

A Numerical Model of Atomic Relaxation and its Applications

*A thesis submitted for the degree of
Doctor of Philosophy*

by

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To Melissa

My love.

Declaration

Except where otherwise indicated, the material presented in this thesis is, to the best of my knowledge, original and has not been submitted in whole or part for a degree in any university.

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Abstract

Atomic relaxation is important in many diverse disciplines, from atomic and nuclear physics to medical physics. A Monte Carlo model of the atomic relaxation is presented in this thesis. Atomic transition probabilities are taken from the Evaluated Atomic Data Library (EADL). Transition energies are calculated using the relativistic self-consistent-field Dirac-Fock method for any specified distribution of atomic vacancies. This CPU-intensive computational approach is adopted to account for the effect of spectator vacancies resulting from non-radiative transitions that occur during the atomic relaxation. The shake theory from Carlson and Nestor is adopted to account for simultaneous ionisation of two electrons in the near closed-shell atomic systems. The model has been extensively benchmarked against the best calculations and available experimental data. The model is applicable to the atomic relaxation resulting from inner-shell ionisation for a wide range of atomic numbers, with particular emphasis on radioactive atoms. In addition, the model can be used to study the femtosecond evolution of the atomic relaxation.

A novel technique, which combines the atomic-relaxation modelling and a Bayesian statistical approach, has been developed to analyse the perturbation in the charge-state distribution of fission fragments due to the decays of nanosecond isomers. This study is, to the author's knowledge, the first theoretical attempt to study the effect of isomeric decays on the charge distributions of fission fragments. This technique has been demonstrated to be applicable for the extraction of isomeric ratios in fission fragments. Furthermore, this study has laid the groundwork to hunt for unknown nanosecond isomers that are low-lying excited states of fission fragments.

Finally, the impact of adopting a rigorous approach to assess transition energies in the atomic-relaxation modelling on dosimetric evaluation of Auger-electron emitting medical radioisotopes, is also investigated. This study showed that the sub-micrometre dimension is where a realistic Auger-energy spectrum is essential for radiobiological studies of Auger-emitting radioactive or irradiated atoms. Therefore, the Monte Carlo model developed for this thesis can provide significantly improved energy spectra to existing radiobiological models for better research outcomes.

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Introduction

1.1 Overview

In 1923, a 26-year-old French physicist named Pierre Victor Auger reported his observation of multiple electron events following photoionisation by low-energy photons, now known as “Auger effect” [1], when he was working in the laboratory of Jean Perrin, who was the winner of the Nobel Prize in Physics 1926 for his discovery of sedimentation equilibrium. The ejection of Auger electrons and the emission of characteristic X-rays constitute a process for an excited atom to release its excess energy and this phenomenon is known as “atomic relaxation”. The atomic relaxation can occur whenever an atom is ionised by irradiation with photons or charged particles. In addition, several nuclear decay modes, such as electron capture (EC) and internal conversion (IC) of nuclear transitions, can interact with atomic shells and ionise an atom to initiate the atomic relaxation. Figure 1.1 illustrates the relaxation of an atom by Auger and X-ray emissions following the K-shell ionisation by a nuclear decay. In fact, the atomic relaxation is responsible to dissipate the extra decay energy that has been transferred to the atom from the nucleus. For example, 85% of the total decay energy released in the EC decay of ^{125}I is dissipated through the atomic relaxation. Since most Auger electrons are short in range, localised Auger-emitting radioisotopes can be utilised in cancer therapy for minimising collateral damage to normal tissues adjacent to the targeted tumour. Auger-emitting radioisotopes have shown promising results in *in vitro* and *in vivo* studies over the last decade [2, 3, 4, 5, 6]. Moreover, a first-in-human trial using ^{111}In -carrying radiopharmaceuticals has been performed recently [7].

Emission spectra are essential for the dosimetric evaluation of Auger-emitting radioisotopes. Calculated Auger multiplicities for common Auger-emitting radioisotopes in the literature are shown in Figure 1.2. A large scatter in the calculated Auger multiplicities is seen for the common Auger-emitting radioisotopes, especially for high- Z elements, and the available data for the less common Auger emitters is very scarce. Moreover, there is growing interest in the use of high- Z nanoparticles as radiosensitisers in medical physics, in which the detailed emission spectrum of high- Z elements resulting from the irradiation by keV photons and MeV protons is needed [8, 9, 10]. Most existing models neglect spec-

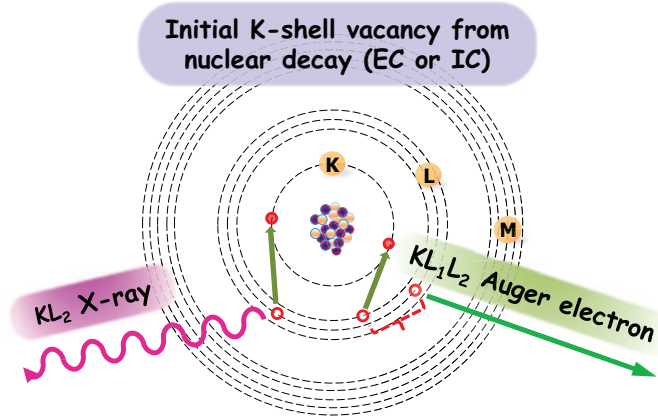


Figure 1.1: Relaxation of an atomic vacancy in the K shell created by a nuclear decay via a K-shell X-ray or Auger electron. The radiative transition involves filling the vacancy in the K shell with an electron from the L_2 subshell and emitting a KL_2 X-ray ($K\alpha_2$ in spectroscopic notation). A secondary vacancy is produced in the L_2 subshell, which will be filled by another transition in the subsequent steps of the atomic relaxation. In contrast, a non-radiative transition fills the K-shell vacancy and results in the ejection of an electron, and the outgoing electron is called the KL_1L_2 Auger electron. Two secondary vacancies are produced in the transition, one in the L_1 subshell and another one in the L_2 subshell. Similarly, these secondary vacancies will be filled in the subsequent steps. Radiative and non-radiative transitions are competing with each other whenever a vacancy is created.

tator vacancies, which can be created after every Auger transition. Most of these models also derive transition energies using neutral binding energies. These assumptions could potentially lead to inaccurate estimates of the emissions of low-energy X-rays and Auger electrons. The inconsistencies in the literature led to the call for the development of an improved model of atomic relaxation by the International Nuclear Data Committee of the International Atomic Energy Agency [11]. Thus, the primary theme of this thesis is to develop a realistic atomic-relaxation model to better estimate the emission spectra due to inner-shell ionisation.

With the availability of a realistic atomic-relaxation model, several topics that involve the emission of Auger electrons and X-rays can then be investigated in detail. For example, the model can be used to model the shift in the charge-state distribution of fission fragments measured at the LOHENGRIN mass separator [12] compared to semi-empirical predictions. The shift has long been described as a consequence of the emission of conversion and Auger electrons following the decays of nanosecond isomers [13], but little effort has been made to fully understand these distributions from underlying principles

This thesis thus has two aims:

- *Development of an atomic-relaxation model*

The first aim of this thesis is to develop a Monte Carlo model of the atomic relaxation, which is named as **BrIccEmis**. In order to account for the effect of spectator vacancies, **BrIccEmis** calculates transition energies based on the relativistic self-consistent-

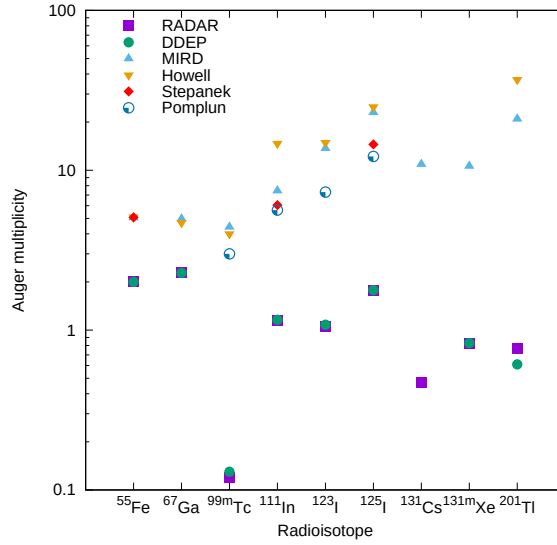


Figure 1.2: Auger multiplicities for several Auger-electron emitting radioisotopes reported in the literature (see Table 3.6 for details). Note the logarithmic scale on the y-axis in this figure.

field Dirac-Fock (DF) method for any given distribution of vacancies. An innovative data storage system has been implemented to handle the huge number of possible vacancy distributions that can occur during the simulation. This leads to a significant reduction of the simulation time, and thus enables **BrIccEmis** to be readily applied to a wide range of atomic numbers. The extensive computation time required to account for the arbitrary distribution of vacancies is one of the main factors behind the neglect of spectator vacancies in the existing models. In addition, **BrIccEmis** also accounts for the shake-off process in the near closed-shell atomic systems. In the pursuit of more realistic modelling of the atomic relaxation, **BrIccEmis** is further improved through benchmarking against available experimental energy spectra and charge-state distributions resulting from inner-shell ionisation. Although **BrIccEmis** is mainly aimed at Auger-emitting radioisotopes, the simulation of the atomic relaxation initiated due to other means, such as photoionisation, can also be performed with a manual specification of the initial-vacancy distribution.

- *Applications of the atomic-relaxation model*

The second aim of this thesis is to apply **BrIccEmis** to investigate several Auger-related topics. The first focus is to model the perturbed charge-state distribution of highly-ionised fission fragments (mean charge $\approx 21+$) resulting from the decays of nanosecond isomers. We demonstrate a novel technique, which is based on the **BrIccEmis** simulation and a Bayesian approach, to analyse the charge distribution and extract isomeric ratios of fission fragments. This study aims to develop a robust method for extracting useful physics quantities from the perturbed charge-state distribution and to lay the foundation for deriving the conversion coefficient and thus

the multipolarity of the transitions from unknown nanosecond isomers in fission fragments.

The second focus is to investigate the impact of new decay spectra generated from **BrIccEmis** on the medically relevant quantities such as dose point kernels (DPKs - radial dose distribution around a unit source) and S -values (absorbed dose per unit cumulated activity) in a joint study with a research group at the University of Oxford. This study pinpointed the spatial domain where accurate Auger-energy spectra is essential for future radiobiological studies of Auger-emitting radioactive or irradiated atoms. As a result of this study, we wish to encourage the use of energy spectra calculated from realistic atomic-relaxation models in radiobiological modelling, especially when the dominant radiation effect is due to the emission of Auger electrons.

1.2 Thesis outline

This thesis is divided into three parts. In the first part of the thesis, the required knowledge for the study of atomic relaxation is presented and the details of the Monte Carlo atomic-relaxation model, **BrIccEmis**, is described

In Chapter 2, the necessary background for modelling the atomic relaxation following nuclear decay is reviewed. In particular, the concepts of electron capture and internal conversion of nuclear transitions as well as the terminologies for the atomic relaxation are presented. In addition, the shake-off theory of Carlson and Nestor [14] is introduced. An overview on the current state of the existing atomic-relaxation models is then given.

In Chapter 3, the details of **BrIccEmis** are presented. The structure of the model is first introduced before the required input nuclear and atomic physics data are described. Then follows a description of several approximations used in the model and a discussion of their impacts on the simulation.. In addition, a protocol for the time-dependent simulation of the atomic relaxation is introduced.

The second part of this thesis is concerned with the benchmarking of the model.

In Chapter 4, energy spectra simulated using **BrIccEmis** are benchmarked against experimental Auger-electron energy spectra resulting from nuclear decay and photoionisation. In addition, the calculated Auger and X-ray transition energies in the singly-ionised systems are compared with the best calculations and evaluations. In this comparison, the importance of the shake-off process in the atomic relaxation is highlighted. The simulated average radiation spectra are also compared with the results from existing models.

In Chapter 5, simulated charge-state distributions are benchmarked against experimental data for the M_5 photoionisation in xenon and ^{131m}Xe decay. The impact of different

ways of selecting the propagating vacancy in the simulation is also discussed. In addition, the impact on the simulation of an alternative input atomic physics data, which is derived from the DF method, is discussed.

The last part of this thesis is concerned with the applications of the model. **BrIccEmis** is applied to study the perturbed charge-state distributions of fission fragments. Then the impact of new decay spectra generated using **BrIccEmis** on the medically relevant quantities is investigated.

In Chapter 6, a novel technique that combines the **BrIccEmis** simulation and a Bayesian approach to analyse the perturbed charge-state distributions of fission fragments resulting from the decays of nanosecond isomers is described. The semi-empirical calculations of the mean charge and width of the unperturbed charge-state distribution, which is established during the fragment's passage in a solid target, are briefly reviewed. Then the technique is used to study the charge-state distributions of three barium isotopes produced from a fission reaction. In addition, the impact of the secondary effects that have been neglected in the simulation is discussed.

In Chapter 7, the impact of **BrIccEmis**-derived energy spectra on dose point kernels (DPKs) and S -values for fourteen Auger-emitting radioisotopes is investigated. First, the definitions of the DPKs and S -values, which are commonly used in nuclear medicine, are introduced. Then, the impact of adopting a rigorous approach to assess transition energies in the atomic-relaxation modelling on the DPKs and S -values is discussed. Furthermore, the impact of the assumption on whether valence-vacancy neutralisation happens during the vacancy cascade, which is one of the main factors contributing to the large variation of the Auger multiplicities reported in the literature, is also discussed.

Finally, the main results of this thesis are summarised in Chapter 8 and a brief outlook for future improvements and further applications of **BrIccEmis** is presented.

Background theory

Overview

In this chapter, the physics related to atomic relaxation will be reviewed and an overview of existing models will be provided. Inner-shell ionisation creates an atomic vacancy and leaves the residual atom in an excited state. Such a vacancy can be created by photoionisation, electron bombardment, nuclear decay, and other processes. In this thesis, we will primarily focus on atomic ionisation due to nuclear processes such as electron capture (EC) and internal conversion (IC). The atomic vacancy created by EC or IC is known as the initial (or primary) vacancy. The initial vacancy will initiate a process of atomic relaxation that gives rise to the emission of X-rays and Auger and Coster-Kronig electrons.

2.1 Radioactive decay processes

2.1.1 Electron capture (EC)

In the EC process, the atomic nucleus decays by absorbing an atomic electron and emitting a neutrino

$$(Z + 1, A) + e^- \rightarrow (Z, A) + \nu_e, \quad (2.1)$$

and the energy condition determining whether EC can occur from the atomic X shell ($X=K, L, M, N, \dots$) is given by [15]:

$$E_\nu = Q^+ - E_i - E_X > 0, \quad (2.2)$$

where Q^+ is the energy of disintegration corresponding to the difference in atomic masses between parent and daughter nuclear ground states, E_i is the energy of the final nuclear state in the daughter nucleus, and E_X is the binding energy of the capture electron in the X shell (or the subshell) in the daughter atom. If the transition energy, $Q^+ - E_i \gg E_K$, then K -shell electron capture dominates [16] (see Figure 2.1). The released energy, E_ν , will be shared by the emitted neutrino and, if applicable, the bremsstrahlung photon or shake-off electron. Moreover, EC decay competes with positron emission or beta plus decay

(β^+) when

$$Q^+ - E_i \geq 2m_0c^2, \quad (2.3)$$

with

$$2m_0c^2 = 1.021998 \text{ MeV}, \quad (2.4)$$

m_0 being the electron rest mass.

The electron capture rate for the X -shell is governed by the fractional capture probabilities, $P_K, P_L, P_M, \dots$, which are normally calculated by means of ratios [16, 17].

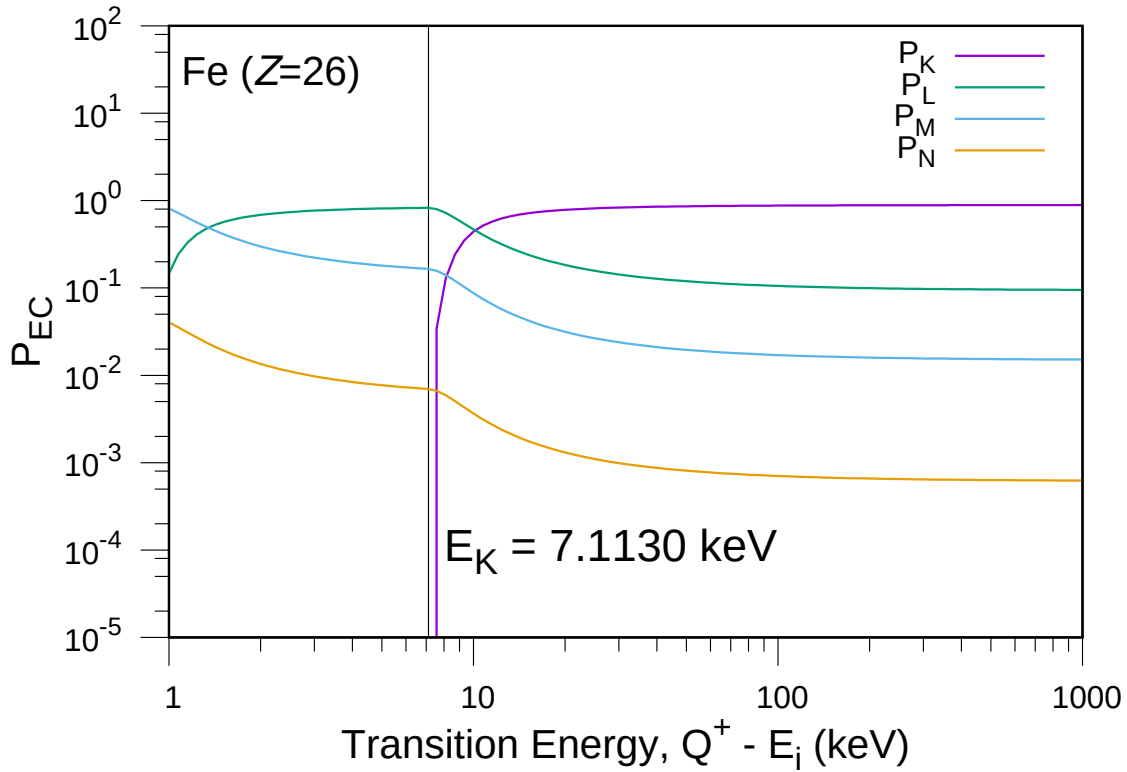


Figure 2.1: Capture probabilities in Fe ($Z=26$) as a function of transition energy. P_K is zero for $Q^+ - E_i < E_K$. The binding energy of the L_1 -subshell of Fe is 0.8461 keV. The plot is generated using Eq. (3.2) to (3.6) below.

2.1.2 Internal conversion (IC)

An electromagnetic (EM) transition occurs when an excited nuclear state de-excites towards a lower nuclear state that may, or may not, be the nuclear ground state. Normally, the energy released in an EM transition is carried away by a γ -ray with energy:

$$E_\gamma = (E_i - E_f) - E_r, \quad (2.5)$$

where $E_i - E_f$ is the energy difference between the initial and final nuclear states of the γ

transition and E_r is the recoil energy of the nucleus in its final state which is given by:

$$E_r = \frac{E_\gamma^2}{2M_N c^2}, \quad (2.6)$$

with M_N as the mass of recoiling daughter nucleus. In most cases, the recoil energy is negligible except for high γ -ray energies and low atomic numbers. When the nuclear transition energy is greater than $2m_0c^2$, an electron-positron pair can be emitted with the pair carrying away energy:

$$E(e^\pm) = E_\gamma - 2m_0c^2, \quad (2.7)$$

with a distribution of energies extending from 0 to $E(e^\pm)$ for each of the two particles.

The de-excitation energy of the nucleus can also be transferred directly to an X -shell electron ($X = K, L_1, L_2$, etc) which is ejected from the atom, carrying away the energy:

$$E_{ic,X} = E_\gamma - E_X, \quad (2.8)$$

where E_X is the binding energy of the X atomic shell. This ejection of an electron is known as internal conversion (IC). The IC coefficient, α_{ic} , which governs the rate of internal conversion, is defined as the ratio of the conversion electron emission probability, P_{ic} , to the γ -ray emission probability, P_γ :

$$\alpha_{ic} = \frac{P_{ic}}{P_\gamma}. \quad (2.9)$$

The internal conversion coefficient of the electron in the X -shell is defined by:

$$\alpha_X = \frac{P_{icX}}{P_\gamma}. \quad (2.10)$$

Transitions involving IC are only possible if $E_{ic,X} > 0$. For example, the 2.1726 keV transition from the decay of ^{99m}Tc can only convert electrons from the M_1 or higher subshell as the transition energy is lower than the atomic binding energy of K- and L-shells (Figure 2.2).

IC depends on the atomic subshell where the converted electron originates, the atomic number Z , the nuclear transition energy, and the multipolarity of the transition. It increases with multipolarity and decreases with increasing transition energy, becoming negligible for high energies where electron-positron pair production becomes important (Figure 2.2). In this thesis, the low-energy regime is the main focus where IC is important. Therefore, electron-positron pair production will be neglected unless specified from here on.

The nuclear transition multipolarity is specified by the type ("electric" or "magnetic") and the angular momentum, of quantum number L , carried off by the photon during the transition. If $L = 0$, the transition is called monopole, $L = 1$ dipole, $L = 2$ quadrupole, etc. The vector relation between L and the total angular momentum quantum numbers of

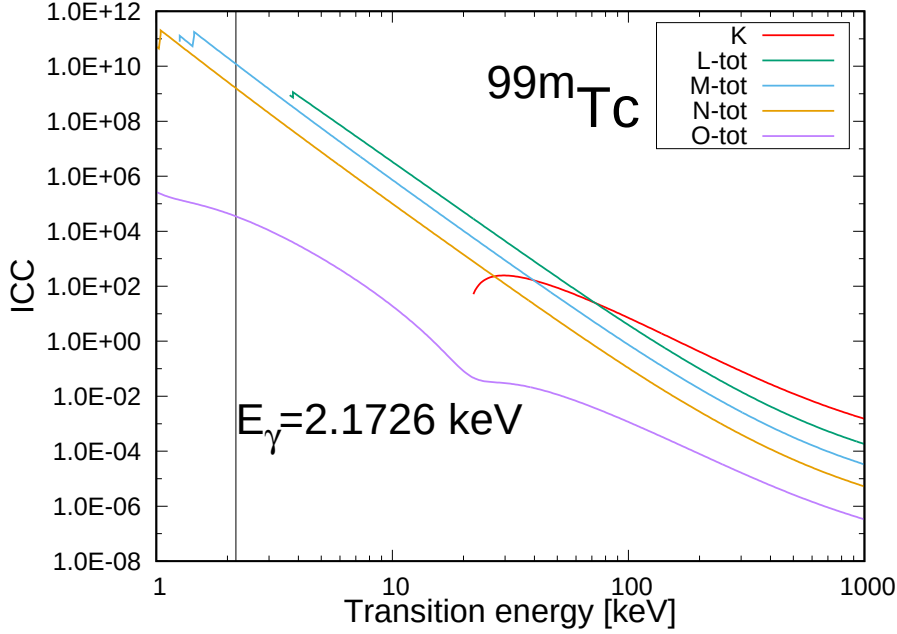


Figure 2.2: Theoretical internal conversion coefficient (ICC) for $E3$ transition in the decay of $^{99m}\text{Tc}_{43}$ as a function of transition energy (keV) from BrIcc [18]. The dashed line marks the position of the transition energy, $E = 2.1726$ keV. E_γ is lower than the binding energies of K- and L-shell.

the initial and final nuclear states of the transition (J_i and J_f) :

$$|J_i - J_f| \leq L \leq |J_i + J_f| . \quad (2.11)$$

If the parity changes and L is odd (even) then the EM transition is electric (magnetic). If the parity does not change and L is odd (even) then the EM transition is magnetic (electric). Unless $J_i = J_f$, the lowest value of L (i.e. $L = |J_i - J_f|$) generally dominates but there is some mixing with higher multipole orders. For example, if the lowest multiple order is a magnetic transition of multipole order L (ML), then competition with electric transition of multipole order $L + 1$ ($EL + 1$) is likely to be significant. The conversion coefficient for a mixed $M1+E2$ transition is related to the conversion coefficients of pure $M1$ and $E2$ transitions by [19] :

$$\alpha(E2/M1) = \frac{\alpha(M1) + \delta^2 \alpha(E2)}{1 + \delta^2} , \quad (2.12)$$

where the multipole mixing ratio, δ^2 , is the ratio of photon emission probabilities of the $E2$ transition to that of the $M1$ transition.

The total probability of an EM transition arising from a nuclear disintegration is the

sum of the γ -emission and IC probabilities:

$$P_T = P_\gamma + P_{ic} + P_{e^\pm} = P_\gamma(1 + \alpha_{ic} + \pi). \quad (2.13)$$

where π is the electron-positron pair conversion coefficient. The probability of X -shell conversion per EM transition is:

$$P_{ic_X} = P_T \times \frac{\alpha_X}{1 + \alpha_{ic} + \pi} = P_\gamma \alpha_X. \quad (2.14)$$

2.2 Atomic relaxation

Vacancy creation in one of the atomic shells leads to a reorganisation of the atomic shells resulting in emission of X-rays or electrons. The distribution of the initial vacancies depends on the nature of the production process. Here, we are mainly interested in the reorganisation of atomic shells after radioactive decay, which means that we mostly consider initial single-vacancy states.

2.2.1 Radiative and non-radiative transitions

When an atomic vacancy is filled by an electron transition from a higher shell, the energy released in this transition could be carried away by a characteristic X-ray, or it is transferred to an outer-shell from which an electron is ejected. The first process is named as a *radiative* transition while the latter process is regarded as *non-radiative* transition. If a non-radiative transition leads to two vacancies in one or two different principal shells, it is called the “*Auger effect*”¹ [23]. In case that one or two vacancies created are in the same principal shell where the initial vacancy is located, the process is called a *Coster-Kronig* (CK) transition [24]. CK transitions may be non-radiative or radiative.

For radiative transitions, two notation systems are in use:

1. Siegbahn: transitions are characterised according to the initial vacancy and a Greek letter as subscript (K_{α_1} , K_{β_1} , L_{α_1} , L_{β_1} , etc.)
2. IUPAC²: transitions are characterised according to the (sub)shells involved (KL_3 , KM_3 , L_3M_5 , L_2M_4 etc.)

¹It is important to note that the first paper that touched on the topic of Auger electrons was published by Lise Meitner [20] when she was studying the energy spectra of β -emitters. Therefore, it has been claimed that Meitner should have got the credit for the discovery of Auger electrons [21]. The issue of whether Meitner’s name should have been included was examined by Olivier Hardouin Duparc [22]. Duparc concluded that “... the Auger effect has rightly been attributed to Auger” because Auger’s observations of multiple-electron events were the central theme of his PhD thesis at Jean Perrin’s laboratory while Meitner’s concern was the exact nature of the β radiations.

²This notation is proposed by the International Union of Pure and Applied Chemistry (IUPAC) in 1991 [25].

Non-radiative transitions are usually denoted by the (sub)shells involved (i.e. IUPAC notation). For example, let us consider a transition that involves the filling of an initial vacancy in the K shell by an electron in the L₂ subshell. If the energy released is transferred to an electron in the L₃ subshell, which is ejected, this is called a KL₂L₃ Auger electron. Similarly, KYZ refers to an electron emitted from the Z shell [26].

The numerical model of atomic relaxation **BrIccEmis** that the author has developed for this thesis is based on atomic transition probabilities from the Evaluated Atomic Data Library (EADL) [27] which was tabulated using the IUPAC notation. Therefore, the IUPAC notation will be used throughout this thesis. A nomenclature list of atomic transitions can be found in Appendix A.1.

Transition energies

The X-ray energy emitted for an XY transition (X = K, L, M, etc., Y = L, M, N, etc.) is written as:

$$E_{XY} = E_X - E_Y, \quad (2.15)$$

where E_X and E_Y are the atomic binding energies of the shells X and Y, respectively. Similarly, the electron energy E_{XYZ} emitted for a XYZ transition is given by:

$$E_{XYZ} = E_X - E_Y - E_Z^Y, \quad (2.16)$$

where E_X and E_Y are the atomic binding energies for shells X and Y. E_Z^Y is the binding energy of an electron on the Z shell when the atom is already ionised with a vacancy in the Y shell. This process will produce two vacancies in the Y and Z shells from the initial vacancy in the X shell. Semi-empirical calculations of Auger-electron energies for elements $10 \leq Z \leq 100$ for KLL up to N₅OO transitions were published by Larkins [28]. The agreement with experimental values is generally within 1-2 eV with maximum discrepancies near 5 eV for both vapor- and solid-phase systems. However, the uncertainties in Auger-electron energies for high atomic numbers could be in the range 10-20 eV due to absolute uncertainties in binding energies used in the tabulation [28].

Transition probabilities

An atom with a vacancy in one of its shells is in an excited state. The associated lifetime of the excited state τ is related to the level width Γ by $\Gamma\tau = \hbar$ where $\hbar$ is the reduced Planck's constant. The total decay rate of the excited state A is the inverse of the lifetime $A = \tau^{-1}$ (or $\Gamma = A\hbar$). If different decay processes from the excited state are assumed to be independent, then the level width is the sum of the radiative width, Γ_R , the Auger width, Γ_A and the Coster-Kronig width, Γ_{CK} :

$$\Gamma = \Gamma_R + \Gamma_A + \Gamma_{CK}. \quad (2.17)$$

Yields for the various processes are defined as follows (see Appendix A.1 for detailed definitions):

$$\begin{aligned} \text{Radiative or fluorescence yield} \quad \omega &= \Gamma_R/\Gamma, \\ \text{Auger yield} \quad a &= \Gamma_A/\Gamma, \\ \text{Coster-Kronig yield} \quad f &= \Gamma_{CK}/\Gamma. \end{aligned} \quad (2.18)$$

The sum of all three yields is unity:

$$\omega + a + f = 1. \quad (2.19)$$

K shell

For a given radioisotope, n_K , the total number of K initial vacancies created following a nuclear decay, is:

$$n_K = \sum P_{EC} P_K + \sum P_\gamma \alpha_K, \quad (2.20)$$

where P_{EC} is the electron capture intensity, P_K is the K-shell capture probability, P_γ is the intensity of γ -transition and α_K as the K-shell conversion coefficient. The K shell has no subshells therefore CK transitions are not possible. The sum of the radiative yield and the Auger yield is unity:

$$\omega_K + a_K = 1. \quad (2.21)$$

The two yields can be written as :

$$\omega_K = \frac{N_{X_K}}{N_{X_K} + N_{A_K}} \quad a_K = \frac{N_{A_K}}{N_{X_K} + N_{A_K}}, \quad (2.22)$$

where N_{X_K} is the number of K X-rays emitted and N_{A_K} the K-Auger electrons ejected.

The K-shell fluorescence yield, ω_K , depends exclusively on the atomic number Z , except for low values of Z , where it may be influenced by the chemical state [16]. It increases with Z and approaches 1 for high Z (see Figure 2.3). Moreover, Bambynek et al. [32] have shown that experimental K-shell radiative yields can be fitted with the following semi-empirical relation:

$$\left\{ \frac{\omega_K}{1 - \omega_K} \right\}^{1/4} = \sum_{i=0}^3 B_i Z^i, \quad (2.23)$$

where $B_0 = 0.015 \pm 0.010$, $B_1 = 0.0327 \pm 0.0005$, $B_2 = 0$, and $B_3 = -(0.64 \pm 0.07) \times 10^{-6}$.

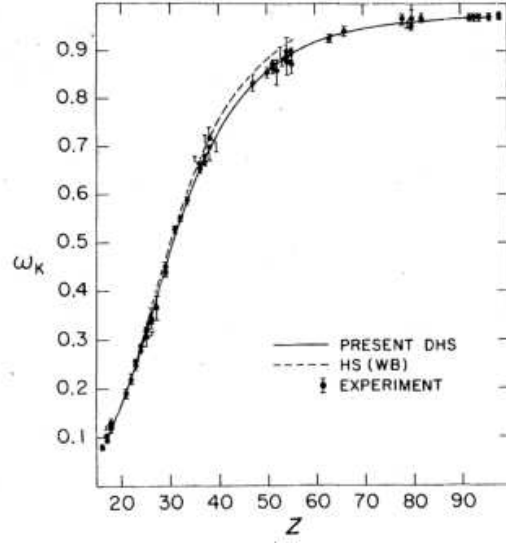


Figure 2.3: Atomic K -shell fluorescence yields as a function of atomic number. “Present DHS” are relativistic Dirac-Hartree-Slater calculations [29] and “HS(WB)” are non-relativistic Hartree-Slater calculations [30, 31]. Experimental data adopted in the review paper by Bambenek *et al* [32] are also shown. Image extracted from [29].

L shell

The vacancies in the L shell can be created by L-electron capture, by emission of an L-shell conversion electron, and by transfer of an L-shell electron to fill a K-shell vacancy. So the total number of L-shell initial vacancies corrected for K-shell transfer is given by:

$$n_L = n_{L_1} + n_{L_2} + n_{L_3} = \sum P_{EC}P_L + \sum P_\gamma\alpha_L + n_K n_{KL}, \quad (2.24)$$

where P_L is the L-shell capture probability and α_L is the L-shell conversion coefficient, n_K is the total number of K initial vacancies calculated using Eq. (2.20), and n_{KL} is the number of L vacancies created after an L-shell electron is used to fill a K-shell initial vacancy.

Moreover, additional vacancies could be created in L_2 and L_3 shells due to L-shell CK transitions. If N_{L_i} are the relative numbers of the initial vacancies in each subshell (i.e. $N_{L_i} = \frac{n_{L_i}}{n_L}$), then the mean radiative yield $\bar{\omega}$ and mean Auger yield $\bar{a}$ are given by:

$$\bar{\omega} = \sum_{i=1}^3 N_i v_i \quad \bar{a} = \sum_{i=1}^3 N_i \mathbf{a}_i, \quad (2.25)$$

where the coefficients v_i and $\mathbf{a}_i$ are the effective L-shell radiative and Auger yields, respectively, which result per initial vacancy in the L_i subshell, Eq. (2.24). The effective radiative

yields v_i are related to the subshell radiative yields ω_i and the CK yields by:

$$\begin{aligned} v_1 &= \omega_1 + f_{12}\omega_2 + (f_{13} + f_{12}f_{23})\omega_3, \\ v_2 &= \omega_2 + f_{23}\omega_3, \\ v_3 &= \omega_3. \end{aligned} \tag{2.26}$$

Similarly, the effective Auger yields $\mathbf{a}_i$ can be expressed in terms of the subshell Auger yields a_i and the CK yields:

$$\begin{aligned} \mathbf{a}_1 &= a_1 + f_{12}a_2 + (f_{13} + f_{12}f_{23})a_3, \\ \mathbf{a}_2 &= a_2 + f_{23}a_3, \\ \mathbf{a}_3 &= a_3. \end{aligned} \tag{2.27}$$

and the subshell yields obey the following relations:

$$\begin{aligned} \omega_1 + a_1 + f_{12} + f_{13} &= 1, \\ \omega_2 + a_2 + f_{23} &= 1, \\ \omega_3 + a_3 &= 1. \end{aligned} \tag{2.28}$$

Figure 2.4 shows that for $Z < 90$ with a single vacancy on the L shell or a higher shell, non-radiative processes dominate ($\omega < 0.5$) [33].

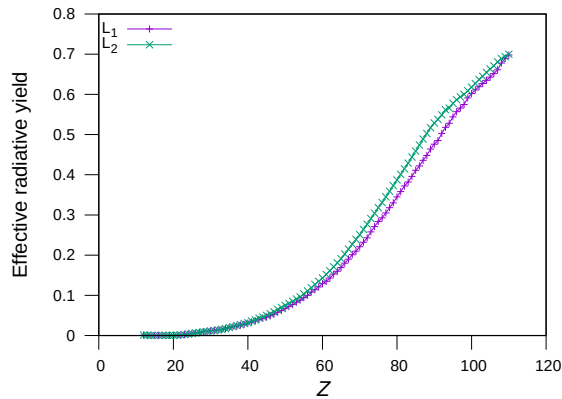


Figure 2.4: Theoretical effective fluorescence yield of L_1 and L_2 subshells from Krause [33].

The sum of the mean radiative and the mean Auger yields for a given initial vacancy distribution is:

$$\overline{\omega_L} + \overline{a_L} = 1. \tag{2.29}$$

For $30 \leq Z \leq 96$, Cohen [34] summarised the status of mean L-shell radiative yield

data and presented the following semi-empirical relation

$$\left\{ \frac{\overline{\omega_L}}{1 - \overline{\omega_L}} \right\}^{1/4} = \sum_{i=0}^3 B_i Z^i, \quad (2.30)$$

with the following coefficients

$$\begin{aligned} B_0 &= 0.177650, \\ B_1 &= 2.98937 \times 10^{-3}, \\ B_2 &= 8.91297 \times 10^{-5}, \\ B_3 &= -2.67184 \times 10^{-7}. \end{aligned} \quad (2.31)$$

M shell and higher shells

The formalism to describe radiative, Auger and CK transitions and their yields in the M shell or higher shells may be derived in analogy to that given for the L-shell. However, by the time the M-shell vacancies are created, the atom is likely to have more than one vacancy due to the emission of Auger and CK electrons and the corresponding formulae become very complicated.

2.2.2 Vacancy cascade

The number of vacancies can multiply due to the dominance of non-radiative transitions in the atomic relaxation process. Exception are when the hole is in the K shell of an element where $Z > 30$ or for some L shells of very heavy elements. Auger transition probabilities are largest when the overlap between the orbitals is greatest [35]. The full atomic relaxation is a multi-step phenomenon that involves a series of vacancy-filling processes to reorganise the atomic structure. It is therefore also given other names such as “vacancy cascade” or “Auger-electron cascade”.

The vacancy cascade begins after the creation of initial vacancy and it will continue until all vacancies are filled by the emissions of radiative and non-radiative transitions, or until no more atomic transitions are energetically possible. Atomic excited states, except those low-lying states which can only proceed with low-energy radiative transitions, generally have lifetimes in the order of femtoseconds, thus the vacancy cascade has a lifetime of 10^{-16} to 10^{-13} s [36, 37, 38].

Although the fundamental picture of the individual atomic transitions remains similar to the one described in Section 2.2.1, the atomic structure will continuously change as the vacancies migrate towards the outer shells. Therefore, the deterministic description of the total number of emitted X-rays and electrons like the one described in Section 2.2.1 is no longer sufficient. The stochastic model of vacancy cascade was first suggested by Krause and Carlson in the 1960s when they were studying the charge distribution of rare

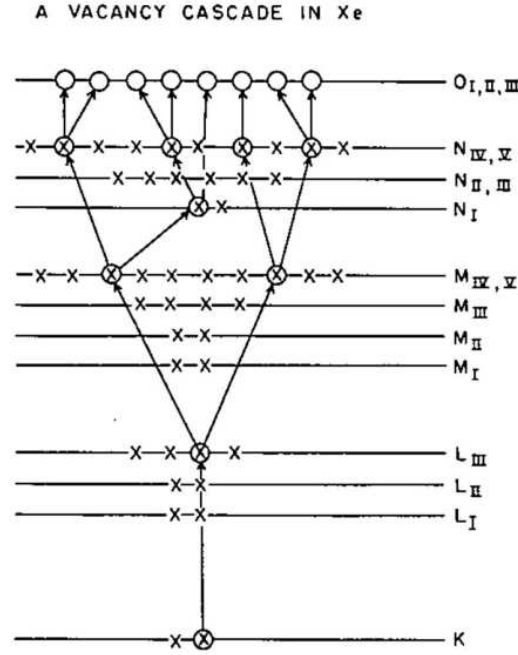


Figure 2.5: Typical vacancy cascade in xenon following K-shell ionisation with ultimate loss of all electrons in the O shell. Electrons are represented by $\times$, vacancies by $\circ$ and filled vacancies by $\otimes$. The first step is a KL_3 X-ray, it is then followed by seven non-radiative transitions. The final result is 8 vacancies in the O shell (assuming there the atom is isolated). Image is extracted from [36].

gases following inner-shell ionisation [36, 39, 40, 41]. The model was built by combining knowledge of atomic radiations in the neutral atom and a Monte Carlo (MC) technique. The calculated charge distributions were in good agreement with the experimental results. It has since been accepted by the scientific community that the mechanism of the vacancy cascade is indeed stochastic in nature. A schematic diagram of a typical vacancy cascade in xenon is shown in Figure 2.5.

Interest in using Auger-electron emitting radioisotopes for targeted radionuclide therapy grew in the 1980s due to the desirable property of Auger and CK electrons in minimising the penetrating radiations to neighbouring cells (i.e. selective delivery of ionising radiations) [42, 43, 44, 45]. Charlton and Booz [46] applied the stochastic model of atomic relaxation to generate the electron spectrum following the decay of ^{125}I which could emit many Auger and CK electrons due to two vacancy cascades. First, one following the EC decay of the parent nucleus, and second, one after IC of the 35.5 keV γ -transition with the conversion rate of 91.4% (see Figure 2.6). Since then, modelling of the vacancy cascade has primarily been used to seek understanding of the Auger effect in a biological medium. Most existing models of the vacancy cascade approximate the Auger transition energies using methods that rely on neutral-atom binding energies, which will produce a discrete distribution of energy lines rather than a continuous distribution. However, a continuous

Auger-energy distribution is an inevitable result of the vacancy cascade phenomenon. In his review paper on the topic [35], Carlson, who was the pioneer in understanding the vacancy cascade, summarised as follows:

... as the vacancy cascade proceeds, the variety of possible Auger processes increase. In the case of a heavy element, the last stages of the vacancy cascade will produce almost a continuum of electrons.

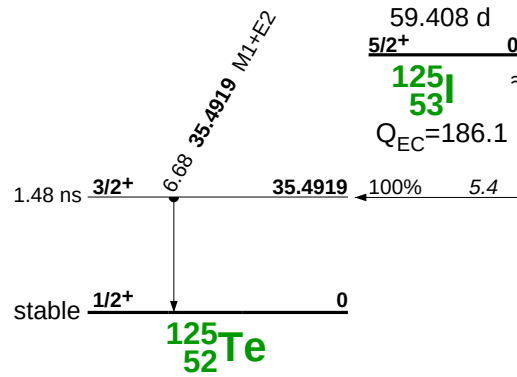


Figure 2.6: Decay scheme of ^{125}I

It should be mentioned that most Monte Carlo radiation transport codes, such as PENELOPE [47], MCNP [48], FLUKA [49], and Geant4 [50], implement a partial vacancy cascade assuming static atomic structure (i.e. using transition energies and probabilities calculated from a single-vacancy approximation) in the handling of energy dissipation following atomic ionisation by photons or charged particles.

2.3 Shake-off and shake-up

2.3.1 Shake theory

Removal of an electron from an atom will change the effective nuclear charge viewed by the remaining electrons. This results in a sudden change in the central potential which could excite an atomic electron into the continuum (shake-off) or into a lesser-bound discrete excited state (shake-up) [14]. Both shake-off and shake-up may result from processes that remove an atomic electron such as photoionisation, electron capture, internal conversion, as well as radiative and non-radiative transitions. Moreover, β^- and α decays can also shake off atomic electrons [51]. The shake-off electron shares energy with the primary radiation that creates it. Therefore, shake-off electrons contribute to the continuous energy distribution from 0 eV up to the lower energy side of the main peak. Figure 2.7 shows a schematic diagram of such an energy distribution. In contrast, the shake-up effect is

less noticeable since it does not alter the charge state of an ion and it mainly reduces the kinetic energy of the primary radiation (thus producing a satellite line).

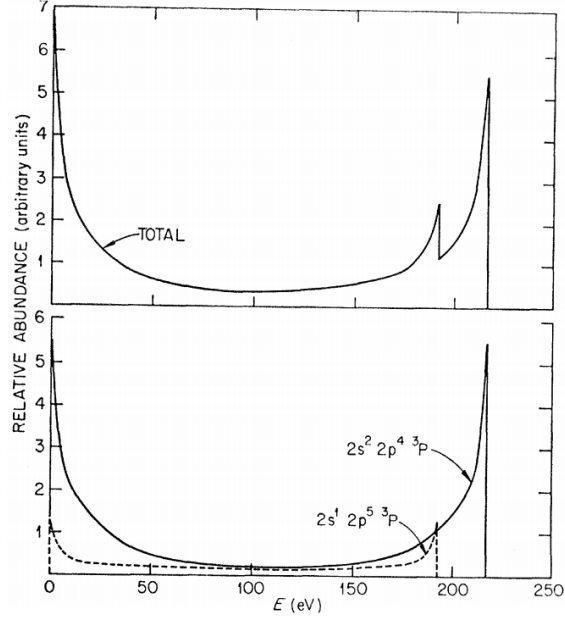


Figure 2.7: The overall distribution of electron energies for multiple electron ejection following the photoionisation of the L shell of neon. The lower panel shows the contributions from the simultaneous ionisation of two electrons from the $2p$ subshell and one each from the $2s$ and $2p$ subshells. The continuous background mainly comes from the energy-sharing process between the shake-off electrons and the photoelectrons. The upper panel shows the sum of the contributions. Image extracted from [52].

The probability of electron shake-off is mainly calculated based on the sudden approximation [14, 51, 53, 54]. In the sudden approximation, the probability for an electron initially represented by the wave function ψ_i to be found in a given final wave function ψ_f is given as the square of the overlap integral between ψ_i and ψ_f [14]:

$$P_{i \rightarrow f} = \left| \int \psi_f^* \psi_i d\tau \right|^2, \quad (2.32)$$

where the initial state is the neutral atom and the final state is an ion created by the primary ionisation event and the atomic potential is different from that for the initial state. The shake-off and shake-up processes obey monopole selection rules which means there is a change only in the principal quantum number and all other quantum numbers should retain the original values.

Another method of calculating the probability of shake-off and shake-up is to calculate the probability that an atomic electron will remain in an orbital with the same quantum numbers as the initial state and subtract this value from unity. Therefore, the probability of promoting an orbital electron designated by n and l , where n and l are the principal and orbital angular momentum quantum numbers, to a higher bound state or to the continuum,

is given by [14]:

$$P_{nl} = 1 - \left[\left| \int \psi_{nl}'^* \psi_{nl} d\tau \right|^2 \right]^N - P_F, \quad (2.33)$$

where ψ_{nl} represents the single-electron wave function of the orbital nl in the neutral atom and $\psi_{nl}'^*$ represents the orbital nl in the ion with a single vacancy created in a given subshell. These wave functions are calculated using the Hartree-Fock or Dirac-Fock method in the single-configuration framework. N is the number of electrons in orbital nl . The quantity P_F is a correction that arises from the condition that electron shake-up transitions to occupied levels are forbidden due to the Pauli principle. When the principal quantum number of the highest occupied state is x , P_F is given by [14, 53]:

$$P_F = \sum_{n'=1}^x N \frac{N'}{2(2l+1)} \left| \int \psi_{n'l}'^* \psi_{nl} d\tau \right|^2, \quad (2.34)$$

where $n' \neq n$, and N' is the number of electrons in the orbital designated by $n'l$. According to calculations by Carlson and Nestor [14] for the rare gases, the following two generalisations about the probabilities calculated using Eq. (2.33) can be made:

1. P_{nl} increases with n for a given inner-shell vacancy,
2. For a given orbital nl , P_{nl} is negligible if the vacancy is created in a higher shell than the orbital; it is significant for a vacancy in the same n quantum number, but becomes substantially larger for a vacancy in the $n - 1$ principal shell.

It is important to note that Eq. (2.33) includes both shake-off and shake-up processes. Shake-up is relatively unimportant for the inner shells, but for the valence shell electrons, it is equally important as the shake-off.

Calculations by Carlson and Nestor [14, 51] and Mukoyama [53] are based on the single-configuration framework. Calculations using the multiconfiguration framework, which accounts for electron-electron correlation, have been proposed by Dyll [55] for noble gases and recently the method has been applied by Lowe et al. [56] to transition metals which have open $3d$ ($M_{4,5}$) shells. Theoretical values calculated using the multiconfiguration framework are generally in much better agreement with experimental data.

2.3.2 Satellites, hypersatellites and multiple-electron ejections

X-ray *satellites*, especially those accompanying K X-rays, have been observed in many experiments [57, 58, 59]. These satellite lines appear when the transitions occur in the presence of spectator vacancies. For example, a K_{α_1} radiative transition from L_3 to the K shell in the presence of a spectator vacancy in the L_2 subshell will give rise to the $KL_3^{L_2}$ satellite. Removal of an electron from an inner shell reduces the screening of the atomic

potential. Because of this, the binding energies of the inner shells are increased more than the outer shells. So X-ray satellites emitted from a multiply-ionised atom will have slightly higher energy than the parent X-ray lines [59, 60]. If the spectator vacancies are in the same shell as (or lower than) the filling vacancy, the transition energy could be significantly enhanced and these transitions are usually referred to as the *hypersatellites* [61, 59]. For example, a K_{α_1} transition filling an empty K shell gives rise to a $KL_3^{[K]}$ hypersatellite line ($1s^{-2} \Rightarrow 1s^{-1}2p_{3/2}^{-1}$). The energies of hypersatellites are higher than those of normal satellites. For satellites of inner-shell transitions, the spectator vacancies are normally created as a result of the shaking process following the inner-shell ionisation. Since the probability of removing an electron from an inner shell is very small relative to the probability of promoting a valence electron through shake-off or shake-up, the intensities of hypersatellites are normally lower than those of satellites.

The radiative Auger effect (RAE) occurs when an X-ray is emitted simultaneously with an electron as a result of shake-off. Åberg and Utriainen [62] reported the first observation of RAE. Similar to the description provided in Section 2.3, the total energy released in RAE is shared between the electron and, in this case, the X-ray photon. It has been observed that the electrons have almost zero energy in RAE, which means the peak energy of RAE photons lies very near to the low-energy side of the parent line [63, 64, 65]. Unlike the shake-off accompanying other hole-creation processes (i.e. IC, photoionisation, Auger, etc.), RAE is only important if a radiative transition involves an electron from the valence shell [66]. Since the X-ray transition does not create a new vacancy, the valence electrons do not experience a sudden change in the central potential if it involves only the inner shells. Therefore, the RAE is mainly observed for low- Z elements [60, 67]. Figure 2.8 shows the decomposition of the satellites, hypersatellites and RAE structures in the spectrum of K_{α} and K_{β} X-rays in Vanadium.

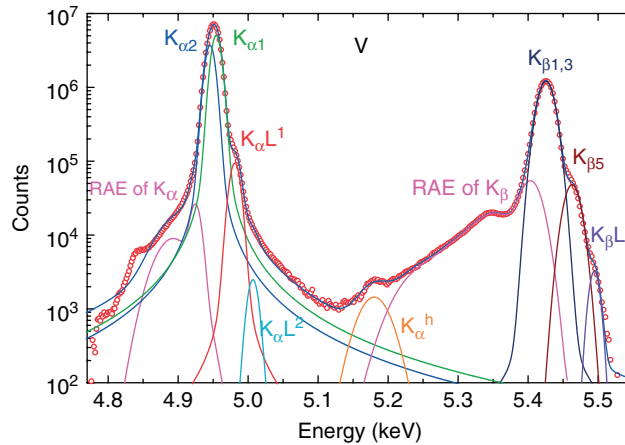


Figure 2.8: Satellites, hypersatellite and radiative Auger effect (RAE) structures of the K X-rays of vanadium emitted following photoionisation. Image extracted from [59].

Auger satellites are formed in a similar way to the X-ray satellites, but they appear on the low-energy side of the parent Auger lines while Auger hypersatellite lines are higher in energy than the parent lines [36, 68, 69, 70]. Unlike the X-ray satellites, Auger satellite lines are usually harder to quantify. Since Auger transitions are usually dominant in higher shells, their *actual* transition energies could be significantly different from the energies calculated using the *single-vacancy approximation*. In fact, the energy spectrum of outer-shell Auger electrons is almost a continuum. Figure 2.9 shows the kinetic energy spectra of outer-shell Auger electrons measured using the multi-electron coincidence technique [71].

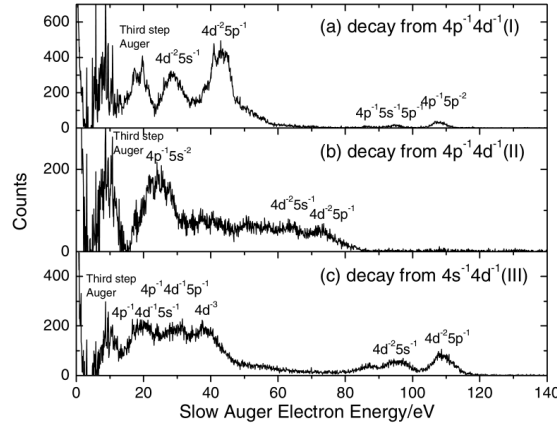


Figure 2.9: Kinetic energy spectra of N-shell Auger electrons following M_5 -Auger transition. Image extracted from [71].

Shake-off results in two-electron ejection and this phenomenon is sometimes called the *double Auger* decay [72, 73, 74, 75, 76]. In fact, Carlson and Krause [72] first assigned the shake-off process as the double Auger decay before they eventually characterised the phenomenon. The total shake-off probability of a given orbital, P_{nl} calculated using Eq. (2.33) includes the shake-off of two or more electrons, which is usually small using the Binomial theorem ($< 3\%$ of single shake-off). When two shake-off electrons are ejected, the phenomenon is sometimes called the *triple Auger* decay [77].

2.4 Overview of existing atomic relaxation models

Since the first application of a vacancy cascade model for medical applications by Charlton and Booz [46], several programs have become available to compute detailed atomic radiation spectra, mainly for radiation protection and dosimetric purposes. This section gives an overview of the current state of these programs.

2.4.1 Existing programs for calculating atomic spectra

In the modelling of the vacancy cascade, there are two different approaches being used. One is the so-called *deterministic approach* and the other is to base the calculations on

Monte Carlo techniques.

In the deterministic approach, the contributions from filling each vacancy are computed using closed formulae, similar to Eq. (2.22). This approach has very small computational requirements and it is mainly used for the cascade up to M shell. The approach has been used by the Radiation Dose Assessment Resource (RADAR) [78, 79] and the Decay Data Evaluation Project (DDEP) [80], and the Medical Internal Radiation Dose (MIRD) committee [81]. The deterministic approach is quite reasonable and simple for transitions involving vacancies on the K and L shells where the number of spectator vacancies is small.

A set of approximating formulae were derived by Dillman [82] to evaluate the L- and M-shell atomic radiations. Dillman used a rather coarse approach, which assumed that these radiations carry a low total energy and may be treated as a “*single group*”. His work led to the development of the EDISTR code [82] to evaluate the complete spectrum of atomic radiations. Recently, Endo et al. [83] have further improved the EDISTR code based on the RELAX code [84] to become the EDISTR04 code which is used by the MIRD committee [81].

The alternative approach, Monte Carlo calculations, proves to be better suited to the inclusions of all possible paths in the relaxation process. This technique was pioneered by Krause and Carlson [85] in their study of the reorganisation of the atomic structure of krypton following photoionisation. Subsequently, the technique was translated by Charlton and Booz to study the biological effect of Auger-emitting radionuclides [46]. Since then, it has been adopted by Howell [38], Stepanek [86] and Pomplun [87, 88] in their calculations. Pomplun’s model was built on the model of Charlton and Booz [37]. This method allows the incorporation of all atomic shells with the potential to produce low-energy Auger electrons with high radiotoxicity. It has high computational requirements since it includes all atomic shells in the calculation. Because of this, most of the existing Monte Carlo codes generate atomic radiation spectra based on relatively low number of samples, typically 10,000 decays, which might not be enough to obtain a converging solution for atoms with high atomic number, Z .

Common in both approaches is the necessity to know all relevant transition energies and transition rates. Table 2.1 summarises the information about existing programs. The last column shows a brief summary of the model to be discussed in Chapter 3.

Table 2.1: Comparison of existing programs. This table has been partially published in Ref. [89].

	RADAR	DDEP	Eckerman & Endo (MIRD)	Howell	Stepanek	Pomplun	Present study
	[78, 79]	[80]	[81]	[38]	[86, 90]	[88, 87]	
Nuclear decay/structure data ^(a)	ENSDF	DDEP	ENSDF	ENSDF	ENSDF	ICRP38	ENSDF
Conversion coefficients	[91]	[18, 92]	[92, 93]	[92]	[86]	[91, 94, 95]	[18]
Electron capture probabilities	[96]	[97]	[98, 99]	[96, 100]	[96, 100]	[96]	[97]
Atomic shells	K, L	K, L	K – O	K – O	K – N	K – N	K – O
Atomic transition rates ^(b)	[32, 101]	[102, 103, 104]	[27]	[44, 105, 106, 107] (A)	[27]	[105, 106, 107] (A)	[27]
	RADLST	EMISSION	EDISTR04	[102, 108] (X)		[33, 109] (X)	
Atomic transition energies ^(c)	NAB [110]	SE [28]	NAB [27]	Z/Z + 1 (A) NAB (X)	DF	DF [111]	DF [112]
Shake-off	No	No	No	No	No	Yes/No	Yes/No
Vacancy propagation ^(d)	DET	DET	DET	MC	MC	MC	MC
Charge neutralisation	No	No	Yes	Yes	Yes/No	Yes/No	Yes/No

(a) ENSDF: Evaluated Nuclear Structure Data File [113]; DDEP: Decay Data Evaluation Project [80],

ICRP38: International Commission on Radiological Protection [114].

(b) Computer codes: RADLST by Burrows [101], EMISSION by Schönfeld & Janßen [115],

EDISTR04 by Endo, Yamagichi & Eckerman [83]; (A) – Auger electrons, (X) – X-rays.

(c) Transition energies deduced from: NAB: neutral atom binding energies; SE: semi-empirical Auger energies,

Z/Z+1: approximated from neutral atom binding energies [116]; DF: relativistic self-consistent Dirac–Fock calculations.

(d) Approach to treat vacancy cascades: DET: deterministic, using closed formulae; MC: Monte Carlo.

All six calculations listed in Table 2.1 use transition rates from existing tabulations based on a combination of experimental data, systematics (obtained by interpolation and extrapolation), as well as theoretical calculations, which often use different assumptions, wave functions, and so forth. The two most cited works concerning the atomic transition rates used by these calculations are Bambynek et al. [32] and the Evaluated Atomic Data Library (EADL) [27]. Transition energies and probabilities presented in these compilations are for cases where there is a single vacancy in one of the atomic shells. In an effort to compensate for this limitation, the calculations by existing programs normally employ various corrections. One of these is the so-called Krause-Carlson correction [85], which takes into account the effect of multiple spectator vacancies (see Section 3.4.3). Shake-off and shake-up are usually neglected due to limited theoretical calculations (mainly available for noble gases) and the increased complexity required to evaluate the vacancy cascade.

Atomic binding energies are affected by the changes in atomic structure occurring during the vacancy cascade. Some of the calculations listed in Table 2.1 use neutral atom binding energies (NAB) to estimate transition energies based on formulae similar to Eq. (2.15) and (2.16) or semiempirical values (SE) from Larkins [28]. Others use the $Z/Z + 1$ rule [116] to estimate the Auger transition energies. This rule uses the electron binding energies pre-calculated for atoms in ground state, i.e. without any consideration of the realistic electron vacancy distribution during the vacancy cascade. Therefore the application of this rule can lead to overestimation in energies of transitions between the outermost atomic subshells [117]. Only Stepanek [86], Pomplun [87, 88] and the model developed for this thesis, adopted theoretical binding energies calculated from the relativistic self-consistent Dirac-Fock method.

2.4.2 Input data of existing programs

For nuclear structure data, the Evaluated Nuclear Structure Data File (ENSDF), is adopted by most programs except DDEP [80] and Pomplun [87, 88]. Pomplun uses decay data from publication 38 of the International Commission on Radiological Protection (ICRP) [114] to determine the distribution of initial vacancies. Most programs except Stepanek [86, 90] use internal conversion coefficients (ICC) tabulated in the 60s and 70s by Hager [91], Rösler [92], Band [93, 95] and Dragoun [94]. Recently, DDEP replaced some ICC with values from BrIcc [18], which are also adopted in this work. Stepanek builds his own unpublished ICC database called the INCOCO library [86, 90].

The electron capture probabilities are generally extracted from Nuclear Data Sheets [96, 100] and Bambynek et al. [98]. Howell and Stepanek assume that L_3 capture is negligible and only M_1 and N_1 contribute to M- and N-shell capture rates. In addition, they also assume that the contribution of N_1 capture is only 10% of the M_1 capture. They have to make these assumptions because the Nuclear Data Sheets they used only provide

capture probabilities P_K , P_L and P_{M+} . From Table 3.1, the calculated P_N is not always 10% of P_M . Only MIRD [81] calculates the capture probabilities for the K-shell to the O_8 -subshell using the data from Refs. [98, 99].

All Monte Carlo codes cover the atomic transitions from K to N-shell and they generally adopt transition probabilities from EADL [27], in which K and L-shell transition rates are adopted from Scofield [102] and Chen [105], or McGuire [106, 107] (M and N-shell transitions). For programs using the deterministic approach, RADAR and DDEP adopt transition rates from other publications which only consider K and L shells but MIRD adopts transition rates and energies from EADL.

See Table 2.1 for details.

2.4.3 Assumptions on the state of the atom

In the evaluation of Auger energy spectra, there are two general assumptions on the state of the atom: (a) isolated atom and (b) condensed phase. Distinction between the two assumptions is the inclusion or neglect of *charge neutralisation* during the vacancy cascade. An atom in the condensed-phase approximation is assumed to have an unlimited source of *continuum* electrons from the environment so that any vacancy created in the valence shell is filled immediately. This assumption is sometimes called *fast neutralization* while the isolated-atom approximation is called *slow neutralisation* [38, 86, 88, 118]. Recently, indications have been found that charge neutralisation in a molecular environment could happen as fast as the vacancy cascade (10^{-16} to 10^{-13} s) [119, 120]. In the proposed model for this thesis, we will consider both isolated-atom and condensed-phase approximations for comparison.

Common to both assumptions is the necessity to address charge neutralisation between successive nuclear events, such as between the electron capture and internal conversion processes. Lifetimes of the intermediate nuclear state are usually at least a few orders of magnitude longer than the lifetimes of the vacancy cascade [38]. It is therefore reasonable to assume that the internal conversion (IC) process takes place *after* the daughter atom is fully relaxed following an electron capture (EC) event, so it takes place in a neutral daughter atom. This means the vacancy cascade following an EC event happens in an excited state of the *daughter* atom (i.e. *parent's* electronic structure with a missing inner-shell electron) whereas the cascade following IC process happens in a neutral daughter atom. This assumption is violated only in rare cases wherein the nuclear level half-life of the daughter nucleus is much shorter than the time needed for the atom to be fully neutralised. For example, a shift of 20 ± 7 eV has been observed for the K conversion electron of the 963 keV transition [$T_{1/2}(963 \text{ keV level}) = 30 \text{ fs}$] in the electron capture decay of $^{152\text{m}}\text{Eu}$ [121].

Depending on the nuclear level scheme, the radioactive decay may produce multiple

electromagnetic transitions which, depending on the conversion coefficient, may proceed with the emission of multiple conversion electrons. There is a finite, usually very small, probability that a second conversion electron is emitted before the vacancies created in the previous conversion process can be fully neutralised. Based on the average nuclear and atomic half-lives, it is a very unlikely event, and it will not be considered in the model of this thesis.

2.5 Summary

A vacancy cascade model was first proposed by Krause and Carlson [85] in the 1960s to study the charge-state distribution that resulted from the atomic relaxation processes following inner-shell ionisation. It was subsequently applied to study the local energy deposition by Auger-emitting radioisotopes following the introduction of Auger-based targeted therapy [43, 44]. There are several existing programs that calculate atomic radiation spectra based on the vacancy cascade model (see Table 2.1). However, the International Nuclear Data Committee (INDC) of the IAEA has pointed out that there are inconsistencies across all existing programs and there is an urgent need to develop a computational route to “determine the energies and emission probabilities of the low-energy X-rays and Auger electrons to a higher degree of detail and consistency than is available at present” [11]. Moreover, one of the pioneers in microdosimetry of low-energy electrons, Nikjoo, has pointed out that “both the data base and the computer codes used to simulate the decay of Auger-emitting radioisotopes are not available to the generality of the community” [118]. Although Stepanek’s code [90] is made available to the public, it has rarely been used or cited.

In order to address these issues raised by the scientific community, a stochastic model of the vacancy cascade has been developed at the Australian National University, which will be described in detail in the next chapter.

A new model for atomic relaxation - BrIccEmis

Overview

In this chapter, details of a stochastic model of the vacancy cascade, which is developed in this thesis and named as **BrIccEmis**, are presented. The general structure of the model is described before presenting an introduction to the required input data for the vacancy-cascade simulation. The calculation of transition energies based on the relativistic self-consistent-field Dirac-Fock method in the implementation of the **RAINE** code [112] is detailed and several approximations used in the vacancy-cascade modelling are also discussed.

The relevant published paper to this chapter is:

- “*A stochastic cascade model for Auger-electron emitting radionuclides*”,
Boon Q. Lee, H. Nikjoo, J. Ekman, P. Jönsson, A. E. Stuchbery and T. Kibédi,
International Journal of Radiation Biology **92**, pp.641-53 (2016).
This is Ref. [122] in the bibliography.
- “*Atomic Radiations in the Decay of Medical Radioisotopes: A Physics Perspective*”
B. Q. Lee, T. Kibédi, A. E. Stuchbery, and K. A. Robertson, Computational and
Mathematical Methods in Medicine **651475**, 14 pages (2012).
This is Ref. [89] in the bibliography.

3.1 General structure

A Monte Carlo model of the vacancy cascade, **BrIccEmis** was initially designed to simulate the vacancy cascade accompanying nuclear decay but it can be applied to other types of inner-shell ionisation if the distribution of initial vacancies is given. Figure 3.1 shows the general flow chart of **BrIccEmis** to generate X-ray and Auger energy spectra following atomic relaxation.

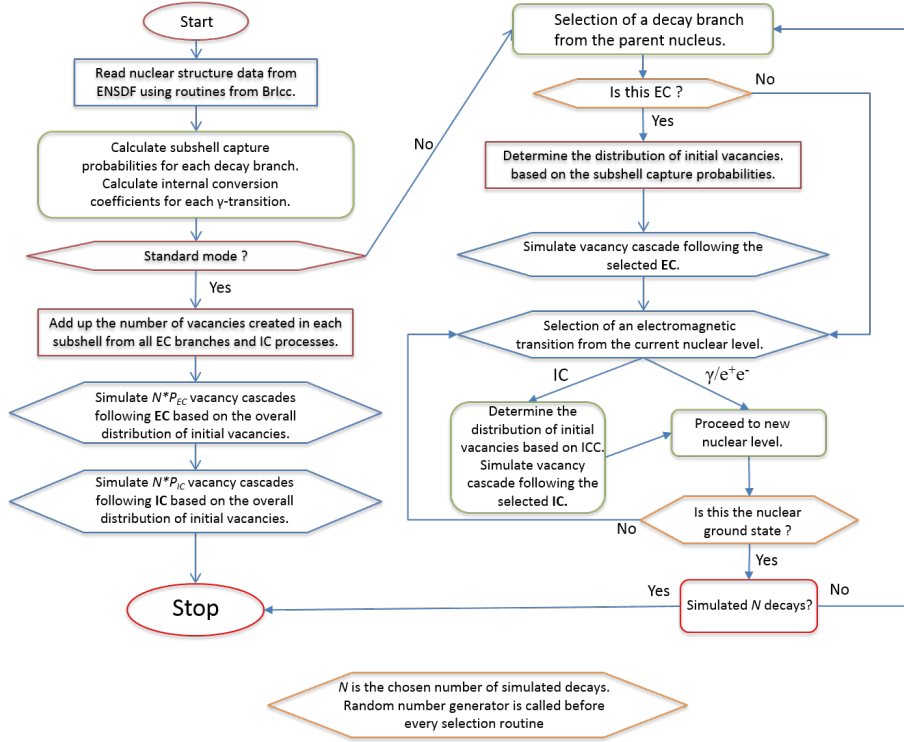


Figure 3.1: Flow chart of the *BrIccEmis* code. P_{EC} is the total electron capture branching ratio and P_{IC} is the total emission probability of conversion electron of the given radioisotope.

BrIccEmis uses routines from *BrIcc* [18] to extract nuclear structure data from the Evaluated Nuclear Structure File (ENSDF) [113]. The extracted information is used to determine the distribution of initial vacancies using the methods to be described in Section 3.2.1 and 3.2.1. There are two ways of simulating the vacancy cascade in *BrIccEmis*. The first one is named as “**Standard**” mode and the other one, which also simulate intra-nuclear cascade (INC), is called “**INC-VC**”

Standard mode is the widely adopted method of simulating the full atomic relaxation of radioisotopes. It has been used by all existing models detailed in Table 2.1. The method requires only the overall distribution of initial vacancies resulting from a radioactive decay by means of electron capture (EC) and/or internal conversion (IC), i.e. intra-nuclear cascades are ignored. The **Standard** mode is a two-step process. First, the EC decay and the corresponding vacancy-cascade simulation will be carried out. In the second step, the electromagnetic (EM) decay of the daughter nucleus and the atomic relaxation following IC will be simulated. In the calculations of energy spectra, shake-off and shake-up processes (vacancy creation) as a result of β decay are usually neglected since they mainly affect the valence shell [51]. All existing models, to the author’s knowledge, neglect the so-called “atomic structure effect” which was first revealed in the measurement of K X-rays of Holmium when Borchert et al. [123] compared the energy shift due to two different ways of creating a K-shell atomic vacancy, i.e. EC decay and photo-ionisation (see Section 3.4.2).

This effect increases the energies of X-rays and Auger electrons (in the order of several eV) due to an extra electron inherited from the parent atom in EC decay. **BrIccEmis** decouples the vacancy cascades accompanying EC and IC so as to account for this effect.

INC–VC mode allows a more detailed study since it tracks all radiation emitted accompanying each radioactive decay by simulating the intra-nuclear cascade as well. Examples of independent decay for radioiodines (^{123}I , ^{124}I , ^{125}I) using this mode are given in Appendix A.2. In the **INC–VC** mode, a random number is generated in the first step to determine the EC branch by which the parent nucleus decays. A vacancy cascade is then initiated based on the subshell capture probabilities of the selected EC branch. After the first vacancy cascade, a nuclear transition is selected randomly to proceed from the excited level that the previously chosen EC branch has decayed to. If an IC process is selected, another vacancy cascade is initiated following the ejection of the conversion electron. The simulation of the nuclear decay is concluded when the daughter nucleus has reached its nuclear ground state through a cascade of nuclear transitions. The atom is assumed to recover its neutral atomic configuration in between two successive nuclear transitions in the model (see Section 3.4.7).

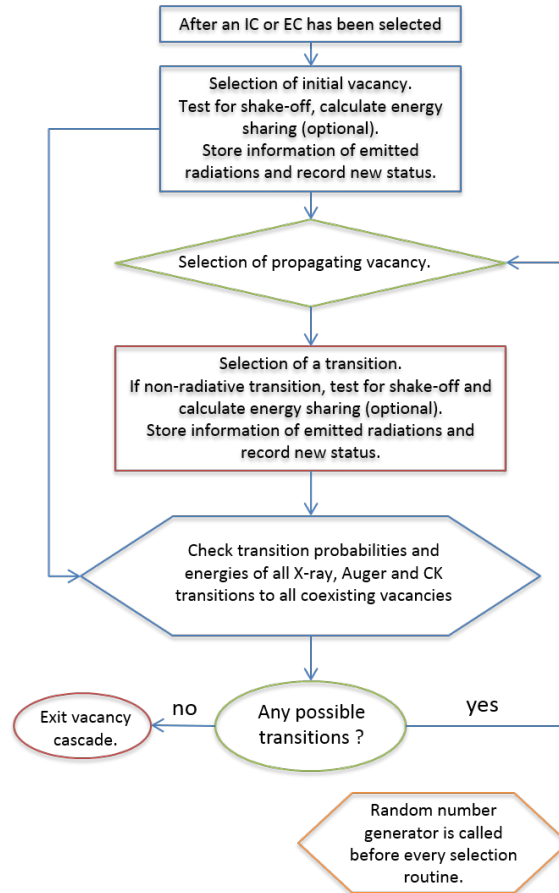


Figure 3.2: Flow chart of the PROP component of the **BrIccEmis** code.

Figure 3.2 shows the general flow chart of the vacancy cascade module of the **BrIccEmis**

code, which is currently named as **PROP**. **PROP** is called after a distribution of initial vacancies from an EC or IC process has been determined. In the first step of **PROP**, a random number is generated to select the location of the initial vacancy as a result of the selected EC or IC process. If the shake-off option is switched on, **PROP** will test for ejection of valence electrons as a result of the shake-off accompanying removal of an inner-shell electron by nuclear decay (see Section 3.2.3). Next, energies of all radiative and non-radiative transitions to fill the chosen initial vacancy that are tabulated in the Evaluated Atomic Data Library (EADL) [27] are checked and any energetically forbidden transitions are discarded. A transition energy is calculated as the difference between the total energies of the atom before and after a transition using the **RAINE** code ([112]), which is based on the relativistic self-consistent Dirac-Fock (DF) method (see Section 3.3). Since all atomic transition probabilities from the EADL are calculated for singly ionised atoms, a semi-empirical correction, which was first used by Krause and Carlson [85], is applied to modify them according to the actual number of electrons available in the participating subshells (see Section 3.4.3). Next, a random number is generated to determine the atomic transition that fills the chosen initial vacancy out of all energetically allowed transitions. If the transition is non-radiative, then an extra vacancy will be added to the system which may be filled in the subsequent transitions based on the atomic transition rates from EADL [27]. When there are multiple co-existing vacancies, a multiple-vacancy protocol will be deployed (see Section 3.4.4).

The protocol is similar when there are multiple vacancies. Energies of all transitions from the EADL are checked for each vacancy and any energetically forbidden transitions are discarded. Then a transition is selected from the list of energetically allowed transitions to fill a propagating vacancy based on their transition rates. The procedure to select the propagating vacancy will be discussed in Section 3.4.4. Eventually, all vacancies will have been propagated to the valence shell, or there will be no more energetically possible transitions to fill the remaining inner-shell vacancies. The program will then exit the **PROP** component and continue according to the protocol described in Figure 3.1.

3.2 Input data

3.2.1 Nuclear data

Nuclear structure data required for **BrIccEmis** are extracted from ENSDF [113]. ENSDF is maintained regularly and its use will ensure that the most up-to-date information is used to evaluate the nuclear event. **BrIccEmis** reads ENSDF using routines from **BrIcc** [18] to get all the necessary nuclear information for the simulation of the vacancy cascade following nuclear decay. This section describes the method used in **BrIccEmis** to determine the distribution of initial vacancies following an EC or IC process.

Electron Capture (EC) Probabilities

In order to obtain subshell EC probabilities, the most desirable way is to use experimental data. However, experimental values of sufficient accuracy are usually not available for many transitions. Alternatively, one can perform *ab initio* calculations of capture probabilities using EC theory from the fundamental paper presented by Bambynek et al. [98]. This is a consistent and accurate but very computationally expensive method of calculating the capture rates.

In view of these difficulties, Schönfeld reported a simple and practical way to calculate electron capture probabilities [17]. He compiled tables of theoretical cross section data required for the calculation of capture probabilities based on the same materials which were referred to by Bambynek et al. [98]. The tables contain a consistent and complete set of data to calculate electron capture probabilities for allowed and non-unique first forbidden transitions of radioisotopes with atomic numbers in the range of $3 \leq Z \leq 102$. Schönfeld [17, 97] estimated that the relative standard uncertainty of the values for K-shell, P_K , and L-shell, P_L , capture probabilities is smaller than 0.3% and 3%, respectively. Therefore, BrIccEmis adopted the Schönfeld's method [17] and his cross section data [97] to determine the subshell capture probabilities.

Leaving aside the effect of β^+ decay¹, which may compete with electron capture, the basic relation between the fractional electron capture probabilities is:

$$P_K + P_L + P_M + P_N + P_O = 1, \quad (3.1)$$

where P_X is the probability of capturing an electron in the X shell. If the ratios P_L/P_K , P_M/P_L , etc., are known, P_K can be calculated from

$$P_K = \left\{ 1 + \frac{P_L}{P_K} \left[1 + \frac{P_M}{P_L} \left(1 + \frac{P_N}{P_M} \left(1 + \frac{P_O}{P_N} \right) \right) \right] \right\}^{-1}. \quad (3.2)$$

Other parameters in Eq. (3.1) are then calculated using the following relations:

$$P_L = P_K \left(\frac{P_L}{P_K} \right), \quad (3.3)$$

$$P_M = P_L \left(\frac{P_M}{P_L} \right), \quad (3.4)$$

$$P_N = P_M \left(\frac{P_N}{P_M} \right), \quad (3.5)$$

$$P_O = P_N \left(\frac{P_O}{P_N} \right). \quad (3.6)$$

The dominant contributions to P_L , P_M , P_N and P_O come from L_1 , M_1 , N_1 and O_1 subshells; capture probabilities of L_2 , M_2 and N_2 have to be taken into account whereas

¹In most cases, the EC/ β^+ ratio is given in the ENSDF.

the contributions from other subshells can be neglected. The ratios in Eq. (3.3)-(3.6) are calculated using:

$$\frac{P_L}{P_K} = k_{LK} \left(\frac{\Delta E - E_{L_1}}{\Delta E - E_K} \right)^2, \quad (3.7)$$

$$\frac{P_M}{P_L} = k_{ML} \left(\frac{\Delta E - E_{M_1}}{\Delta E - E_{L_1}} \right)^2, \quad (3.8)$$

$$\frac{P_N}{P_M} = k_{NM} \left(\frac{\Delta E - E_{N_1}}{\Delta E - E_{M_1}} \right)^2, \quad (3.9)$$

$$\frac{P_O}{P_N} = k_{ON} \left(\frac{\Delta E - E_{O_1}}{\Delta E - E_{N_1}} \right)^2, \quad (3.10)$$

where the transition energy ΔE is given by $\Delta E = Q^+ - E_f$ with the decay energy Q^+ and energy of the nuclear level towards which the electron capture occurs, E_f , and E_K , E_{L_1} , E_{M_1} , E_{N_1} , and E_{O_1} are the atomic binding energies of the electrons in the subshell indicated by the subscript. The values of atomic binding energies used in *BrIccEmis* for Eq. (3.7)-(3.10) are extracted from [18, 124]. Equations (3.7) to (3.10) are only valid for closed atomic shells. The K (L_1 , M_1 , N_1 and O_1) shell is closed for elements with $Z \geq 2$ (4, 12, 20 and 38) although there are exceptions for N_1 (Cr and Cu) and O_1 (Nb, Mo, Ru, Rh, Pd and Ag) [97]. The values of the quantities k_{LK} , k_{ML} , k_{NM} and k_{ON} are taken from [97]. They are calculated from the ratios of the radial wave functions, $g_{L_1}^2/g_K^2$, etc., the ratios for the correction factors for overlap and exchange, $B(L_1)/B(K)$, etc., and the subshell ratios, P_{L_2}/P_{L_1} [17, 98]. For example, k_{LK} is given by:

$$k_{LK} = \frac{g_{L_1}^2}{g_K^2} \times \frac{B(L_1)}{B(K)} \times \left[1 + \frac{P_{L_2}}{P_{L_1}} \right]. \quad (3.11)$$

The uncertainties in these values are of the order of 1% but they increase with decreasing atomic number, Z [17]. For example, the uncertainty in k_{LK} for $Z = 4$ is estimated to be 25% [97]. The evaluation of k_{LK} , k_{ML} , k_{NM} and k_{ON} is detailed in Ref. [17].

After the ratios are made known, the fractional capture probabilities can then be calculated using Eqs. (3.3)-(3.6). Subshell capture probabilities (up to O_1) for allowed and non-unique first-forbidden transitions can be evaluated using:

$$P_{L_1} = \frac{P_L}{\left(1 + \frac{P_{L_2}}{P_{L_1}} \right)}, \quad P_{L_2} = P_{L_1} \frac{P_{L_2}}{P_{L_1}}, \quad (3.12)$$

$$P_{M_1} = \frac{P_M}{\left(1 + \frac{P_{M_2}}{P_{M_1}} \right)}, \quad P_{M_2} = P_{M_1} \frac{P_{M_2}}{P_{M_1}}, \quad (3.13)$$

$$P_{N_1} = \frac{P_N}{\left(1 + \frac{P_{N_2}}{P_{N_1}} \right)}, \quad P_{N_2} = P_{N_1} \frac{P_{N_2}}{P_{N_1}}, \quad (3.14)$$

$$P_{O_1} \approx P_O. \quad (3.15)$$

Subshell ratios P_{L_2}/P_{L_1} , P_{M_2}/P_{M_1} and P_{N_2}/P_{N_1} , which are equivalent to the ratios of the electron radial wave functions required in Eq. (3.12) to (3.14), are extracted from the 1995 PTB internal report [97]. There are no data for the O-shell capture ratio P_{O_2}/P_{O_1} so the author makes the assumption written in Eq. (3.15). This assumption will prohibit any possible initial vacancy in the O₂-subshell in high- Z radioisotopes but P_O is usually at least 2 orders of magnitude smaller than P_K and P_L so the imperfect treatment of splitting of P_O into P_{O_1} and P_{O_2} will not cause significant uncertainties.

Calculated EC probabilities of several radioisotopes using the Schönfeld method are presented in Table 3.1.

Table 3.1: Calculated electron capture probabilities P_K , P_L , P_M , P_N and P_O of several radioisotopes. Q^+ , E_f and P_{EC} are taken from ENSDF. No β^+ branch for these isotopes.

	Q^+ (keV)	E_f (keV)	P_{EC}	P_K	P_L	P_M	P_N	P_O
⁵⁵ Fe	231.21(18) ^a	0	1.00(0)E+0	0.8853	0.0983	0.0157	0.0007	
		126.0	1.30(10)E-9	0.8786	0.1040	0.0167	0.0007	
⁶⁷ Ga	1000.8(12)	0	9(9)E-3	0.8836	0.0989	0.0164	0.0011	
		93.310(5)	5.25(11)E-1	0.8834	0.0991	0.0164	0.0011	
		184.576(6)	2.27(1)E-1	0.8832	0.0993	0.0164	0.0011	
		393.527(7)	2.36(3)E-1	0.8824	0.0999	0.0165	0.0011	
		887.693(9)	2.81(4)E-3	0.8680	0.1119	0.0188	0.0013	
¹¹¹ In	862(5)	396.16(5)	5(5)E-5	0.8521	0.1182	0.0247	0.0050	
		416.63(5)	1.00000(5)E+0	0.8515	0.1187	0.0248	0.0051	
¹²⁵ I	185.77(6)	35.4925(5)	1.00E+0	0.8007	0.1561	0.0349	0.0079	0.0005
¹³¹ Cs	355(5)	0	1.00E+0	0.8336	0.1304	0.0288	0.0067	0.0005

^a 231.21(18) means 231.21 ± 0.18 .

Internal Conversion Coefficients (ICC)

Theoretical ICC for atomic numbers of $Z = 5$ -110 are taken from BrIcc, a tabulation released in the year of 2008 [18]. The ICC values in BrIcc are based on the RAINE code that performs the relativistic self-consistent-field (SCF) Dirac-Fock (DF) calculations using the so-called “frozen orbital” (FO) approximation [112]. The FO approximation accounts for the atomic hole left behind by the conversion electron assuming that there is insufficient time for a rearrangement of the atomic orbitals when the conversion electron is escaping the atom. Most of the previous ICC calculations have assumed that the atomic vacancy created in the conversion process is filled instantly (i.e. “no-hole (NH) approximation”). In the NH approximation, the electron wave functions for the initial and final states used in the ICC calculations are solved in the SCF of a neutral atom. When the kinetic energy of the conversion electron, E_k , is higher than 1 keV, one would argue that the atomic hole should be taken into account, as the average time required to fill the hole is significantly longer than the time it takes for the electron to escape from the atom. This argument applies to all elements and atomic shells [125]. Moreover, high-precision experimental conversion coefficients indicate that the effect of the atomic vacancy should be taken into account

[126]. In general, the inclusion of the atomic hole in the calculations of ICC increases the ICC values [125].

A comparison between theoretical ICC calculated using the FO approximation from *BrIcc* and experimental ICC that are known to better than 5% accuracy, shows that the average difference, $\bar{\Delta}_{ICC}(Theory - Exp)$ is -0.72 ± 0.21 % [18].

3.2.2 Atomic data

Atomic transition probabilities

The evaluated Atomic Data Library (EADL) [27] tabulates atomic transition probabilities calculated using the relativistic Dirac-Hartree-Slater (DHS) method within the jj coupling scheme for atoms with $Z = 5$ to 100 (See Appendix A.3 for a brief review of different coupling schemes). These probabilities are calculated for the singly-ionised systems. The values reported in the EADL are mainly taken from the publications by Scofield [102, 127, 128], for radiative transitions, and Chen et al. [29, 105, 129, 130, 131], for non-radiative transitions. The transition probabilities are tabulated in the form of relative intensities (i.e. branching ratios from an atomic vacancy) for each (sub)-shell.

Three major assumptions used in the EADL tabulations, which may limit the accuracy of resulting spectra, are:

- The transition probabilities available in the EADL database are calculated by assuming that the (sub-)shell from which the electron will be removed contains the full number of electrons, as in the neutral atom,
- The EADL database does not include the probability of shake-off and shake-up,
- Transitions between some outer atomic subshells, though weak transitions, are not included in the current version of the EADL database.

A review paper published in 2010 by Augelli et al. [132] suggested that some contents of the EADL database do not reflect the state-of-the-art. This statement is based on the finding by Pia et al. [133] that Hartree-Fock (HF) calculations appear more accurate than the DHS method used by the EADL for the radiative transitions. Augelli et al. [132] also reported that there are some accidental errors in the assembly of the library. The flawed values are affected by errors amounting to orders of magnitude differences with respect to original publications by Scofield [102, 127, 128]. Nevertheless, they also stressed that EADL is still an invaluable tool for Monte Carlo simulation, thanks to its wide collection of atomic parameters in one consistent environment [132].

Since the EADL contains the most complete atomic data available to date, and it describes the total transition rates correctly, it is adopted in the *BrIccEmis* code. The K and L-shell radiative rates in the EADL are accurate to about 10% but the inaccuracies

can be bigger than 30% for transitions involving outer-shells with energy below 100 eV. EADL's Auger transition widths, Γ_A , for inner-shell vacancies are known to better than 15% if the inner shells do not decay by Coster-Kronig (CK) transitions; if CK transitions are in competition, the Auger widths can be larger than experiments by a factor of 2 [27]. These uncertainties directly affect the radiative and non-radiative yields (see Eq. 2.17). In BrIccEmis, the relative intensity of six KLL Auger transitions in the jj coupling scheme for $18 \leq Z \leq 96$ are corrected based on the interpolated values from Chen et al. [134], which are calculated in the intermediate coupling scheme and so account for the effect of configuration interaction. Nevertheless, the relative intensity of KLL to all K-shell transitions is kept fixed as the absolute KLL transition rates are not sensitive to different calculational methods. Table 3.2 compares the KLL transition rates from Chen et al. [134] and EADL [27] for selected elements. Both methods mostly agree to within 4% for $Z \leq 50$. For $Z \geq 50$, the deviation can be beyond 7%, but the KLL transitions do not contribute significantly ($< 10\%$) to the total in these elements.

Table 3.2: Comparison of KLL transition rates from Chen et al. [29] and EADL [27] for selected atomic numbers Z .

Z	Chen IC-CI (s^{-1})	EADL (s^{-1})	IC-CI/EADL (%)	KLL RI ^a (%)
18	7.373×10^{14}	7.124×10^{14}	103.5	74.5
36	1.074×10^{15}	1.071×10^{14}	100.3	26.9
54	1.307×10^{14}	1.215×10^{14}	107.6	7.2
80	1.962×10^{14}	2.033×10^{14}	96.5	2.5
96	2.973×10^{14}	3.140×10^{14}	94.7	1.8

^aRelative intensity (RI) of KLL to all K-shell transitions is extracted from the EADL.

Level widths

Radiative, Γ_R , and non-radiative, $\Gamma_{NR} = \Gamma_A + \Gamma_{CK}$ widths described in Eq. (2.17) are also tabulated in the EADL. The total width, $\Gamma = \Gamma_R + \Gamma_{NR}$ for each subshell is a useful atomic parameter to control the selection of the propagating vacancy (see Section 3.4.4) and estimate the duration of the vacancy cascade. Moreover, these widths can also be used to estimate the natural line widths of X-ray and Auger transitions.

The accuracy of total widths of K and up to N₇ shells reported in the EADL has been extensively studied by Campbell and Papp [135]. They concluded that the EADL predictions appear satisfactory for atomic subshells, where CK contributions do not contribute or contribute only to a small extent to the overall widths; for these subshells, they provided estimates of uncertainty in the range of 5-15%, while for other subshells, the uncertainties are estimated to be in the range of 25-30%. This conclusion is consistent with the estimates given by the authors of EADL [27].

3.2.3 Shake-off

Carlson and Nestor [14] calculated the total shake-off probability from a given atomic orbital as the result of a sudden vacancy in one of the atomic shells using the terminology detailed in Section 2.3. They calculated the shake-off probabilities for neon, argon, krypton and xenon as a function of the location of the initial vacancy. Additionally, Mukoyama and Taniguchi [53] calculated similar shake-off probabilities for elements with Z between 2 and 36, and for all subshells as a function of an initial vacancy in the K and L shells. Both calculations agreed well with each other. These shake-off probabilities are calculated assuming the problem can be treated with single-electron wave-functions rather than as a many-body problem. The calculated probabilities are no longer valid when sudden vacancies in a given shell have promoted additional electron ejection from the same shell. In these cases, electron correlations must be included for the calculations to agree with experiments. Recent calculations by Lowe et al. [56] and Pham et al. [136] have shown that electron correlations, especially for open-shell atomic systems, are required to reproduce experimental data. For example, Lowe et al. calculated the shake-off probabilities from the $3d$ subshell for Sc ($Z=21$) to be 42%, which agrees well with experiments, but is about 7-times bigger than the calculations by Mukoyama and Taniguchi [53].

Another critical assumption used in the sudden approximation is that the outgoing electron, which gives rise to additional electron ejection from outer shells, must have sufficient velocity. Mukoyama and Taniguchi [53] have shown that the agreement between theory and experimental data from inner-shell photoionisation depends on the photon energy. Moreover, Carlson [35] has shown that the shake-off probability following the photoionisation of the K-shell in neon increases with photon energy up to a threshold energy after which it becomes constant.

In *BrIccEmis*, the shake-off probabilities for noble gases are adopted from the publication by Carlson and Nestor [14]². These values are only valid when the energy available for electron shake-off is substantially greater than the threshold for electron shake-off, E_0 :

$$C = E/E_0 \gg 1, \quad (3.16)$$

where E is the energy of outgoing primary radiation, i.e. conversion electron, Auger electron, photoelectron, etc., without the shake-off process. Carlson [35] provided an empirical value of 3 for C which was derived from the shake-off accompanying photoelectrons. *BrIccEmis* does not account for the shake-up effect (i.e. assuming pure shake-off). The validity of this assumption will be discussed later in Section 3.4.5.

²Kochur et al. [137] has compiled the shake-off probabilities of non-noble gases for $Z \leq 71$ based on the method of Carlson and Nestor. Only shake-off probabilities for up to the $4d$ (N_5) subshell were reported in their work. However, for elements with $Z \geq 37$, caution must be taken when the values from Ref. [137] are used because significant contributions from $5s$ and subshells above to the total shake-off probability will be neglected.

The shake-off process accompanying EC is neglected as the values from Carlson and Nestor [14] are not applicable for the vacancy creation due to EC. After an electron is captured from the atom in EC decay, outer-shell electrons experience almost no change in the central Coulomb field due to shielding, thus they have negligible probability to escape from their original orbitals. In contrast, the inner-shell electrons, especially those very close to where the vacancy is created, have non-negligible but still small ($< 1\%$) probability to be shaken away from their original orbitals.

3.3 Calculations of transition energies

In order to treat the vacancy cascade correctly, a realistic electron configuration must be taken into account for the calculation of atomic transition energies. An accurate but expensive method of calculating the energies of emitted X-rays or Auger and CK electrons is to solve the relativistic Dirac equations before and after each transition required by the Monte Carlo calculations. This method of calculating the energies was first suggested by Pomplun et al. [37]. In the current model (**BrIccEmis**), the binding energies for all (sub-)shells and the total energy of the bound electrons for any electron configuration are calculated using the **RAINE** code [112]. The **RAINE** code is based on the relativistic self-consistent-field Dirac-Fock (DF) method in the jj coupling scheme (see Appendix A.3 for a brief review of different coupling schemes). The **RAINE** code requires an input in the form of a list of subshell occupation numbers. Since the vacancy cascade is a multi-step process, this way of calculating transition energies permits an energy check of all possible transitions at each propagation step during the cascade.

The atomic relaxation scheme allows an excited atom to return to its ground state by emitting atomic radiations. Each atomic radiation carries a unique amount of energy away. The energy of atomic radiation emitted at the N^{th} step of the vacancy cascade is calculated using the following relation:

$$E^{A/X} \Big|_N = E_T^{S=N} - E_T^{S=N+1} \quad N \in [1, N_{Max}], \quad (3.17)$$

where A denotes a non-radiative transition and X defines a radiative transition, S is the propagation step and E_T is the total energy of the atomic level at a given step, S . The vacancy cascade stops after N_{Max} steps when there are no more kinematically-allowed transitions. For example, $E_T^{S=1}$ is the total energy of the atomic level right after a nuclear decay, while $E_T^{S=N_{Max}}$ is the total energy of the atomic level at the end of the vacancy cascade. Therefore, the residual potential energy can be defined as:

$$E_{residual} = E_T^{neut} - E_T^{S=N_{Max}}. \quad (3.18)$$

The **RAINE** code, which does not take into account the Breit electron interaction and quantum electrodynamics (QED) corrections³, overestimates the atomic binding energies, especially the K shell [140, 141], and this causes the calculated transition energies to be higher than the experimental values. Differences in K-shell binding energies between the **RAINE** code and experimental values from Sevier [18, 124] are increasing with the atomic number Z (see Table 3.3 and Figure 3.3). The overestimation is fairly consistent for other subshells in each atomic principal shell. Furthermore, overestimation seems to be smaller for atoms with closed atomic shells. For example, the overestimation in binding energies of $_{54}\text{Xe}$ is smaller than $_{53}\text{I}$. It is important to note that the experimental values tabulated by Sevier [124] originated from several different experimental techniques which required either (a) conversion from their original units, such as x-ray units (xu) or Angströms ($\text{\AA}$), to eV energy units, or (b) reliance on calibration standards of varying accuracies for those data obtained directly in eV units. Therefore, there would always be some intrinsic uncertainties in the reported neutral atom binding energies.

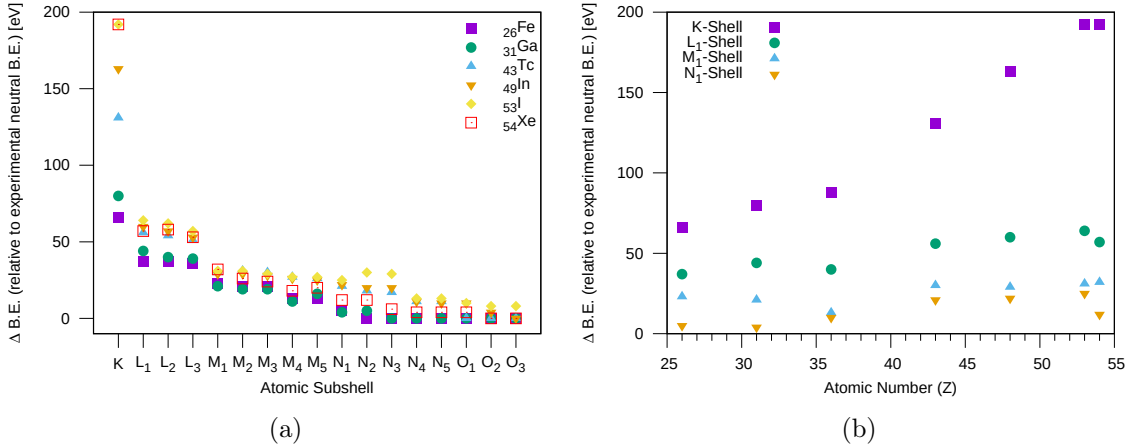


Figure 3.3: Vertical axes indicate the difference between **RAINE**'s [112] neutral binding energies and experimental data (in eV). Experimental binding energies are extracted from [18, 124]. The difference in binding energies is shown as a function of (a) atomic subshell and (b) atomic number for selected subshells.

Data storage

The stochastic nature of the vacancy cascade can result in many possible electron configurations. For example, simulation of the vacancy cascade in Hg has produced, so far, 8.69 million unique configurations⁴. As mentioned earlier, the total electron energy of

³A group at the University of Melbourne, which is led by Prof. Chris Chantler, has been successful in improving the current state of the calculation of QED corrections for multi-electron atoms [138] and getting convergence of the Breit interaction in the self-consistent and configuration-interaction approaches [139]. These theoretical advances would potentially help guide the new development of the **RAINE** code in order to account for the Breit interaction and QED corrections properly.

⁴A unique configuration here is defined as a unique combination of subshell occupation numbers. For example, $1s^2 2s^2 2p_{1/2}^2 2p_{3/2}^3$ and $1s^2 2s^2 2p_{1/2}^1 2p_{3/2}^4$ are two different unique configurations in the fluorine-like

Table 3.3: Differences in calculated atomic shell binding energies (B.E.) and experimental results [18, 124] ($\Delta B.E. = \text{Experimental B.E.} - \text{Theoretical B.E.}$) for radioisotopes investigated in this thesis. All values in the table have been rounded to the nearest eV.

$\Delta B.E. = B.E.(calc.) - B.E.(exp.)$ (eV)							
Subshell	Atomic number, Z						
	26	31	36	43	49	53	54
K	66	80	88	131	163	192	192
L₁	37	44	40	56	60	64	57
L₂	37	40	38	54	57	62	58
L₃	36	39	36	51	53	57	53
M₁	23	21	13	30	29	31	32
M₂	21	19	12	31	29	31	26
M₃	21	19	12	30	28	29	24
M₄	13	11	8	27	26	27	18
M₅	13	16	8	26	25	27	18
N₁	5	4	5	21	22	25	12
N₂		5	10	18	20	30	12
N₃			13	17	20	29	6
N₄				17	20	29	6
N₅				11	10	13	4
O₁					10	10	4
O₂					4	8	0
O₃						8	0

each configuration is calculated by the DF method using the **RAINE** code. **BrIccEmis** uses a tree-like data structure to store the total electron energies of encountered configurations *on-the-fly*. Figure 3.4 shows a schematic description of the data structure used in **BrIccEmis**. Two parameters used to group the electron configurations are (a) the number of existing vacancies and (b) the subshell of the lowest vacancy (or deepest hole) of the given configuration. Each *grandchild* of the tree is an array that stores all electron configurations which share the same grouping parameters. The length of each grandchild array is different and pre-calculated based on possible combinations for its associated electron configurations. A fail-safe routine, which will extend the length of a grandchild, is in place to prevent data overflow during simulation. All configurations that have been calculated will be stored together with their corresponding total electron energies in a binary file which can be assessed in the future simulations.

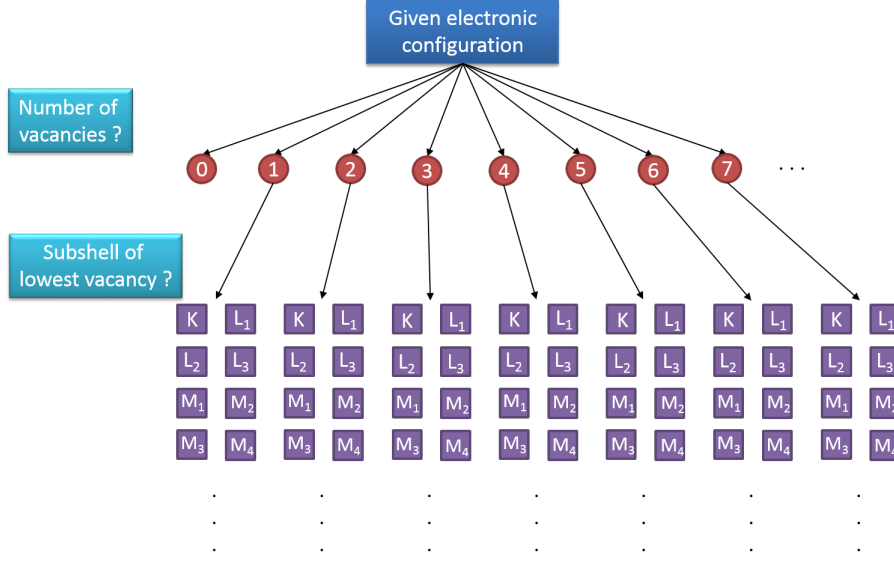


Figure 3.4: Schematic description of the tree-like data structure used to store the total electron energies of all possible configurations.

3.4 Approximations

3.4.1 Subshell electron capture (EC) probabilities of forbidden transitions

The method of calculating subshell electron capture probabilities described in Section 3.2.1 is only applicable to allowed and nonunique first-forbidden transitions. In these types of EC decay, capture from subshells with orbital angular momentum, $l \geq 2$ (i.e. $L_3, M_3, M_4, \dots$) is negligibly small [98]. For unique first-forbidden transitions, the exponent in the energy factor in Eqs. (3.7)-(3.10) should be 4 instead of 2. Moreover, the contribution of subshells with $l \geq 2$, especially L_3 , need to be included. Therefore, $P_L = P_{L_1} [P_{L_2}/P_{L_1} + P_{L_3}/P_{L_1}]$ and P_{L_3}/P_{L_1} for a closed-shell system is [98]:

$$\frac{P_{L_3}}{P_{L_1}} = \frac{p_{L_3}^2 \beta_{L_3}^2}{\beta_{L_1}^2} \cdot \frac{B_{L_3}}{B_{L_1}} \cdot \frac{(\Delta E - E_{L_3})^2}{(\Delta E - E_{L_3})^4}, \quad (3.19)$$

where p_x is the electron momentum and β_x is the Coulomb amplitude of orbital x . **BrIccEmis** is currently unable to calculate P_{L_3} , but if the factors $p_{L_3} \beta_{L_3}$ for different atomic numbers Z become available, then the code will be able to account correctly for the unique first-forbidden transitions. These transitions are treated in the same way as the allowed and nonunique first forbidden transitions for now. For instance, ^{124}I has non-negligible unique first forbidden transitions and the contribution from the L_3 subshell is 1.7% according to Nikjoo et al. [118] (see Table 3.4).

Capture probabilities of higher-order forbidden unique and nonunique transitions decrease very rapidly with increasing degree of forbiddenness [98]. If these types of EC tran-

Table 3.4: Initial vacancy distributions for the decay of ^{124}I . The contribution from L_3 is neglected in `BrIccEmis` as it treats the unique first-forbidden transitions in the same way as the allowed and nonunique first-forbidden transitions. This table has been published in Ref. [122].

Radionuclide	^{124}I		
	Nikjoo [118]	This work	
		EC	IC
Atomic Subshell	EC	EC	IC
K	0.644	0.661	0.00303
L_1	0.105	0.0867	0.00034
L_2	0.003	0.0023	0.00004
L_3	0.017	–	0.00003
M_1	0.004	0.0184	0.00007
M_2	–	0.0005	0.00
M_3	–	–	0.00
M_4	–	–	0.00
M_5	–	–	0.00
N_1	–	0.004	0.00001
N_2	–	0.0001	0.00
N_3	–	–	0.00
O_1	–	0.0002	0.00
Sum	0.773	0.7732	0.00352

sitions are reported in a given ENSDF, they will be treated in the same way as the allowed and nonunique first forbidden transitions, but they generally have very small branching ratios due to low capture rates, so they will not affect the simulation significantly.

3.4.2 Atomic structure effect

Larkins’ tabulation of semi-empirical Auger-electron energies [28] is the “Gold Standard” in the literature for comparison with experimental values. However, in the early 1990s, the Auger group at the Laboratory of Nuclear Problems, JINR, Dubna, observed that the absolute energies of $\text{L}_3\text{M}_4\text{M}_5$ Auger lines from the EC decays of several elements in the lanthanide series, which include ^{155}Gd , ^{159}Tb , ^{165}Ho and ^{171}Yb , are systematically higher than Larkins’ values by around 20 eV [142]. In the subsequent years, the same group measured the differences in absolute energies of the dominant $\text{L}_3\text{M}_4\text{M}_5$ Auger lines in Xe and Gd from EC and β^- decays, respectively [143, 144]. They found that the differences are 9.4 ± 1.1 eV and 16.6 ± 2.9 eV in Xe and Gd, respectively, which could not be explained purely by chemical shifts. Around the same time, Bulgakov et al. [145] proposed that this is due to the vacancy cascade being too fast for the atom to rearrange itself into the electronic ground state of the daughter’s nuclear charge following an EC decay, before an atomic transition occurs. This effect is particularly significant for transitions involving K and L shells where atomic holes have very short lifetimes ($\sim 10^{-17}$ - 10^{-15} s). As mentioned

earlier, this so-called “atomic structure effect” was first revealed in the measurements of K X-rays of Holmium [123]. The impact of this effect is generally small and it only increases the atomic transition energy in the order of eV due to the additional valence electron inherited from the atomic system of the parent nucleus in an EC decay.

When will the atom rearrange itself into the electronic structure of the daughter’s nuclear charge? This remains an open question. At this stage, *BrIccEmis* assumes the vacancy cascade accompanying an EC decay happens in the atomic structure of the parent’s nuclear charge while the vacancy cascade following atomic-hole creation by IC occurs in the electron configuration of daughter’s nuclear charge.

3.4.3 Krause-Carlson statistical correction

The relative transition rates from the EADL are calculated based on the single-hole approximation. However, multiple vacancies may exist during the vacancy cascade thus rendering EADL relative rates no longer valid. Nevertheless, calculation of transition rates from first principles for “random” electron configurations that could be encountered in the vacancy cascade is a huge task and beyond the scope of this thesis. Therefore, *BrIccEmis* adopts a statistical correction, first used by Krause and Carlson [85] and subsequently in all of the Monte Carlo calculations of Auger spectra [37, 46, 88], to adjust the relative rates based on the number of electrons available to a particular transition. The correction is equivalent to keeping transition rates per electron constant and ignoring the changes in the effective Coulomb field whenever an additional inner-shell electron has been removed during the vacancy cascade. This introduces a slight error since Auger transition rates, which are the most likely events during the vacancy cascade, are rather insensitive to such changes [146, 147]. However, this is not true for Coster-Kronig (CK) transition rates, which change more rapidly due to the changes in the effective Coulomb field; but the ratio of the CK transition to the Auger transition remains almost constant [85, 148]. Nevertheless, future study of changes in transition rates due to multiple vacancies using the state-of-the-art atomic structure packages such as *Grasp2K* [149, 150] and *Ratip* [151] as well as *FAC* [152] is important to validate or invalidate this statistical correction.

The correction factor K for a non-radiative transition, XYZ, is

$$K = \left(\frac{O_c(Y)!}{(O_c(Y) - 1)!} / \frac{O_n(Y)!}{(O_n(Y) - 1)!} \right) \times \left(\frac{O_c(Z)!}{(O_c(Z) - 1)!} / \frac{O_n(Z)!}{(O_n(Z) - 1)!} \right), \quad (3.20)$$

where O_c and O_n are the sub-shell occupation numbers of cascading and neutral atoms, respectively, and ! is the factorial. Eq. (3.20) only applies when Y and Z are not in the same subshells. If Y and Z are in the same subshells, the following correction factor K will be used:

$$K = \left(\frac{O_c(Y)!}{2!(O_c(Y) - 2)!} / \frac{O_n(Y)!}{2!(O_n(Y) - 2)!} \right). \quad (3.21)$$

M3	x x x o		
M2	x o	$KL_{12}^L(A)$	1
M1	x x	$KL_{23}^L(A)$	1/4
L3	x o o o	$KL_{33}^L(A)$	0
L2	x x	$KL_{32}^M(A)$	$1/2 * 1/4$
L1	x x	$KM_{33}^M(A)$	$(3!/2!(3-2)!)/ (4!/2!(4-2)!)$
K	x o	$KL_3^X(X)$	1/4

Figure 3.5: Transition rate correction factors based on Eqs. (3.20)-(3.22) for a hypothetical vacancy distribution. An electron is labelled as $\times$ while a vacancy is denoted as an $\circ$. The letters X and A in the parenthesis indicate a radiative and non-radiative transition, respectively. The image is extracted from Ref. [38].

For radiative transitions, XY, the correction factor K is simply the ratio

$$K = O_c(Y)/O_n(Y). \quad (3.22)$$

This factor is also applied for shake-off processes from orbital Y. Examples of using this statistical correction are shown in Figure 3.5.

3.4.4 Selection of the propagating vacancy

There is no simple rule about which atomic vacancy is to be filled first when there are multiple co-existing vacancies. The following describes four options that have been included into BrIccEmis.

A. Highest vacancy

The first option is to select the vacancy that is located in the highest subshell. This way of selecting the propagating vacancy was first proposed by Mukoyama [153]. He concludes that this method always results in a higher mean charge than for a simulation that fills the lowest hole first.

B. Lowest vacancy

The second option is to occupy the inner-shell vacancy before the outer-shell hole is filled. This scheme reduces the relative abundances of highly ionised states compared to the first option [153] because many outer-shell transitions become energetically forbidden by the time when outer-shell holes are selected to propagate. Pomplun [87] claimed that this selection scheme gives the best agreement with experimental charge distributions using his model.

C. Random

The third option is to pick a vacancy to propagate at each step of the cascade at random. This scheme gives a result that is in between *A* and *B*. This way of selecting the propagating vacancy uses the total relative transition rate of a given vacancy, which is equal to unity in a singly-ionised atom, as the weighting factor in the selection process. For example, in a hypothetical scenario where there are two co-existing vacancies, if the relative transition rates of all possible transitions to fill each of the two vacancies are not affected by the Krause-Carlson (KC) correction and energy restriction, they both have equal probability to be selected. It is important to note that the total relative transition rate of a given vacancy diminishes as the cascade progresses due to the implications of the KC correction factor and energy restrictions.

D. Lifetime-weighted

The fourth option is to utilise the natural widths Γ extracted from EADL [27] as the weighting parameter in selecting the propagating vacancy. The natural width of a subshell *X*, Γ_X , is related to the total rate of all transitions that fill a vacancy in *X*, A_X by $\Gamma_X = A_X \hbar$ [154]. According to this selection scheme, an atomic vacancy with a larger natural width will have a higher probability to be chosen to propagate. As the total relative transition rate diminishes due to the implications of the KC correction factor and energy restriction, it is assumed that the natural width decreases in the same way. Additionally, this option allows for time-dependent simulation.

The mean life τ of an ion with multiple vacancies is given by:

$$\frac{1}{\tau} = \sum_i \frac{1}{\tau_i} = \sum_i A_i, \quad (3.23)$$

where τ_i and A_i are the partial mean life and corresponding partial decay rate of a vacancy in shell *i*. The probability that the ion survives for time *t* before the next step of the vacancy cascade is given by an exponential distribution:

$$P(t) = e^{-t/\tau}. \quad (3.24)$$

A random number *RN* is then used to decide whether the ion proceeds to the next step of the cascade at different timepoints based on $P(t)$; if *RN* is bigger than $P(t)$, then an atomic radiation is emitted from the ion at time *t*. One can then calculate the build up of atomic radiations as a function of time. Figure 3.6 shows the evolution of the emission of atomic radiation following the EC decay of isolated ^{125}I ; only the vacancy cascade following the creation of inner-shell vacancies by EC is considered. All transitions that fill the K-shell vacancy proceed extremely fast and complete by 0.5 fs. Outer-shell non-radiative

transitions are completed by 1 ps, while low-energy X-rays could take at least 1 μ s to be fully released.

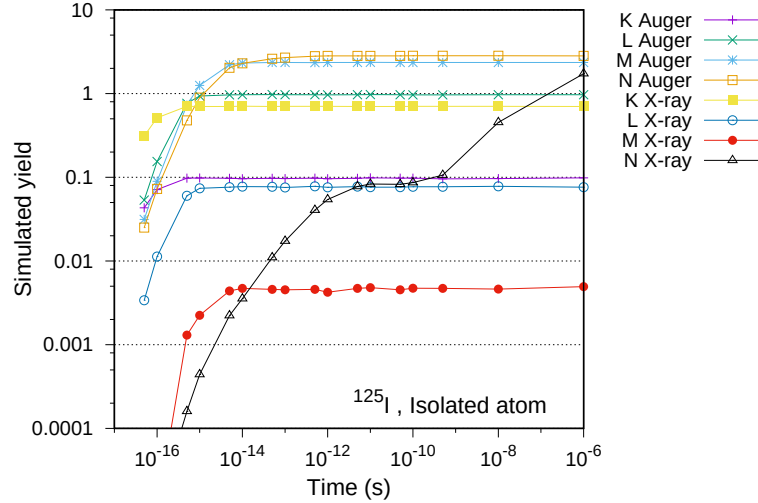


Figure 3.6: Simulated atomic radiation yields, where yield is defined as the number of radiations emitted per decay, following the EC decay of ^{125}I (only the first cascade) as a function of time in the isolated-atom approach. Coster-Kronig transitions are included in the Auger transitions. All values are calculated from 100,000 simulations. This figure has been published in Ref. [122].

Comparison of the residual charge-state distribution simulated using all four selection schemes will be presented in Chapter 5.

3.4.5 Shake-off probabilities

Shake-off accompanying non-radiative transitions

Shake-off probabilities following Auger processes are not well tabulated in the literature. Hence an approximation is needed to utilise the shake-off probabilities following photoionisation (or IC) from the literature. Shake-off probabilities following photoionisation and Auger processes for krypton from Krause and Carlson [85] are shown in Table 3.5. Probabilities of shake-off accompanying Auger transitions $1s-2p2p$ are slightly larger than half of shake-off probabilities following $2p$ photoionisation for most subshells. Since the shake-off probability always diminishes as the vacancy cascade progresses (i.e. atom becomes more ionised), **BrIccEmis** approximates the probabilities of shake-off accompanying Auger transitions, XYZ as the *half* of the average of shake-off probabilities for subshells Y and Z following photoionisation. This crude approximation will no longer be needed when probabilities for shake-off accompanying Auger are made available in the future. The radiative Auger effect (RAE) described in Section 2.3.2 is not considered in **BrIccEmis** as the K X-rays are less dominant for $Z < 30$ where RAE is more noticeable [66]. RAE becomes non-negligible when radiative transitions involving valence electrons are dominant,

but such a case is extremely rare. Even though the K-shell radiative width increases with atomic number, Z , the valence electrons are also getting further away from the K shell, i.e. the radiative transitions involving valence electrons are low in probability, at the same time.

Table 3.5: Probabilities (in percent) for electron shakeoff from the various subshells, n_s , in krypton, as a result of sudden vacancy in orbital, n_0 , due to photoionisation and Auger processes, calculated by Krause and Carlson [85]. For Auger processes, the new vacancies are denoted by the last two subshells involved.

$n_s \backslash n_0$	1s	2s	2p	1s-2p2p
1s	0.004	0.000	0.000	0.003
2s	0.053	0.003	0.007	0.019
2p	0.27	0.026	0.034	0.016
3s	0.25	0.085	0.100	0.042
3p	1.22	0.523	0.59	0.22
3d	3.66	3.39	3.48	2.88
4s	2.23	1.85	1.98	1.46
4p	13.90	12.18	12.37	7.23
Total	21.6	18.1	18.6	11.9

Exclusion of the shake-up effect

Since in this thesis shake-off is mainly used in the study of residual charge-state distributions following the vacancy cascade, *BrIccEmis* uses the approximation in which P_{nl} from Eq. (2.33) is attributed entirely to shake-off processes while it is supposed to be equally distributed between the shake-off and shake-up processes, especially for the valence shell. Also, the Auger transitions involving shaken-up electrons have much lower probabilities than those that do not involve these excited electrons, which means these shaken-up electrons are mostly spectating the subsequent atomic transitions. This approximation is applicable because loosely bound shaken-up electrons may eventually be shaken-off in the subsequent Auger transitions and this approach is also used by Kochur et al. [54]. The situation may be different when the system is highly ionised but P_{nl} values are usually very small in those cases (i.e., valence electrons are then more tightly bound).

3.4.6 Energy distribution of shake-off electrons

The following energy relationship exists for a sudden vacancy created by internal conversion (or photoionisation):

$$h\nu = E_i + E_0 + E_e + E_s, \quad (3.25)$$

where $h\nu$ is the γ -energy, E_i is the ionisation potential for removing the first electron (conversion electron or photoelectron), E_0 is the threshold energy for shake-off (i.e. ion-

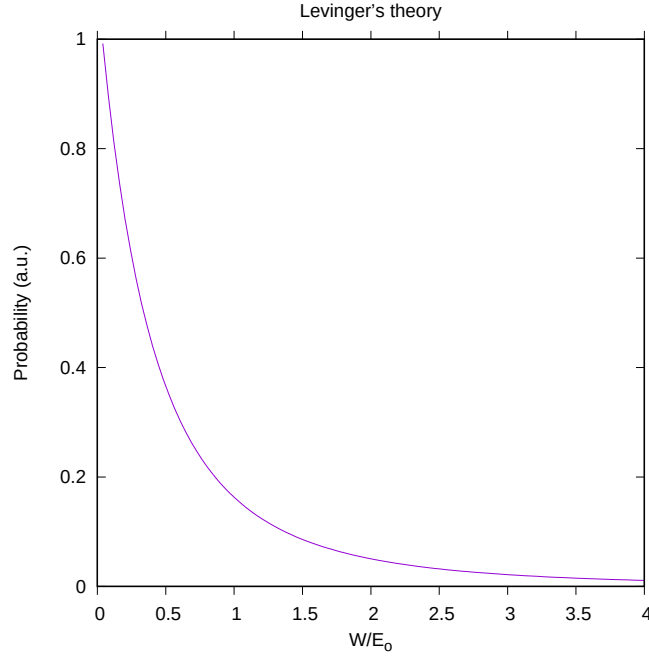


Figure 3.7: Calculated shake-off energy spectrum following atomic ionisation using Levinger's theory. W is the positive energy of the emitted shake-off electron; E_0 is the threshold energy for electron shake-off.

isation potential for removing the electron from its original orbital), E_e and E_s are the kinetic energies of the first and shake-off electrons, respectively. Strictly speaking, the two electrons emitted are indistinguishable. See Figure 2.7, which is extracted from Carlson [52], for the energy spectrum following L-shell photoionisation in neon. Krause et al. [155] showed that the theoretical kinetic energy distribution from shake-off electrons for β decay, which was derived by Levinger [156], compares favourably with experiment. According to Levinger's theory, the transition probability in terms of the positive energy W of the shake-off electron:

$$P(n')dW = 2^6 Z^{-4} (1 - e^{-2\pi n'})^{-1} n'^8 (n'^2 + 1)^{-4} e^{-4n' \cot^{-1} n'} dW, \quad (3.26)$$

where

$$n' = (E_0/W)^{1/2}, \quad (3.27)$$

E_0 is in atomic units ($e^2/a_0 = 27.2114$ eV). Figure 3.7 shows the $P(W)$ vs W based on Eq. (3.26).

BrIccEmis adopts Levinger's theory to approximate the energy of shake-off electrons when it is considered in simulations. For the shake-off accompanying Auger transitions, the following energy relationship exists:

$$E_A = E_0 + E_A^s + E_s, \quad (3.28)$$

where E_A^s and E_A are the Auger transition energies with and without the shake-off process, respectively. The energy distribution of the shake-off electron that accompanies an EC decay is can be approximated using Eq. (3.26), but the electron is now sharing its energy with a neutrino rather than an electron. Nevertheless, BrIccEmis ignores such shake-off transitions due to their small probability (see Section 3.2.3).

3.4.7 Fast and slow neutralisations

Note: Shake-off electrons are neglected in this section.

The two aforementioned neutralisation schemes during the vacancy cascade have been discussed in Section 2.4.3. There is no consensus in the literature on which approach should be adopted, due to the lack of experimental evidence for the time-scale of electron transfer from DNA or proteins to an Auger-emitter. BrIccEmis does not make any preference and it can simulate the vacancy cascade using either neutralisation scheme for comparison. However, it is assumed that charge neutralisation happens between successive nuclear transitions, which are usually at least one order of magnitude longer in the lifetimes (> 1 ps) than most atomic transitions, apart from outer-shell X-rays, regardless of whether fast or slow neutralisation is considered, unless the code is used for the study of residual charge-state distributions. This assumption is not valid in rare cases wherein the level half-life of the daughter nucleus ($\sim 10^{-14}$) is comparable to the time-scale of the vacancy cascade. An example of one of these rare cases is reported in Bulgakov et al. [121].

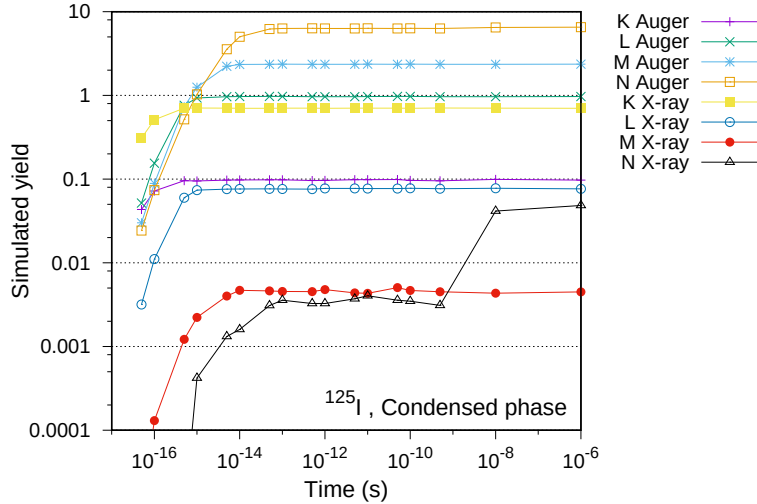


Figure 3.8: Simulated atomic radiation yields, where yield is defined as the number of radiation emitted per decay, following the decay of ^{125}I as a function of time in the condensed-phase approach. Coster-Kronig are included in the Auger transitions. All values are calculated from 100,000 simulations.

If an atom is in vacuum, the isolated-atom assumption is the only applicable approach and it serves well in understanding the residual distribution of charge states following the vacancy cascade. However, if the atom is in a molecular environment, whether or not charge

neutralisation happens *during* the vacancy cascade becomes important. Figure 3.8 shows the evolution of the emission of atomic radiations following the EC decay of condensed-phase ^{125}I (i.e. fast neutralisation); only the first cascade is considered. In comparison to the vacancy cascade simulated for isolated atom (Figure 3.6), there are significantly more N-shell Auger and CK transitions emitted, while the low-energy X-rays are suppressed. For the condensed-phase assumption, **BrIccEmis** assumes that any atomic hole created in the valence shell, which usually has a binding energy ranging from zero to several eV, is immediately filled after its creation during the vacancy cascade. For most elements, only the valence shell has no transition rate reported in EADL [27] because these transitions are energetically forbidden. However, for some transition metals and lanthanides, transitions that fill an atomic vacancy in the d or f shell are missing from EADL, which might be due to computational limitations at the time of tabulation, or energy restriction. For example, no transitions to fill $N_{4,5}$ ($4d_{3/2,5/2}$) subshells of cadmium, which have neutral binding energies of 9.7 and 9 eV [124], are reported in EADL. Thus the simulation of the vacancy cascade using values from EADL will always leave the cadmium atom in an ionised state. Since the condensed-phase assumption is mainly used to ensure that the atom is completely neutralised at the end of the cascade process [38, 84], **BrIccEmis** assumes vacancies in subshells where no known atomic transitions reported in EADL, i.e. the vacancies cannot be filled by means of atomic transitions, to be part of the valence shell so they will be immediately neutralised if they are ionised in the condensed-phase assumption.

Table 3.6: Comparison of calculated Auger and Coster-Kronig (CK) electron yields for selected medical radioisotopes. Assumptions used in each program can be found in Table 2.1. Values in the last column are calculated from 100,000 simulations. This table has been partially published in Refs. [89, 122, 157, 158].

	RADAR ^a	DDEP ^a	Eckerman & Endo (MIRD)	Howell	Stepanek	Pomplun	BrIccEmis ^b
	[78, 79]	[80]	[81]	[38]	[86, 90, 117]	[87, 88]	
Charge neutralisation	No	No	Yes	Yes	No/Yes	No/Yes	No/Yes
Auger and CK electron yield per nuclear decay							
^{55}Fe (2.73 y)	2.00	2.00	5.05	5.10	-/5.07		4.14/4.95
^{67}Ga (3.2612 d)	2.28	2.28	4.96	4.70			4.61/4.85
$^{99\text{m}}\text{Tc}$ (6.007 h)	0.12	0.13	4.41	4.00		3.00 ^c /-	3.52/4.39
^{111}In (2.805 d)	1.14	1.16	7.43	14.7	-/6.05	5.63 ^d /-	5.84/7.17
^{123}I (13.22 h)	1.06	1.08	13.7	14.9		7.30 ^c /-	7.38/12.3
^{125}I (59.4 d)	1.77	1.78	23.0	24.9	8.92/14.5	12.2/18.3	11.9/20.0
^{131}Cs (9.689 d)	0.47		10.9				6.68/10.1
$^{131\text{m}}\text{Xe}$ (11.84 d)	0.82	0.83	10.6				6.11/9.48
^{201}Tl (3.04 d) ^(g)	0.77	0.61	20.9	36.9			10.9/18.8

^aOnly transitions from K and L shells.

^bResults simulated using the random selection of propagating vacancy.

^cShake-off electrons included.

^dValue extracted from Stepanek et al. [117].

Table 3.6 compares the calculated Auger and Coster-Kronig (CK) electron yields for selected medical radioisotopes using both approaches in **BrIccEmis** to the literature. In

general, values calculated using *BrIccEmis* agree well with the existing programs that simulate the full vacancy cascade, apart from Howell [38]. For ^{111}In , Howell's Auger and CK electron yield (14.7) is more than double the current value (7.17). This is due to the fact that Howell extended EADL data for ^{111}In to include transitions that fill $N_{4,5}$ subshells, which are missing in EADL as mentioned earlier, by applying the so-called "Kassis rule" [44]. The Kassis rule estimates the transition probabilities based on the number and distribution of electrons available for the transition. No quantum mechanical considerations are made in this first approximation. Stepanek was able to reproduce a similar result to Howell's data when the author used the $Z/Z + 1$ rule (not shown, see Ref. [117]). Similarly, the overestimation in electron yield of the decay of ^{201}Tl is likely to be due to the use of the $Z/Z + 1$ rule.

For radioiodines, Howell's values are higher than all other programs because he included the O-shell non-radiative transitions based on the Kassis rule. However, these transitions are calculated to have negative energies using the relativistic self-consistent Dirac-Fock (DF) method. The calculations were performed using the *Grasp2K* and *Ratip* programs by Jörgen Ekman of Malmö University (private communication). Therefore, these O-shell non-radiative transitions are unphysical and should not be included in the simulation of the vacancy cascade.

Impact on energy spectrum

Figure 3.9 shows the kinetic energy spectra of photons and electrons for the decays of ^{123}I , ^{124}I and ^{125}I in the isolated-atom and condensed-phase conditions. ^{123}I ($T_{1/2} = 13.27$ hours) decays by electron capture exclusively to the nuclear excited states in ^{123}Te followed by γ emission (84%) and IC (16%) and its decay scheme is shown in Figure 3.10a. Similarly, ^{125}I ($T_{1/2} = 59.4$ days) decays by EC followed by γ emission (7%) and IC (93%) to ^{125}Te , and its decay scheme is shown in Figure 2.6. On the other hand, ^{124}I ($T_{1/2} = 4.19$ days) decays via either positron emission (22.7%) or EC (77.3%) to the nuclear excited states (65%) or the nuclear ground state (35%) of ^{124}Te . The excited states of ^{124}Te de-excite in many steps to the ground state mostly by γ emission. A small number of conversion electrons (0.35%) is also generated during this de-excitation. The decay scheme of ^{124}I is shown in Figure 3.10b. Initial-vacancy distributions of ^{123}I and ^{125}I decays are shown in Tables A.5 and A.6, respectively, while the distribution of initial vacancies created in the decay of ^{124}I is reported in Table 3.4.

No difference between the isolated-atom and condensed-phase conditions is observed for energies above 1000 eV. However, there are large differences at energies below 100 eV. This is the region of N-shell transitions, which strongly depend on the electron population of the valence shell. This observation is consistent with the results published by Pomplun [87].

Table 3.7 shows the average spectrum of ^{124}I for the isolated-atom and condensed-phase

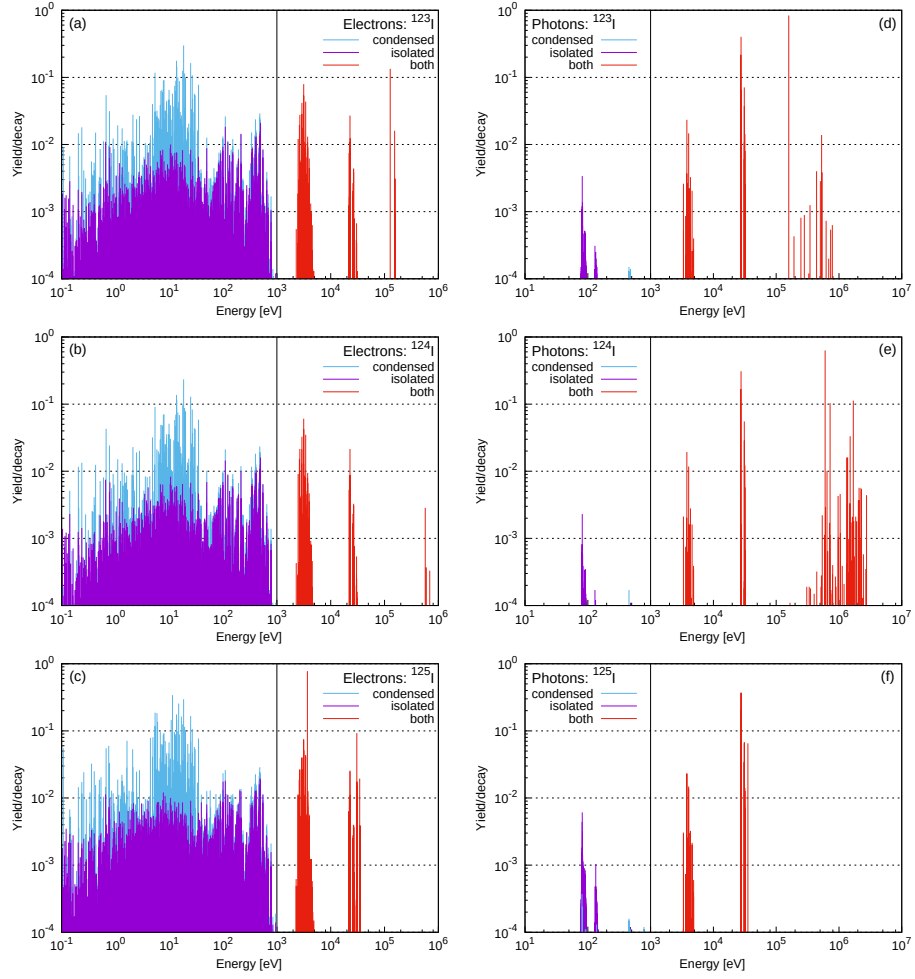


Figure 3.9: Energy spectra of photons and electrons of ^{123}I (a, d), ^{124}I (b, e) and ^{125}I (c, f) calculated in the isolated-atom (light blue) and condensed-phase (purple) approximations. Since the two approximations have negligible differences for energies above 1000 eV, results from the isolated-atom are used to present both approximations in this region (red). At energies below 1000 eV, electron spectra for the condensed phase are plotted behind their counterparts while photon spectra for the same approximation are plotted in front of their counterparts. These spectra show that there are large discrepancies at energies below 100 eV. This figure has been published in Ref. [122].

approximations. Nuclear radiations with yields of less than 10^{-2} have been excluded. The two approximations deviate from each other only for N-shell transitions. In general, the use of the condensed-phase approximation leads to an enhancement in the emission of N-shell Auger and Coster-Kronig transitions. In comparison, some N-shell non-radiative transitions become energetically forbidden in the isolated-atom condition due to the build up of charges during the vacancy cascade. This leads to the greater yields of N-shell X-rays as they remain energetically possible near the end of the vacancy cascade. These observations are well represented in Figures 3.6 and 3.8.

Impact on multiplicity distributions

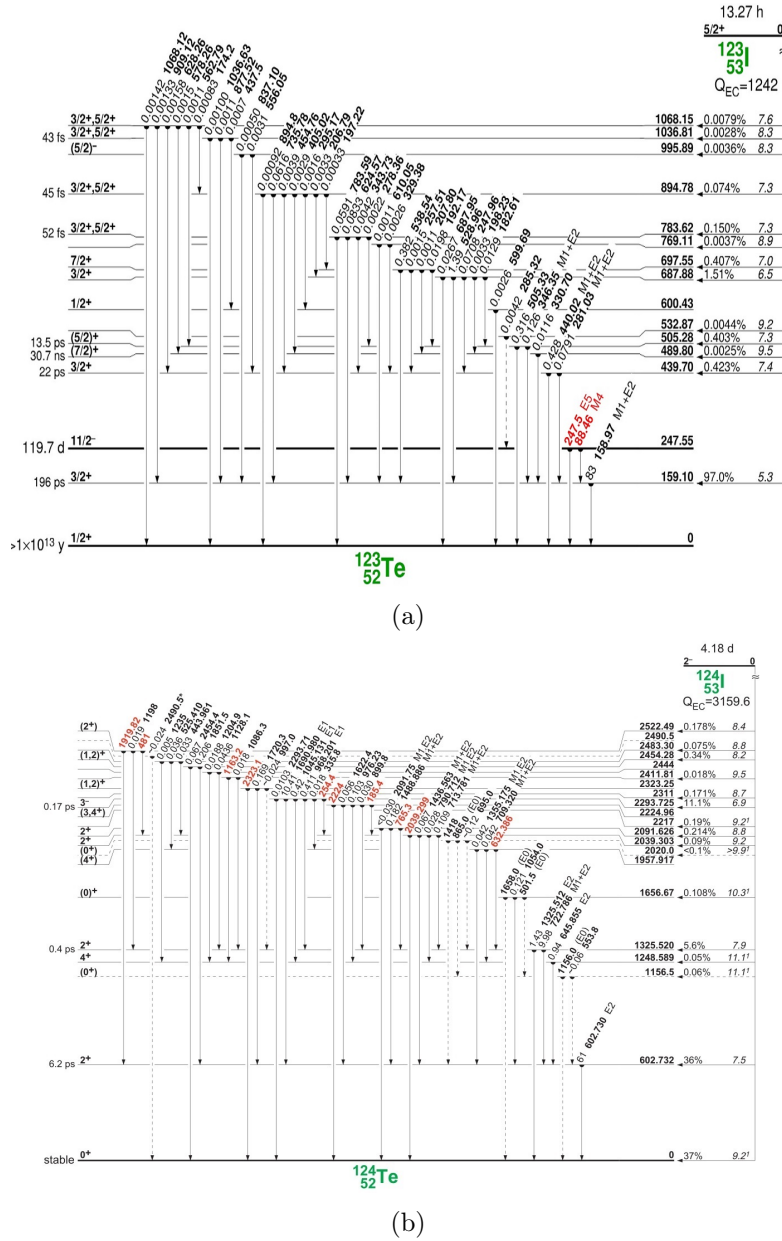


Figure 3.10: Decay schemes of (a) ^{123}I and (b) ^{124}I . Images extracted from Ref. [99]

Figure 3.11 displays the multiplicity distributions of emitted electrons and photons for ^{123}I , ^{124}I and ^{125}I in the isolated-atom and condensed-phase approximations. Figure 3.11a - 3.11c shows that many more electrons can be emitted per decay of radioiodine when the atoms are allowed to have an extra supply of electrons from the environment during the vacancy cascade in the condensed-phase approach. In comparison, there is no difference between the two approximations for the multiplicity distributions of the emitted photons. It is important to note that most of the low-energy radiative transitions proceed very slowly ($\geq 10^{-9}$ s) and they are excluded. If these transitions were taken into account, the isolated-atom approximation would be expected to yield more photons thus widening the

Table 3.7: Average spectrum per decay of ^{124}I for isolated-atom and condensed-phase approximations. Values calculated from 100,000 simulations using the random choice of propagating vacancies and with the cascade stopped when no vacancy has mean lifetime shorter than 10^{-12} s. This table has been published in Ref. [122].

Process	Isolated atom		Condensed phase			
	Average energy (eV)	Yield	Average energy (eV)	Yield	$\Delta E/E$ (%)	$\Delta I/I$ (%)
γ_1	$6.02 \cdot 10^5$	$6.30 \cdot 10^{-1}$	$6.03 \cdot 10^5$	$6.31 \cdot 10^{-1}$	0	+0.2
γ_2	$6.46 \cdot 10^5$	$1.02 \cdot 10^{-2}$	$6.46 \cdot 10^5$	$1.00 \cdot 10^{-2}$	0	-2.0
γ_3	$7.23 \cdot 10^5$	$1.02 \cdot 10^{-1}$	$7.23 \cdot 10^5$	$1.03 \cdot 10^{-1}$	0	+1.0
γ_4	$1.33 \cdot 10^6$	$1.59 \cdot 10^{-2}$	$1.33 \cdot 10^6$	$1.60 \cdot 10^{-2}$	0	+0.6
γ_{21}	$1.69 \cdot 10^6$	$1.14 \cdot 10^{-1}$	$1.69 \cdot 10^5$	$1.13 \cdot 10^{-1}$	0	-0.9
γ_{44}	$1.38 \cdot 10^6$	$1.59 \cdot 10^{-2}$	$1.38 \cdot 10^5$	$1.61 \cdot 10^{-2}$	0	+1.3
γ_{54}	$1.51 \cdot 10^6$	$3.18 \cdot 10^{-2}$	$1.51 \cdot 10^5$	$3.33 \cdot 10^{-2}$	0	+4.7
Auger KLL	$2.26 \cdot 10^4$	$5.41 \cdot 10^{-2}$	$2.26 \cdot 10^4$	$5.44 \cdot 10^{-2}$	0	+0.6
Auger K LX	$2.66 \cdot 10^4$	$2.40 \cdot 10^{-2}$	$2.66 \cdot 10^4$	$2.43 \cdot 10^{-2}$	0	+1.3
Auger KXY	$3.04 \cdot 10^4$	$2.57 \cdot 10^{-3}$	$3.04 \cdot 10^4$	$2.51 \cdot 10^{-3}$	0	-2.3
CK LLX	$2.85 \cdot 10^2$	$1.03 \cdot 10^{-1}$	$2.87 \cdot 10^2$	$1.03 \cdot 10^{-1}$	+0.7	0
Auger LMM	$3.05 \cdot 10^3$	$4.93 \cdot 10^{-1}$	$3.05 \cdot 10^3$	$4.94 \cdot 10^{-1}$	0	+0.2
Auger LMX	$3.68 \cdot 10^3$	$1.39 \cdot 10^{-1}$	$3.68 \cdot 10^3$	$1.38 \cdot 10^{-1}$	0	-0.7
Auger LXY	$4.30 \cdot 10^3$	$9.33 \cdot 10^{-3}$	$4.30 \cdot 10^3$	$9.82 \cdot 10^{-3}$	0	+5.3
CK MMX	$9.71 \cdot 10^1$	$5.22 \cdot 10^{-1}$	$1.04 \cdot 10^2$	$5.33 \cdot 10^{-1}$	+7.1	+2.1
Auger MXY	$4.13 \cdot 10^2$	$1.30 \cdot 10^0$	$4.29 \cdot 10^2$	$1.30 \cdot 10^0$	+3.9	0
CK NNX	$1.89 \cdot 10^1$	$9.30 \cdot 10^{-1}$	$2.18 \cdot 10^1$	$1.37 \cdot 10^0$	+15.3	+47.3
Auger NXY	$2.05 \cdot 10^1$	$1.46 \cdot 10^0$	$1.51 \cdot 10^1$	$4.35 \cdot 10^0$	-26.3	+198.0
X-ray K_{α_1}	$2.76 \cdot 10^4$	$3.11 \cdot 10^{-1}$	$2.76 \cdot 10^4$	$3.11 \cdot 10^{-1}$	0	0
X-ray K_{α_2}	$2.73 \cdot 10^4$	$1.68 \cdot 10^{-1}$	$2.73 \cdot 10^4$	$1.68 \cdot 10^{-1}$	0	0
X-ray K_{β_1}	$3.11 \cdot 10^4$	$5.57 \cdot 10^{-2}$	$3.11 \cdot 10^4$	$5.55 \cdot 10^{-2}$	0	-0.4
X-ray K_{β_2}	$3.18 \cdot 10^4$	$1.71 \cdot 10^{-2}$	$3.18 \cdot 10^4$	$1.80 \cdot 10^{-2}$	0	+5.3
X-ray K_{β_3}	$3.10 \cdot 10^4$	$2.79 \cdot 10^{-2}$	$3.10 \cdot 10^4$	$2.87 \cdot 10^{-2}$	0	+2.9
X-ray KM_+	$3.17 \cdot 10^4$	$1.80 \cdot 10^{-3}$	$3.17 \cdot 10^4$	$1.71 \cdot 10^{-3}$	0	-5.0
X-ray L	$3.94 \cdot 10^3$	$6.00 \cdot 10^{-2}$	$3.93 \cdot 10^3$	$6.02 \cdot 10^{-2}$	-0.3	+0.3
X-ray M	$5.64 \cdot 10^2$	$3.39 \cdot 10^{-3}$	$5.38 \cdot 10^2$	$3.09 \cdot 10^{-3}$	-4.6	-8.8
X-ray N	$9.07 \cdot 10^1$	$7.17 \cdot 10^{-2}$	$1.01 \cdot 10^2$	$1.76 \cdot 10^{-3}$	+11.4	-97.5

photon multiplicity distributions.

A zigzag pattern is observed in the multiplicity distributions of emitted electrons in the condensed-phase approximation (see Figure 3.11a-c). This observation is, in fact, an *artificial* consequence of the approximation. Non-radiative transitions dominate in the atomic shells other than the K shell so it is more likely to have only non-radiative transitions after a non K-shell vacancy is created. The condensed-phase approximation could prevent many non-radiative transitions from becoming energetically forbidden through the neutralisation of valence vacancies. Examples 4 and 12 in Table A.1 (in Appendix A.2) show that only non-radiative transitions are present after a non K-shell initial vacancy is created due to the EC decay of ^{123}I . The number of vacancies is multiplied by two after filling out all the vacancies created in the previous propagation step by non-radiative transitions, thus increasing the number of emitted electrons by an even number. Therefore, the vacancy cascade favours an odd number of emitted electrons in the condensed-phase approximation. Examples 1, 5, 6, 7, 8, 10, 11, 13 in Table A.1 show that the vacancy cascade following EC predominantly emits an odd number of electrons after the emission of a K-Xray. In contrast, the vacancy cascade initiated by IC prefers an even number of emitted electrons after the emission of the conversion electron. The comparison of the multiplicity distri-

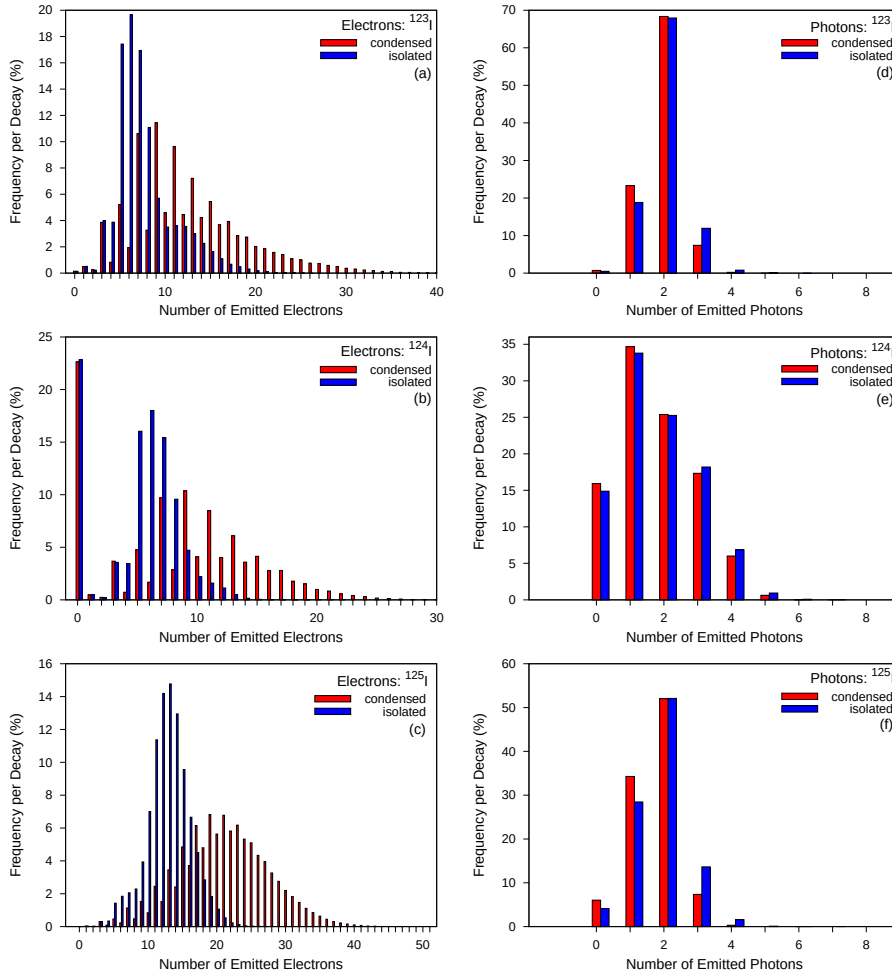


Figure 3.11: Multiplicity distributions of emitted electrons and photons for ^{123}I (a, d), ^{124}I (b, e) and ^{125}I (c, f) in the isolated-atom and condensed-phase approximations. The condensed phase yields wider distributions than the isolated atom for electrons while the two approximations give very similar distributions for photons. This figure has been published in Ref. [122].

butions of emitted electrons calculated in the condensed-phase approximation following the EC and IC for isotopes of iodine, bromine, antimony and barium is shown in Figure 3.12a-d. Only the multiplicity distributions for the isotopes of barium do not exhibit a similar zigzag pattern. This is due to the fact that barium and caesium (daughter element of ^{133}Ba) only have 2 and 1 P-shell valence electrons, respectively, in the condensed-phase approximation. The neutralisation of valence vacancies in the two elements would not be sufficient to prevent some non-radiative transitions from becoming energetically forbidden.

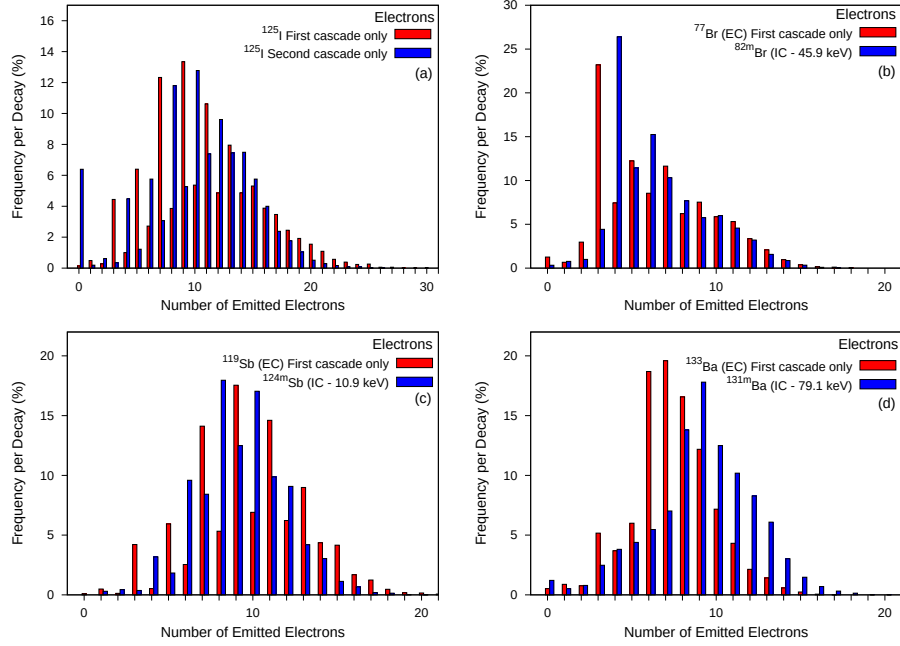


Figure 3.12: Frequency distributions of the number of emitted electrons for isotopes of (a) iodine, (b) bromine, (c) antimony and (d) barium in the condensed-phase approximations. Figures a-c show that the vacancy cascade following EC favours an odd number of emitted electrons while the vacancy cascade following IC prefers an even number of emitted electrons. Unlike the other elements, the multiplicity distributions of barium isotopes do not exhibit a zigzag pattern due to the lack of valence electrons. This figure has been published in Ref. [122].

3.5 Energy balance

Isolated atom

Since `BrIccEmis` evaluates the energy of each possible transition and eliminates all energetically forbidden transitions *before* a transition is selected to continue the vacancy cascade, the energy balance is ensured in the isolated-atom approximation. Therefore, the following energy relationship exists for the vacancy cascade accompanying the internal conversion process:

$$E_{\gamma} = E_{ic} + E_{NR} + E_R + E_s + \Delta V, \quad (3.29)$$

where E_{γ} is the nuclear transition energy, E_{ic} is the energy of the conversion electron, E_{NR} is the total energy of the emitted Auger and Coster-Kronig (CK) electrons, E_R is the total energy of released X-rays, E_s is the energy of shake-off electrons, and ΔV is the potential energy associated with the residual ion. For the vacancy cascade following electron capture, the energy balance equation is

$$Q^+ - E_i = E_X + E_{NR} + E_R + E_s + \Delta V, \quad (3.30)$$

where Q^+ is the disintegration energy, E_i is the energy of final nuclear level in the daughter nucleus and E_X is the binding energy of the capture electron in the X shell.

Condensed phase

In the condensed-phase assumption, the system is assumed to capture electrons from the continuum to fill atomic holes in the valence shell, thus the atom is provided additional energy and it can emit more atomic radiations. For the vacancy cascade following internal conversion, the energy-balance equation is

$$E_\gamma = E_{ic} + E_{NR} + E_R + E_s - \sum_{i=1}^n E_{N,i}, \quad (3.31)$$

where $E_{N,i}$ is the potential energy required to fill a valence vacancy and n is the total number of valence vacancies encountered, which is equal to the total number of electrons emitted, during the cascade process. Similarly, the following equation holds for electron capture decay:

$$Q^+ - E_i = E_X + E_{NR} + E_R + E_s - \sum_{i=1}^n E_{N,i}. \quad (3.32)$$

Several authors [84, 117, 140] treated every neutralisation event in the condensed-phase approximation as the so-called *free-bound* radiative recombination, where a low-energy X-ray carries away the energy that is equal to the binding energy of the neutralised vacancy. However, such a computational treatment could be misleading as femtosecond spectroscopy has revealed that vibrational relaxation is competing with electronic relaxation in electron transfer at the molecular level within the time-scale of the vacancy cascade [119, 120]. The correct treatment of the neutralisation of valence vacancies, which is highly dependent on the environment, is currently beyond the scope of BrIccEmis. Hence, the code only reports bound-bound radiative transitions and assumes the energy is dissipated in the form of *heat* in a neutralisation event.

3.6 Summary

In summary, a Monte Carlo model of vacancy cascade BrIccEmis, which has been developed by the author at the Australian National University for this thesis, has been described in detail. The model is primarily used for simulating atomic energy spectra following nuclear decays, but it can also be used to simulate the vacancy cascade following other ionisation processes such as photoionisation. Nuclear structure information in the format of ENSDF [113] for an isotope is the required input for the model to start a simulation. The model uses atomic transition probabilities from EADL [27] and adopts the Dirac-Fock energies calculated using the RAINE code [112]. Impacts of several approximations used in the model

have also been discussed in this chapter. The handling of the shake-off process, where the shake theory from Carlson and Nestor [14] is adopted, has been described but it is mainly used for the near closed-shell atomic systems. The next two chapters will focus on the validation of the model against experimental data before its applications in fission physics and medical physics are presented in Chapters 6 and 7.

Acknowledgements

The author thanks Dr. Filip Kondev of Argonne National Laboratory for his valuable comments on the development of `BrIccEmis`. In addition, the author is grateful for Dr. Jörgen Ekman of Malmö University for his calculations of atomic transition rates in xenon using the `Grasp2K` and `Ratip` packages.

Energy spectra

Overview

This chapter compares transition energies calculated using the **RAINE** code, which is adopted by **BrIccEmis**, for the singly-ionised systems against the semi-empirical Auger-electron energies as well as the latest theoretical X-ray energies. The simulated energy spectra for the decays of ^{125}I and ^{131}Cs , and the M_5 -subshell photoionisation of atomic xenon will be compared to available experimental data for the benchmarking of **BrIccEmis**. The comparison of simulated average radiation spectra for ^{123}I and ^{125}I decays with the literature values will also be presented. All simulated results shown in this chapter are based on 100,000 simulated events unless specified otherwise.

4.1 Background

Energy spectra of Auger-electron emitting medical radioisotopes are essential for dosimetric calculations to quantify the biological damage delivered to the target [118, 159, 160]. In the past three decades, several authors published calculated emission spectra of selected radioisotopes using either deterministic or Monte Carlo computational methods [38, 81, 86, 87, 118]. However, there is a large scatter in these published spectra, particularly in the yields and energies of outer-shell transitions, as they were calculated based on different approaches toward evaluation of the vacancy cascade. For other less common but emerging radioisotopes such as ^{140}Nd ($T_{1/2}=3.4$ d) and ^{161}Tb ($T_{1/2}=6.9$ d), no emission data have yet been made available. ^{140}Nd is a pure Auger emitter, i.e. ^{140}Nd decays to the ground state of ^{140}Pr and only emits Auger electrons and X-rays, and its short-lived daughter ^{140}Pr is a positron emitter, which can be used for PET (positron emission tomography) while ^{161}Tb is a β /Auger emitter that has been shown to provide an enhanced antitumour effect compared to ^{177}Lu in an *in vivo* study [161]. These radioisotopes have more suitable half-lives and better electron-to-photon dose ratios than most of the commonly used Auger emitters. They can now be produced for medical research at Institut of Laue-Langevin (ILL) and CERN-Medical Isotopes Collected from ISOLDE (MEDICIS) [162, 163]. Thus, a vacancy-cascade model that can generate the energy spectrum for a wide range of elements

in a consistent physics framework is essential for the research into Auger-electron targeted radiotherapy.

The development of **BrIccEmis** was originally initiated by the demand for an improved computational route to address the limitations of the existing programs and databases for the calculations of energy spectra for medical radioisotopes [11]. Chapter 5 will demonstrate that charge-state distributions simulated by **BrIccEmis** are in fairly good agreement with experiments given the complexity of the vacancy cascade. This means that **BrIccEmis** has captured most of the physics of the vacancy cascade. However, it is also evident from the comparison of electron binding energies calculated by the **RAINE** code, which is adopted by **BrIccEmis** for the calculations of transition energies, with accepted values that **RAINE** overestimates for most atomic shells (see Figure 3.3). Therefore, systematic comparisons of energy spectra calculated using **BrIccEmis** with experiments and accepted values are essential for the benchmarking of the model.

In this chapter, a comparison of atomic transition energies calculated in the singly-ionised systems with the accepted values for elements from $Z = 18$ to $Z = 99$, with an interval of 3 elements, is presented. In addition, comparison of the simulated Auger-energy spectra from **BrIccEmis** with the recent measurements for ^{125}I and ^{131}Cs is shown. The simulated energy-correlation spectrum for the M_5 -subshell photoionisation of atomic xenon is also benchmarked against the experimental spectrum published by Suzuki et al. [71]. Last, but not least, the simulated average radiation spectra of ^{123}I and ^{125}I are compared with values published in the literature.

4.2 Auger electrons

In the first part of this section, the Auger energies calculated using the **RAINE** code [112] for the singly-ionised systems are compared to values from Larkins' tabulation [28] of semi-empirical Auger-electron energies as well as the Evaluated Atomic Data Library (EADL) [27], in which the transition energies were derived from theoretical neutral binding energies using the following relation [84]:

$$E_{XYZ} = \text{B.E.}(X) - (\text{B.E.}(Y) + \text{B.E.}(Z)), \quad (4.1)$$

where E_{XYZ} is the energy of the non-radiative transition XYZ and $\text{B.E.}(X)$, $\text{B.E.}(Y)$ and $\text{B.E.}(Z)$ are the binding energies of the X , Y and Z subshells, respectively. In the second part of this section, Auger-energy spectra calculated using the **BrIccEmis** code are compared to several measured spectra.

4.2.1 Comparison with tabulations

Larkins' tabulation of semi-empirical (SE) Auger-electron energies for $10 \leq Z \leq 100$ is the most comprehensive source for detailed comparisons. Auger energies in the Larkins' tabulation are for solid systems and referenced to the Fermi level, with the exception of the elements Ne, Cl, Ar, Br, Kr, Xe and Rn. In contrast, the transition energies from the **RAINE** code and EADL are for the atomic system and referenced to the vacuum level. It is important to note that the jj coupling scheme is used in both the **RAINE** code and EADL but Larkins assumed the intermediate coupling (see Appendix A.3 for brief review of different coupling schemes). For example, the KL_2L_3 Auger transition has two energy lines in the intermediate coupling, i.e. KL_2L_3 ($^3P_1$) and KL_2L_3 ($^1D_2$), but only has one state in the jj coupling. In this section, the energy of an Auger transition from the Larkins' tabulation used for comparison with the **RAINE** code and EADL is the average value of all intermediate coupling states.

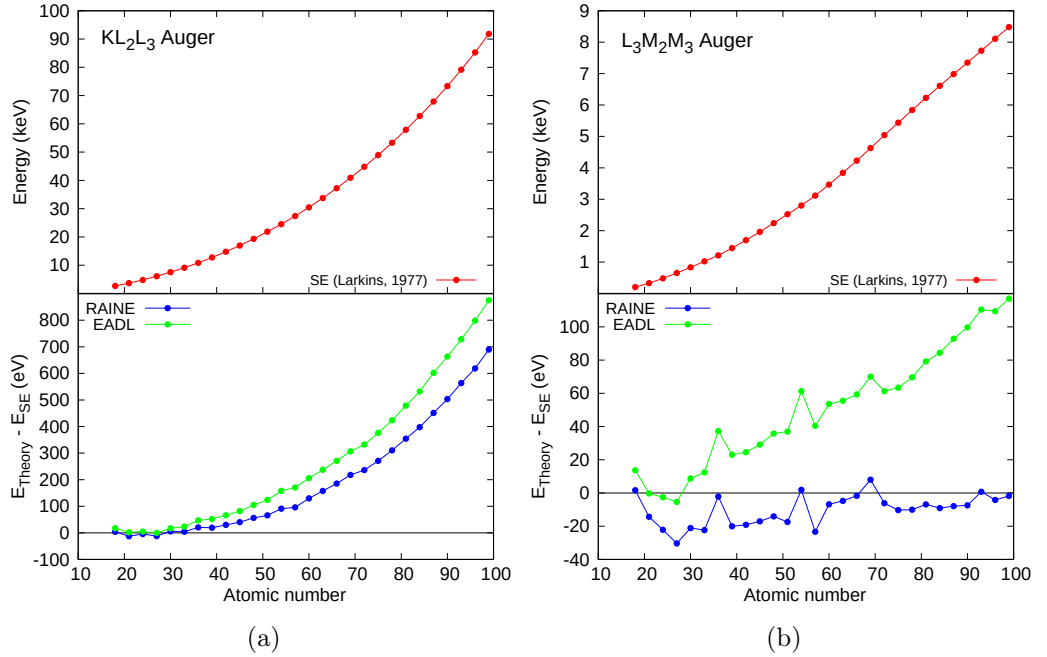


Figure 4.1: Comparison of the semi-empirical (SE, red) (a) KL_2L_3 and (b) $L_3M_2M_3$ Auger energies from Larkins tabulation with the theoretical energies from the **RAINE** code (blue) and EADL (green) for elements from $Z = 18$ to $Z = 99$. The top panels show the semi-empirical values and the bottom panels show the differences between the theories and the semi-empirical method.

Figure 4.1a compares the SE KL_2L_3 Auger energies to the theoretical values from the **RAINE** code and EADL. The bottom panel of the figure shows that both the **RAINE** code and EADL overestimate the KL_2L_3 Auger energies compared to the SE method. The overestimation is increasing with the atomic number and it reaches 700 and 900 eV ($< 1\%$ of the transition energies) at $Z = 99$ for the **RAINE** code and EADL, respectively. Figure 4.1b shows a similar comparison for the $L_3M_2M_3$ Auger transition. In this case, the **RAINE**

code constantly underestimates the Auger energies while EADL overestimates compared to the SE method. The difference between EADL and SE is increasing with the atomic number and it reaches 120 eV for einsteinium ($Z = 99$). The **RAINE** code agrees well with SE for $Z = 18, 36$ and 54 , which are noble gases, when SE values are also referenced to the vacuum level. Interestingly, there are spikes for noble gases as well as elements $Z = 69$ (Tm), 81 (Tl) and 93 (Np), in the differences between the SE and theoretical energies for the $L_3M_2M_3$ Auger transition. These spikes can be attributed to the effect of atomic closed-shell structure. All noble gases have a closed-shell structure by definition. Thulium (Tm) is one N_7 electron short of completing the closed shell while thallium (Tl) and neptunium (Np) only have one unpaired electron in the P_2 - and P_4 -subshells, respectively.

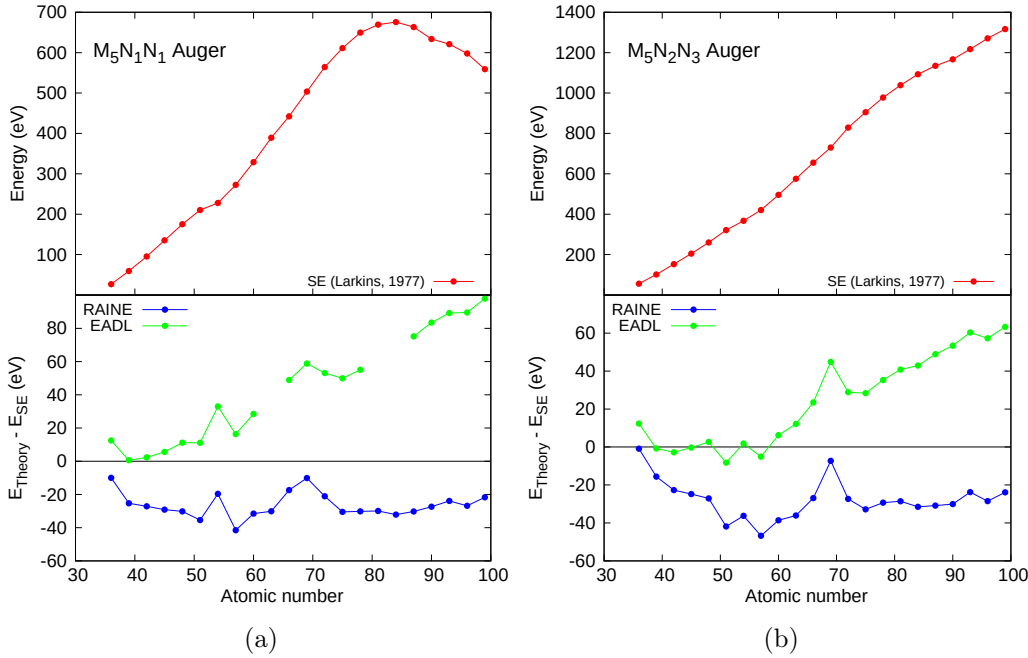


Figure 4.2: Similar to Figure 4.1 but for (a) $M_5N_1N_1$ and (b) $M_5N_2N_3$ Auger transitions of elements from $Z = 36$ to $Z = 99$.

Figure 4.2 compares the SE $M_5N_1N_1$ and $M_5N_2N_3$ Auger energies to the theoretical values from the **RAINE** code and EADL. The bottom panels show a similar behaviour to the one shown in the bottom panel of Figure 4.1b for $L_3M_2M_3$. A similar comparison for $N_5O_1O_1$ and $N_5O_2O_3$ Auger energies is shown in Figure 4.3. The top panel of Figure 4.3a shows a sharp fall in transition energy at $Z = 66$ (Dy). The fall can be attributed to a typo in the Larkins' tabulation. A value of 20.9 eV was reported while the $N_5O_1O_1$ transition energies for the neighbouring elements are in the order of 60 eV. It seems from these figures that EADL is in better agreement with the Larkins' tabulation for M- and N-shell Auger electrons. However, it is important to note that the SE Auger energies from Larkins are referenced to the Fermi level rather the vacuum level for most elements and atoms embedded in solids can have different screening of vacancies.

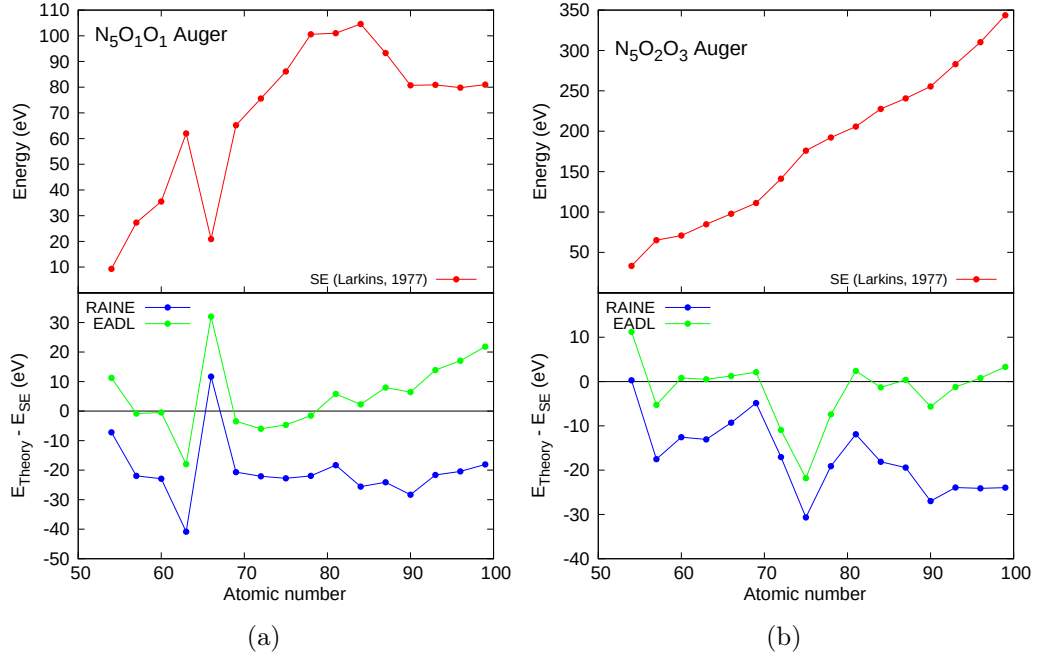


Figure 4.3: Similar to Figure 4.1 but for (a) $N_5O_1O_1$ and (b) $N_5O_2O_3$ Auger transitions of elements from $Z = 54$ to $Z = 99$.

Vapour-metal shifts

Since the semi-empirical (SE) Auger energies reported in the Larkins tabulation [28] were calculated for solid systems, a correction must be applied to modify the energies so they are referenced to the vacuum level. Larkins proposed the following equation to correct the tabulated Auger energies for a phase change:

$$E^{\text{vapour}}(WXY) = E^{\text{solid}}(WXY) + (\Delta\epsilon_W^Z - \Delta\epsilon_X^Z - \Delta\epsilon_Y^Z) - \xi(Z), \quad (4.2)$$

where $E^{\text{vapour}}(WXY)$ and $E^{\text{solid}}(WXY)$ are the Auger transition energies in the vapour and solid phases, and $\Delta\epsilon_W^Z$ is the difference between the vapour- and solid-phase binding energies of the W shell for element Z

$$\Delta\epsilon_W^Z = \epsilon_W^Z(\text{vapour}) - \epsilon_W^Z(\text{solid}), \quad (4.3)$$

which is always positive and $\xi(Z)$ is a solid-state correction term that is a characteristic of an element and independent of the exact nature of the Auger process. Figure 4.4 shows the reported values of $\xi(Z)$ for elements $10 \leq Z \leq 100$ in the Larkins tabulation which were considered to be accurate to within 1-2 eV [28]. Since the $\Delta\epsilon^Z$ term is roughly constant over different subshells [164, 165, 166] and the $\xi(Z)$ term is always positive, the difference between the vapour- and solid-phase Auger energies, i.e. $E^{\text{vapour}}(WXY) - E^{\text{solid}}(WXY)$, should be always negative, and therefore the Larkins' SE energies will be reduced after correcting for a phase change from solid to vapour. The vapour-metal shift has been

experimentally measured to be around 15 eV for zinc, cadmium and ytterbium [167, 168]. Figure 4.5 shows the differences between the theoretical energies from the **RAINE** code and EADL and the SE values from Larkins after they are adjusted for a hypothetical vapour-metal shift of 15 eV. The theoretical energies from the **RAINE** code are now in better agreement with the SE values while the agreement between EADL and Larkins worsens.

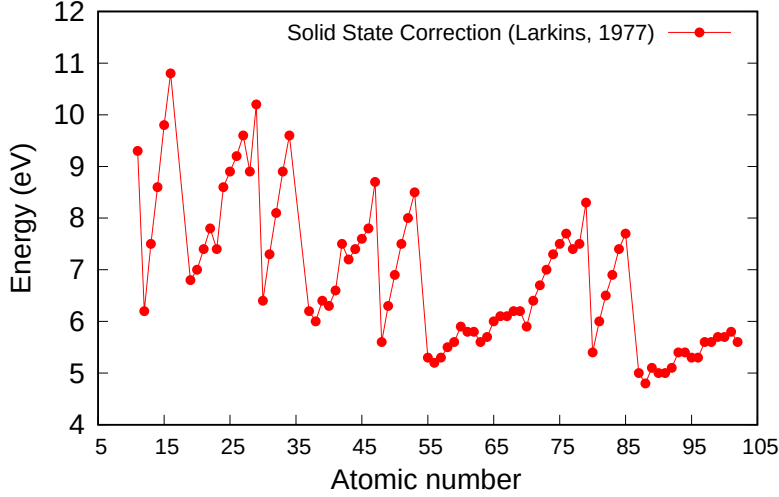


Figure 4.4: Values of the solid state correction term $\xi(Z)$ for elements $10 \leq Z \leq 100$ (except for Ne, Cl, Ar, Br, Kr, Xe and Rn) from the Larkins tabulation [28]. The $\xi(Z)$ term increases from the Group 1 elements to the noble gases and generally decreases with the increase in the principal quantum number of the valence shell.

Auger transitions in xenon

Semi-empirical (SE) energies for Auger transitions in xenon tabulated by Larkins [28] were calculated for the vapour phase and referenced to the vacuum level, which is also the reference level for theoretical energies from the **RAINE** code and EADL. So a comparison of Auger-electron energies in xenon will eliminate the ambiguity about the energy reference level. Figure 4.6a compares the energies of K-shell Auger transitions in xenon from the three sources. The **RAINE** code overestimates the K-shell Auger energies compared to the SE method by about 85-125 eV. The overestimation increases slightly when a transition involves the M-shell. On the other hand, the differences between EADL and the SE method are more inconsistent. KLL Auger energies from EADL are higher than Larkins' values by more than 150 eV but the estimation gradually decreases towards 40 eV for KMM Auger transitions. Figure 4.6b shows that the overestimation in xenon's LMM Auger energies by the **RAINE** code is fairly constant at around 15 eV. The difference between **RAINE** and SE slowly increases when higher subshells of the M-shell are involved in a transition which is consistent with the observation for the K-shell Auger energies. This is due to the smaller overestimation in the $M_{4,5}$ binding energies than the $M_{1,2,3}$ subshells by **RAINE**, so the overestimation of the L-shell binding energy worsens the agreement between

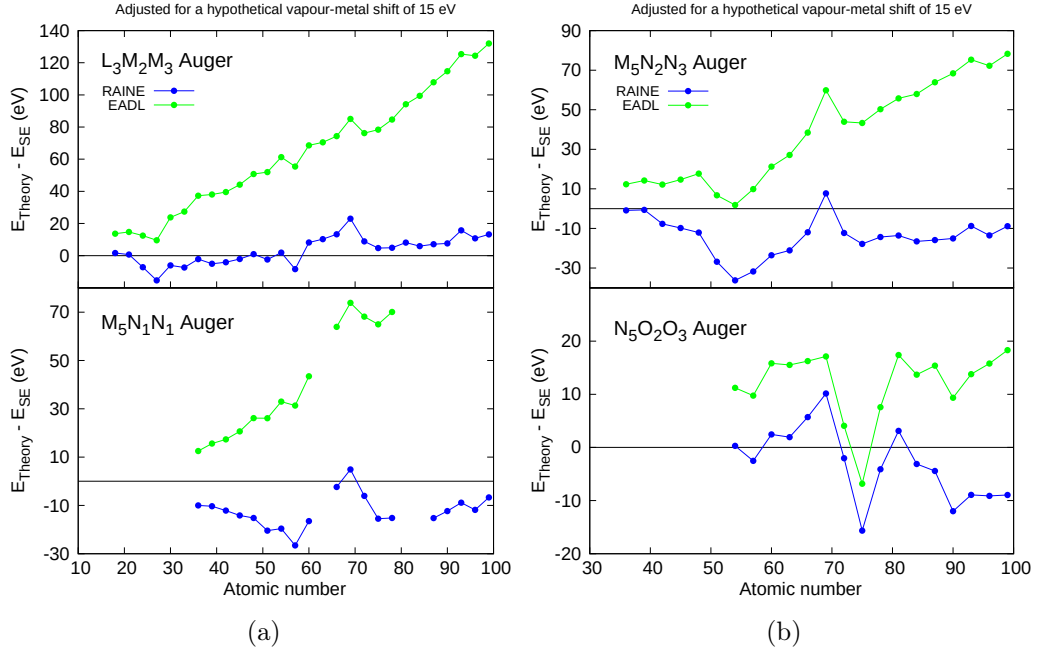


Figure 4.5: Differences between the theoretical energies from the **RAINE** code and EADL and the semi-empirical (SE) energies from the Larkins tabulation for (a, top) $L_3M_2M_3$, (a, bottom) $M_5N_1N_1$, (b, top) $M_5N_2N_3$, and (b, bottom) $N_5O_1O_1$ Auger transitions, after they are adjusted for a hypothetical vapour-metal shift of 15 eV.

RAINE and SE (see Figure 3.3). In contrast, the difference between EADL and SE reduces when a transition involves higher subshells of the M-shell. However, EADL is in worse agreement with SE across all LMM Auger transitions. Interestingly, the overestimation by EADL increases markedly for $L_{2,3}MM$ Auger transitions. This strongly indicates that the overestimation of the $L_{2,3}$ -subshell binding energies by EADL could be much worse than the L_1 -subshell.

Figure 4.7a shows that energies of MNN Auger transitions in xenon calculated by the **RAINE** code are generally lower than the SE values. Similar to K- and L-shell Auger transitions, the differences between **RAINE** and SE for MNN transitions are exhibiting an upward trend when higher subshells are involved albeit towards a better agreement in this case. The discrepancies between **RAINE** and SE are almost constant from M_1NN to M_5NN transitions while the difference between EADL and SE is slowly increasing. Figure 4.7b indicates that energies of N-shell Auger and Coster-Kronig (CK) transitions from **RAINE** and EADL are very similar apart from the $N_{4,5}$ Auger electrons, where **RAINE** agrees well with SE.

Overall, the **RAINE** code is in better agreement with the Larkins' semi-empirical (SE) values than EADL apart from the K-shell transitions. Moreover, discrepancies between **RAINE** and SE are more consistent across all Auger transitions emitted from a particular principal shell.

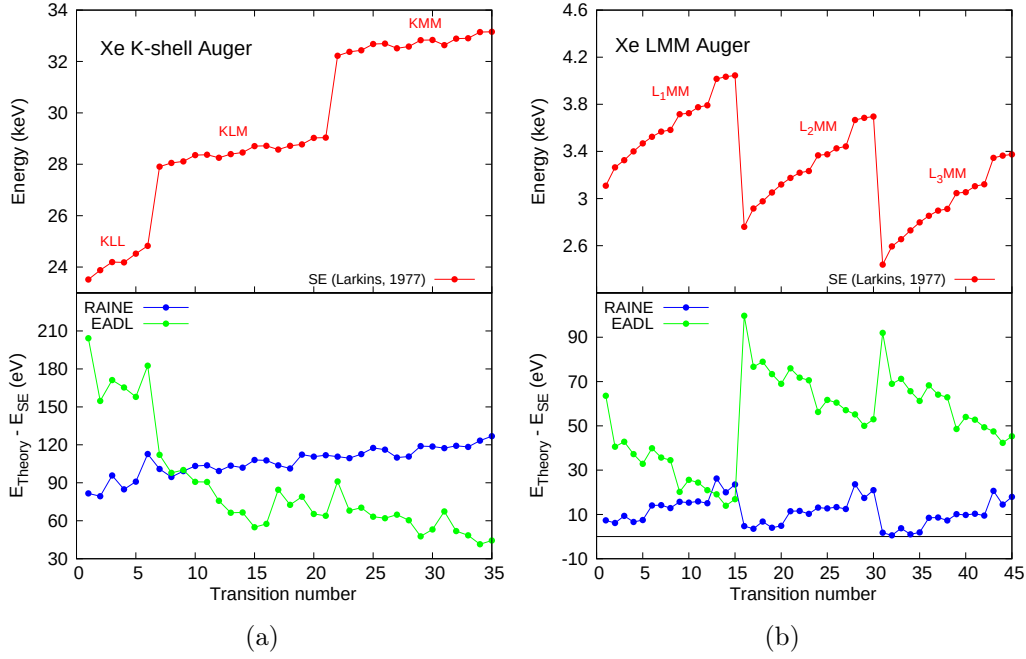


Figure 4.6: Comparison of the semi-empirical (SE, red) (a) K-shell and (b) LMM Auger energies from the Larkins tabulation with the theoretical energies from the RAINE code (blue) and EADL (green) for xenon. The top panels show the semi-empirical values and the bottom panels show the differences between the theories and the semi-empirical method.

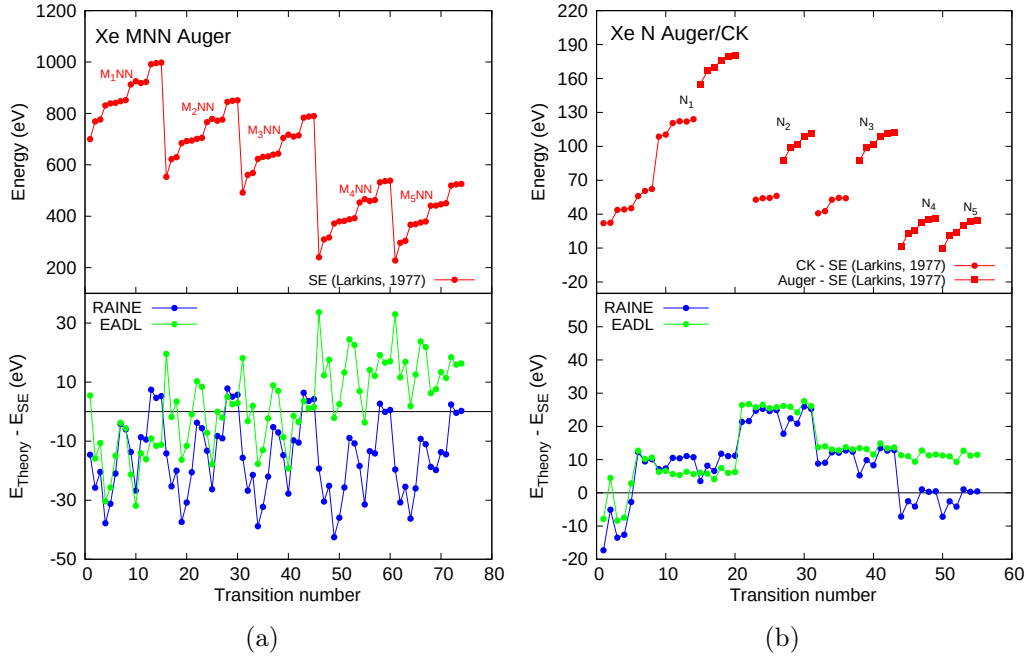


Figure 4.7: Similar to Figure 4.6 but for (a) MNN Auger and (b) N-shell Auger and Coster-Kronig (CK) transitions.

4.2.2 Comparison with experiments

In this section, the Auger-energy spectra calculated using BrIccEmis are compared to the experimental K- and L-shell Auger-energy spectra of ^{131}Cs reported in Ref. [144] as well as

the low-energy electron spectrum (< 4000 eV) of ^{125}I recently measured at the Australian National University (ANU).

^{131}Cs electron capture decay

^{131}Cs ($T_{1/2} = 9.689$ days) decays to ^{131}Xe by electron capture (EC) and its decay scheme is shown in Figure 4.8. In this EC decay, the nuclear ground state of ^{131}Cs decays directly to the nuclear ground state of ^{131}Xe , therefore this is a pure EC decay (or pure Auger emitter in nuclear medicine) and there is no subsequent internal nuclear transition. Auger electrons are only emitted from the atomic relaxation in ^{131}Xe from the vacancy creation by EC. Kovalík et al. [144] measured the K- and L-shell Auger-energy spectra of the ^{131}Cs decay in 1998. In this experiment, a ^{131}Cs source was prepared by evaporation onto a thin aluminium foil.

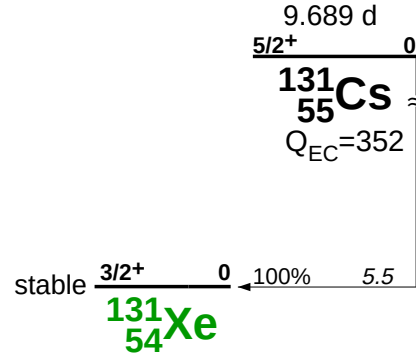


Figure 4.8: Scheme of the electron capture decay of ^{131}Cs to ^{131}Xe . Image extracted from [99]

Figure 4.9 compares the measured KLL Auger spectrum with the theoretical one calculated using **BrIccEmis**, which assumes the electron configuration of Cs, i.e. the so-called “atomic structure effect” (see Section 3.4.2). **BrIccEmis** adopts the jj coupling scheme, therefore it only produces one energy line for KL_1L_2 , KL_1L_3 and KL_3L_3 while the experiment shows that the energy separation between two intermediate coupling states can be up to 51 eV. Moreover, **BrIccEmis** also overestimates the transition energies by about 95 eV, which is consistent with the comparison of transition energies from the **RAINE** code and Larkins (see the bottom panel of Figure 4.6a). Figure 4.10 demonstrates that the theoretical spectrum from **BrIccEmis** agrees well with the experiment after an energy shift of -95 eV is applied. Overall, the relative intensities of individual Auger lines agree well with the experimental intensities.

Figure 4.9 also compares the experimental Auger energies with the semi-empirical (SE) ones from the Larkins tabulation [28]. It seems that there is an energy shift of about 12 eV towards lower energies for the SE Auger lines. For example, the reported experimental energy of $\text{KL}_2\text{L}_3(^1\text{D}_2)$ Auger line is 24521.6 ± 2.0 eV while the SE value of the same

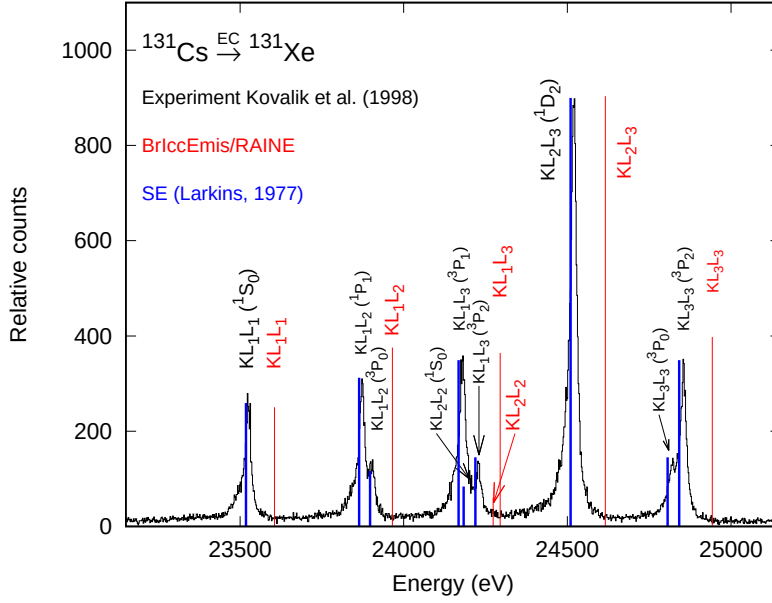


Figure 4.9: Comparison of the experimental KLL Auger-energy spectrum from the decay of ^{131}Cs [144] with the theoretical one calculated using BrIccEmis. The theoretical spectrum is scaled to match the intensity of the experimental $\text{KL}_2\text{L}_3(^1\text{D}_2)$ Auger line. Semi-empirical (SE) Auger energies from the Larkins' tabulation [28] are also plotted for comparison. Since only the transition energies were reported by Larkins, SE Auger lines are scaled to match the intensity of their corresponding experimental counterparts.

transition is 24510.1 eV, i.e. an energy shift of 11.5 eV. This energy shift should be predominantly due to the vapour-metal shift as the experimental energy is referenced to the Fermi level while the SE Auger energy was calculated for the vapour phase of xenon. In addition, this energy shift could also be due to the atomic structure effect, which says that the KLL Auger transitions occur in the electron configuration of Cs rather than Xe. This effect could increase the theoretical KL_2L_3 transition energy by 7.1 eV for the vapour phase based on the Dirac-Fock calculation by the RAINE code. However, it is difficult to establish the magnitude of this effect for the metal phase. A recent measurement of the KLL Auger-energy spectrum from the EC decay of ^{83}Rb to ^{83}Kr showed that this effect could only increase the KLL transition energies by approximately 1-3 eV in the solid systems when the Dirac-Fock calculation indicates that the change in energies should be close to 6.5 eV for the gaseous Kr [166].

Figure 4.11 compares the experimental L-Auger spectrum in the decay of ^{131}Cs to the theoretical one simulated using BrIccEmis¹. All energy lines in the theoretical spectrum have been folded with an approximate lineshape to account for the broadening of Auger lines in the measurement. This lineshape is consisted of a Voigt profile and a constant step function that extends from the centroid of the Voigt profile to 0 eV. The Voigt profile is

¹L-Auger spectrum simulated using BrIccEmis is largely independent of the choice of the propagating vacancy described in Section 3.4.4 since L-Auger electrons are emitted in the first few steps where only a handful of spectator vacancies may exist.

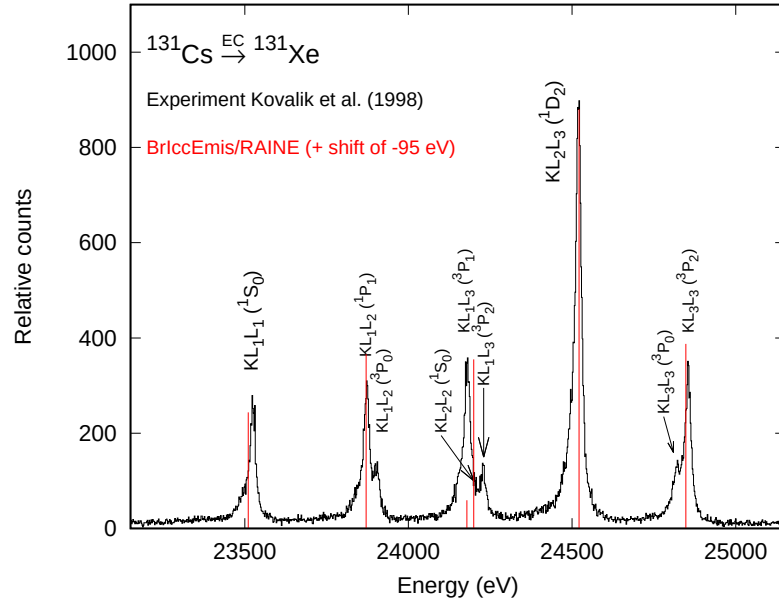


Figure 4.10: Same as Figure 4.9 but an energy shift of -95 eV is applied to the theoretical spectrum from BrIccEmis and Larkins' semi-empirical energy lines are excluded.

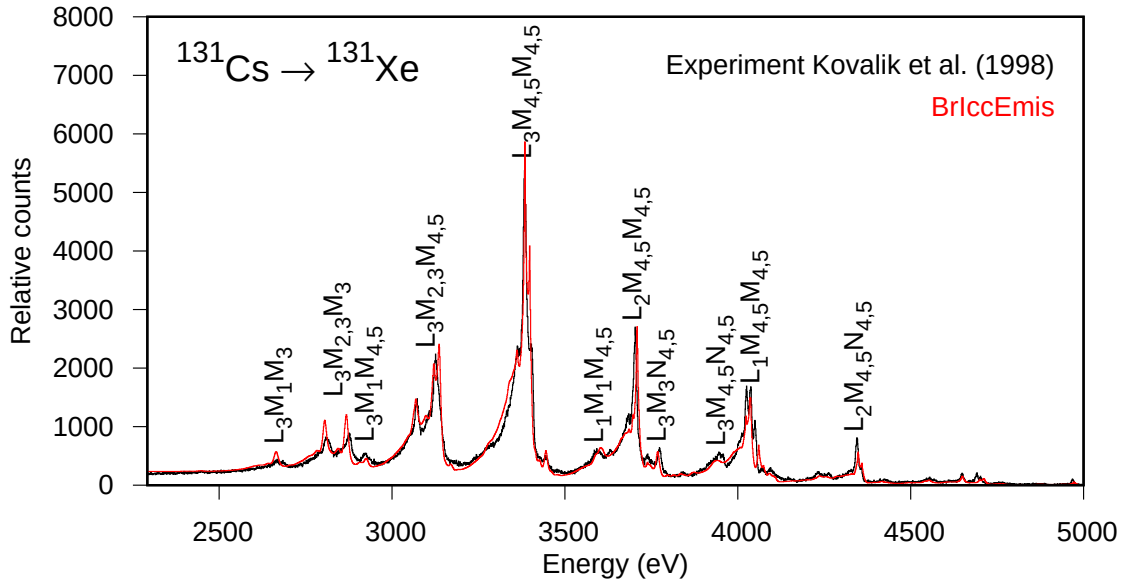


Figure 4.11: Comparison of the experimental L-Auger spectrum from the decay of ^{131}Cs [144] with the theoretical one calculated from BrIccEmis. The theoretical spectrum has been folded with an approximate lineshape (see main text for details), and scaled to match the intensity of the $\text{L}_3\text{M}_{4,5}\text{M}_{4,5}$ Auger group. A similar figure has been published in Ref. [157].

constructed from the convolution of a Gaussian profile, which imitates the spectrometer response function, and a Lorentzian profile resulting from the natural broadening of a transition. For example, the Voigt profile for an Auger electron with kinetic energy of 3374.3 eV is shown in Figure 4.12. Moreover, as an approximation, the Gaussian profile is held fixed over the energy range shown in Figure 4.11. Ideally, the full convolution

of the Gaussian and Lorentzian profiles should be performed to obtain the exact Voigt profile but such a procedure is very time-consuming. The pseudo-Voigt profile, which is a linear combination of the Gaussian and Lorentzian profiles, is used instead for folding the theoretical Auger lines. The η parameter shown in Figure 4.12 is the coefficient for the Gaussian component in the linear combination while $1-\eta$ is the coefficient for the Lorentzian part, and it is determined from the widths of the two individual components using the following relations [169, 170]:

$$\eta = 1.36603 (\Gamma_L/\Gamma) - 0.47719 (\Gamma_L/\Gamma)^2 + 0.11116 (\Gamma_L/\Gamma)^3, \quad (4.4)$$

$$\begin{aligned} \Gamma &= (\Gamma_G^5 + 2.2692691 \Gamma_G^4 \Gamma_L + 2.42843 \Gamma_G^3 \Gamma_L^2 + 4.471631 \Gamma_G^2 \Gamma_L^3 + \\ &= 0.07842 \Gamma_G \Gamma_L^4 + \Gamma_L^5)^{1/5}, \end{aligned} \quad (4.5)$$

where Γ_L and Γ_G are the widths of the Lorentzian and Gaussian components, respectively. The Lorentzian width, Γ_L used in fitting an Auger transition XYZ is approximated using the following relation:

$$\Gamma_L = \Gamma_X + \Gamma_Y + \Gamma_Z, \quad (4.6)$$

where Γ_X is the atomic level width associated with a single vacancy in the X shell. The Γ_X , Γ_Y and Γ_Z used are based on the values provided in the EADL database [27]. The constant step function in the approximate lineshape is to partially account for the interactions of emitted Auger electrons in the source material.

The comparison shown in Figure 4.11 indicates that the folded theoretical spectrum agrees very well with the experiment. The effect of shake-off process has also been considered but it makes no noticeable changes to the folded theoretical spectrum. Overall, it seems from the comparison that a very good agreement between the theory and the experiment has been achieved. However, it is also important to note that the theoretical spectrum is referenced to the vacuum level while its experimental counterpart is referenced to the Fermi level. The comparison of the experimental KLL Auger energy lines to the semi-empirical (SE) calculation of Larkins [28] (see Figure 4.9) seems to suggest that the vapour-metal shift for xenon, i.e. xenon in vacuum versus xenon in a matrix, could be on the order of 12 eV if one neglects the atomic structure effect. Therefore, in the case of the L-Auger energy spectrum, the theoretical spectrum lines up almost perfectly with the experimental one could be due to the slight overestimation in the energies of the LMM Auger group, which is the major component of the L-Auger spectrum, by **BrIccEmis** given that the **RAINE** code overestimates the the LMM Auger energies in xenon by approximately 15 eV compared to the SE energies from Larkins (see the bottom panel of Figure 4.6b). Nevertheless, it is highly encouraging that **BrIccEmis** reproduces the overall structure of the Auger-energy spectrum within 1800 to 5000 eV.

Kovalík et al. [144] reported that the absolute energy of the $L_3M_4M_5(^1G_4)$ transition

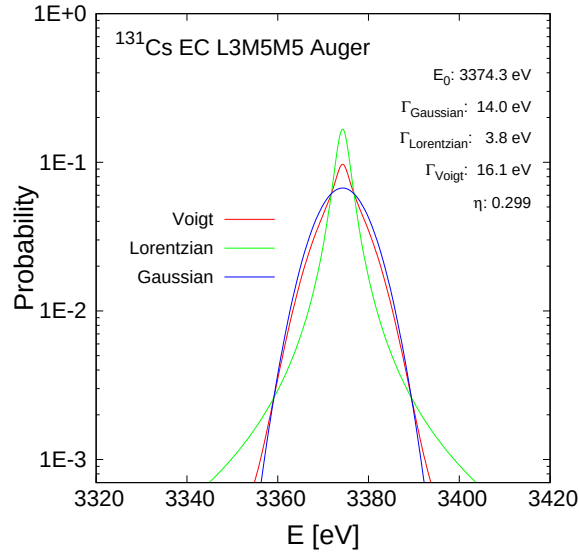


Figure 4.12: Voigt profile of the $L_3M_5M_5$ Auger transition with kinetic energy, $E_0 = 3374.3$ eV. The Gaussian width, Γ_{Gaussian} , is associated with the instrumental resolution and the Lorentzian width, $\Gamma_{\text{Lorentzian}}$, is related to the natural broadening of the transition. See main text for details.

in xenon measured from the β^- decay of ^{131}I ($T_{1/2} = 8.0252$ days) to ^{131}Xe is 9.4 ± 1.1 eV lower than its counterpart when a ^{131}Cs source was used instead. ^{131}I does not decay to the nuclear ground state of ^{131}Xe , so only the conversion process of the γ -transitions from the decays of the nuclear excited states in ^{131}Xe can create atomic vacancies in xenon. Therefore, they suggested that this energy shift could be predominantly caused by the atomic structure effect given that both energies were referenced to the Fermi level and the chemical shifts in electron binding energies for caesium and iodine were found to be about 1 and 3.4 eV, respectively. However, the individual Auger lines in the L-Auger energy spectrum are difficult to resolve since many transitions have overlapping energies, therefore, the reported energy shift of 9.4 eV could be unreliable. Kovalík et al. [144] did not report the absolute energy of the $KL_2L_3(^1D_2)$ transition from the ^{131}I decay. It is important to note that it should be easier to resolve individual lines in the KLL Auger-energy spectrum than in the L-Auger spectrum.

^{125}I electron capture decay

^{125}I ($T_{1/2} = 59.408$ days) decays exclusively to the first excited state of ^{125}Te which then decays predominantly by the internal conversion to the nuclear ground state ($E_\gamma = 35.5$ keV). The decay scheme of ^{125}I is shown in Figure 2.6. Recently, a collaboration between the Australian National University (ANU) and the Australian Nuclear Science and Technology Organisation (ANSTO) was formed to investigate the low-energy Auger electrons emitted from the decay of ^{125}I . In this study, a monolayer of ^{125}I was deposited on a gold substrate

with the source-preparation method inspired by Ref. [171]. This way of preparing a ^{125}I source also enables the measurement of very low-energy electrons (< 1000 eV). Figure 4.13 compares the measured spectrum (preliminary) for electrons below 4000 eV, which also includes the K-shell conversion ($E_e = 3678$ eV) from the decay of the first excited state of ^{125}Te , with the theoretical spectrum calculated from **BrIccEmis**. The theoretical spectrum has been folded with an approximate lineshape similar to the one described in Section 4.2.2². Shake-off processes are also included in the **BrIccEmis** simulation and they are especially important to explain the “shoulder” on the low-energy side of the K-conversion line³.

It seems from Figure 4.13 that the theoretical intensities of L-Auger electrons are not matching well with the experiment. However, it is important to note that the theoretical spectrum is scaled to match the intensity of the K-conversion line, which is labelled as “35K CE” in Figure 4.13, therefore the theoretical intensities of L-Auger electrons can be highly sensitive to the adopted width of the K-conversion line. It is clear from the figure that the experimental width of the K-conversion line is larger than the theoretical width. The adopted theoretical width for the Voigt profile is the combination of the Lorentzian width of 9.8 eV (atomic level width of K-shell⁴) and the Gaussian width of 25 eV. The peak height of the K-conversion line will be reduced when its width is broadened to match the experimental width, and therefore, the theoretical intensities of L-Auger electrons will be in better agreement with the experiment. In terms of the overall structure of the spectrum, a good agreement between the theoretical and experimental spectra is achieved.

Interestingly, the theoretical Auger spectrum that assumes the fast neutralisation, i.e. the condensed-phased approximation (see Section 3.4.7 for details), seems to be in better agreement with the experiment for $M_{4,5}\text{NN}$ Auger electrons at energies below 600 eV. This suggests that the electron transfer from the gold substrate to Te atoms, which are undergoing atomic relaxations following the decays of ^{125}Te , could be in the order of 1-10 fs when most M-shell Auger electrons are emitted (see Figure 3.8). The spectrum calculated using the isolated-atom approximation has an energy shift towards lower energies due to the build up of charges resulting from multiple non-radiative transitions. In this case, the decrease in Auger energies within the isolated-atom approximation results in worse agreement with the experiment.

If one focuses on the very low-energy regime (< 800 eV), it is clear that the theory could not account for the huge intensities of low-energy electrons observed in the exper-

²The theoretical spectrum is simulated using **BrIccEmis** based on the “random-vacancy” approximation described in Section 3.4.4. The choice is made based on the study of residual charge-state distributions discussed in Chapter 5.

³The shake-off probabilities following the creation of vacancies in Te are calculated based on the shake theory from Krause and Carlson [14] (see Section 2.3.1), and the Dirac-Fock wave functions calculated using the **Grasp2K** package. The results of the calculations can be found in Table A.4 in Appendix A.4.

⁴The atomic level width is adopted from EADL [27], which is consistent with the recent evaluation [135].

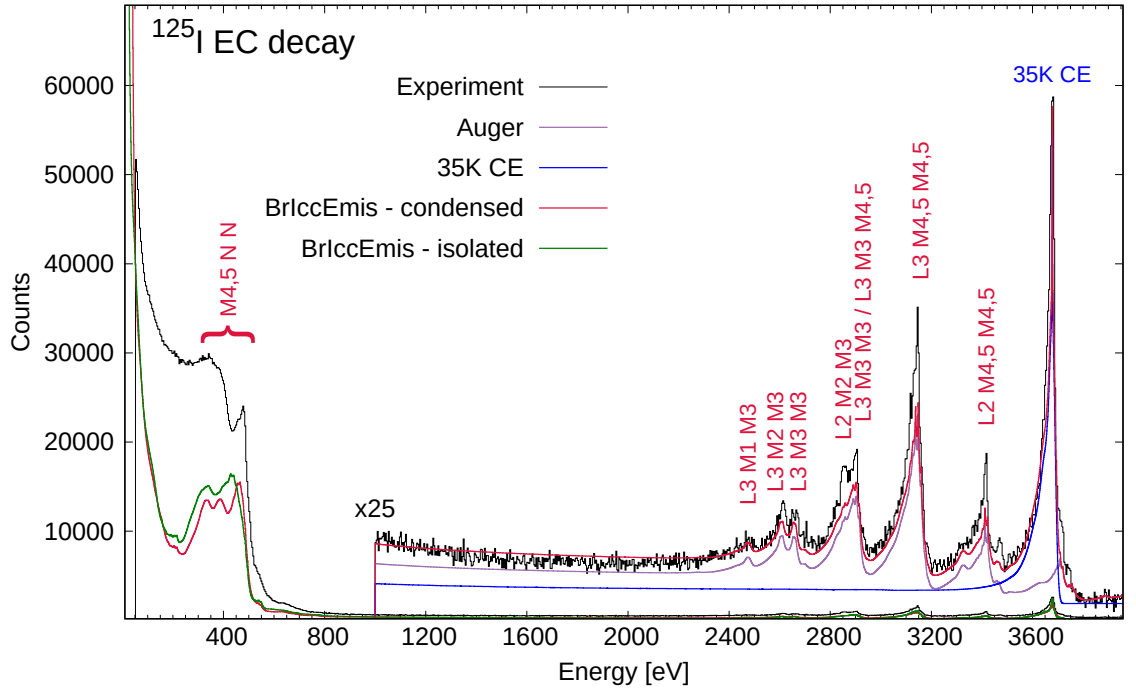


Figure 4.13: Comparison of the (preliminary) experimental low-energy Auger spectrum (50-4000 eV) from the decay of ^{125}I recently measured at ANU with the theoretical one calculated from BrIccEmis. The theoretical spectrum has been folded with an approximate lineshape (see Section 4.2.2 for details) and scaled to match the intensity of the K-shell conversion electrons (labelled as “35K CE”). Two theoretical spectra are shown for < 1000 eV; one assumes the fast neutralisation (condensed) and it is plotted in red and another one, which is shown in green, assumes the slow neutralisation (isolated). From 1000 eV onwards, a separate plot is shown with the intensities scaled up by 25 times and the K-conversion component is shown in blue while the Auger part is displayed in purple. A $1/E$ factor has also been applied to all theoretical spectra to account for the energy dependence of the spectrometer efficiency. The experimental spectrum has been artificially shifted towards higher energies by 6 eV so that the energy of the theoretical K-conversion line matches its experimental counterpart.

iment. This could indicate that extra sources of low-energy electrons are present. These extra electrons are probably coming from the gold substrate following inelastic scattering of the primary radiations emitted from the ^{125}I decays. Moreover, Pronschinke et al. [171] claimed that they observed a 600% enhancement of extremely low-energy electrons (0-10 eV) from a similar gold substrate. However, they neglected the contribution of shake-off electrons when they deduced the increase in the number of electrons within the first 10 eV, i.e. they only considered the contribution of Auger and Coster-Kronig electrons. Nevertheless, the differences between the theory and the experiment at low energies could also be due to the incorrect spectrometer efficiency adopted to scale the theoretical spectrum. The theoretical spectra shown in Figure 4.13 have been scaled by a $1/E$ factor to account for the spectrometer efficiency. Further experiments will be performed to quantify the spectrometer efficiency. Once all spectrometer-related issues are resolved in the near fu-

ture, one can quantify the enhancement of low-energy electrons in absolute terms since the K-shell conversion line can act as the intensity-calibration line for the absolute intensity.

4.3 X-rays

In this section, the X-ray energies calculated by the **RAINE** code are compared to the values from the EADL library [27]. Both of these sources of theoretical transition energies will be compared to the recent theoretical calculations by Deslattes et. al. [172] for several dominant K- and L-shell X-rays and the evaluated M_3N_5 and M_5N_3 X-ray energies from Bearden [173]. The theoretical calculations by Deslattes et. al. [172], which were based on the Dirac-Fock method in the implementation of Desclaux code [111], included the QED and Breit corrections to provide the most accurate theoretical X-ray energies to date. Deslattes et. al. reported that their theoretical K-shell X-ray transition energies deviate from experimental data by < 5 eV for $Z \leq 55$ and < 20 eV for $Z \geq 55$; and for the L-shell X-rays, their values differ from experiments by < 6 eV for $20 \leq Z \leq 100$. On the other hand, the X-ray energies from EADL were derived from theoretical neutral binding energies:

$$E_{XY} = \text{B.E.}(X) - \text{B.E.}(Y), \quad (4.7)$$

where E_{XY} is the energy of radiative transition XY and $\text{B.E.}(X)$ and $\text{B.E.}(Y)$ are the binding energies of X and Y subshells, respectively.

4.3.1 Comparisons with tabulations

Figure 4.14 compares the KL_3 and KM_3 X-ray transition energies from the **RAINE** code, EADL and Deslattes, which will be named as “DF-QED” from here onwards. It is clear from the figure that both **RAINE** and EADL overestimate the K-shell X-ray energies significantly at high- Z where the QED and Breit corrections are required to obtain the accurate electron binding energies [141]. This is also the region where a K-shell atomic vacancy is almost exclusively transported to higher subshells by X-ray transitions [33]. Therefore, the standard Dirac-Fock method implemented in the **RAINE** code will overestimate the total energy released via the K-shell X-rays by about 0.8% at $Z = 100$ due to the lack of QED and Breit corrections. In general, transition energies from **RAINE** are in better agreement with DF-QED than EADL only for $Z \leq 40$. A similar trend is seen for L-shell X-rays. Figure 4.15 shows that EADL is in better agreement with DF-QED for all Z except for L_2M_4 X-rays at $Z \geq 96$.

Figure 4.16 compares Bearden’s evaluated X-ray transition energies [173] with theoretical energies from **RAINE** and EADL. The figure shows that **RAINE** is in slightly better agreement with the evaluation than EADL for M_3N_5 X-rays. **RAINE** deviates from the evaluation for M_5N_3 X-rays at $Z \leq 60$ but the agreement between the two improves at

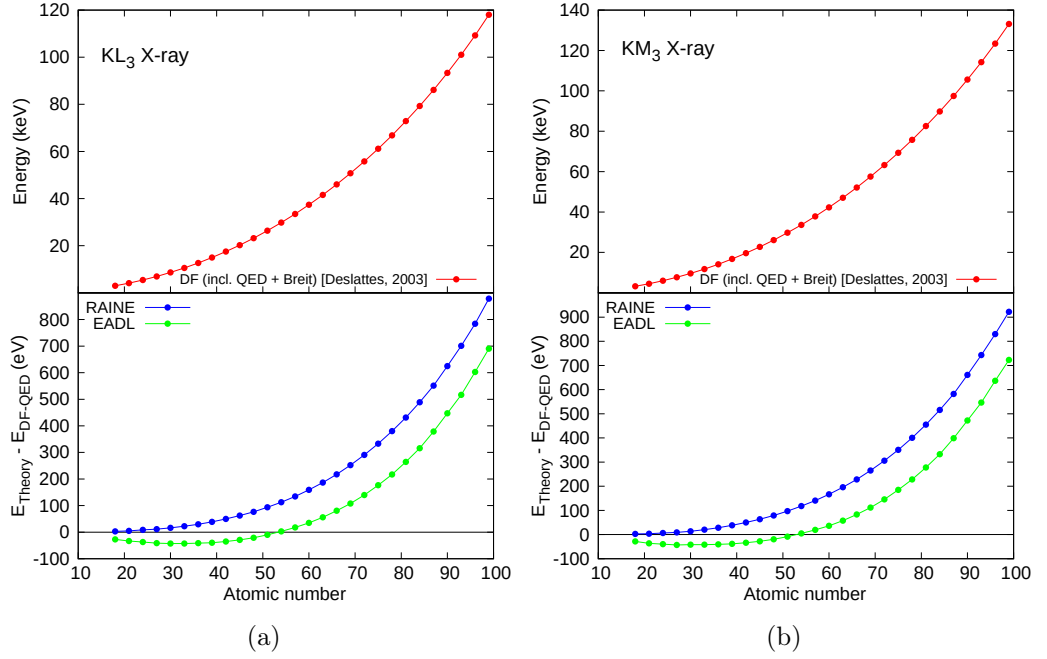


Figure 4.14: Comparison of theoretical X-ray transition energies from Deslattes (red) [172] with those from RAINE (blue) and EADL (green) for (a) KL_3 and (b) KM_3 X-rays. The top panels show the values from Deslattes and the bottom panels show the differences between all three sources.

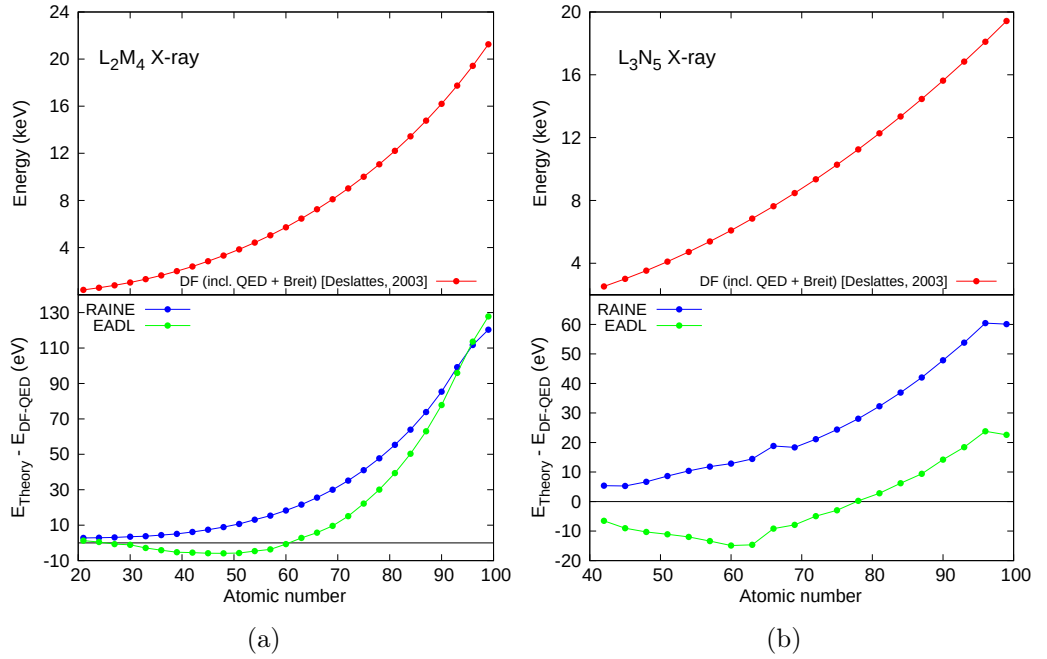


Figure 4.15: Similar to Figure 4.14 but for (a) L_2M_4 and (b) L_3N_5 X-rays.

higher atomic numbers. EADL is in good agreement with the evaluation for the low- Z region but the agreement worsens at higher atomic numbers.

The comparisons shown in Figure 4.17 resonate well with those presented in Figure 4.14 and 4.15, except for L_1 X-rays. The deviations between EADL and DF-QED are within

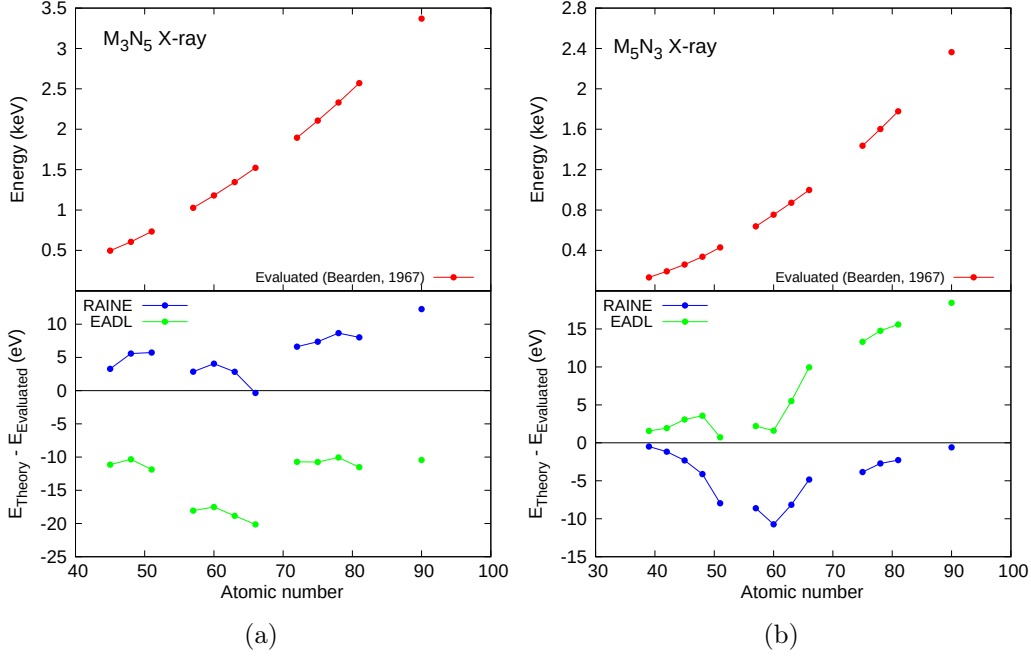


Figure 4.16: Comparison of evaluated X-ray transition energies from Bearden (red) [173] with those from RAINE (blue) and EADL (green) for (a) M_3N_5 and (b) M_5N_3 X-rays. The top panels show the evaluated transition energies and the bottom panels show the differences between all three sources.

20 eV for all K-shell X-rays of xenon while RAINE overestimates the transition energies by at least 100 eV. The differences between EADL and DF-QED for energies of L_1 -subshell X-rays are not consistent with those for L_2 - and L_3 -subshell X-rays. In contrast, the overestimation by RAINE is more constant although it gradually decreases from L_1 to L_3 X-rays.

Overall, EADL provides a better set of X-ray energies for K- and L-shell transitions, but both RAINE and EADL have similar accuracies for M-shell X-rays. This means that the radiative transition energies derived from neutral binding energies and Eq. (4.7) are better than the RAINE code, which implements a Dirac-Fock method but does not account for QED and Breit effects, for the singly-ionised systems.

4.4 2D Auger-energy correlation spectrum

Recently, Suzuki et al. [71] reported a two-dimensional energy correlation spectrum between so-called “fast” ($280 \leq E_e \leq 600$ eV) and “slow” ($0 \leq E_e \leq 160$ eV) Auger electrons that are in coincidence with the M_5 photoelectrons following photoionisation of xenon using 924 eV synchrotron radiation in a vacuum chamber. This measurement revealed the energy spectrum due to the short vacancy cascade following the creation of an initial vacancy in the M_5 -subshell. The reported experimental correlation spectrum is shown in the bottom panel of Figure 4.18a and it is very important for the benchmarking of a vacancy-cascade

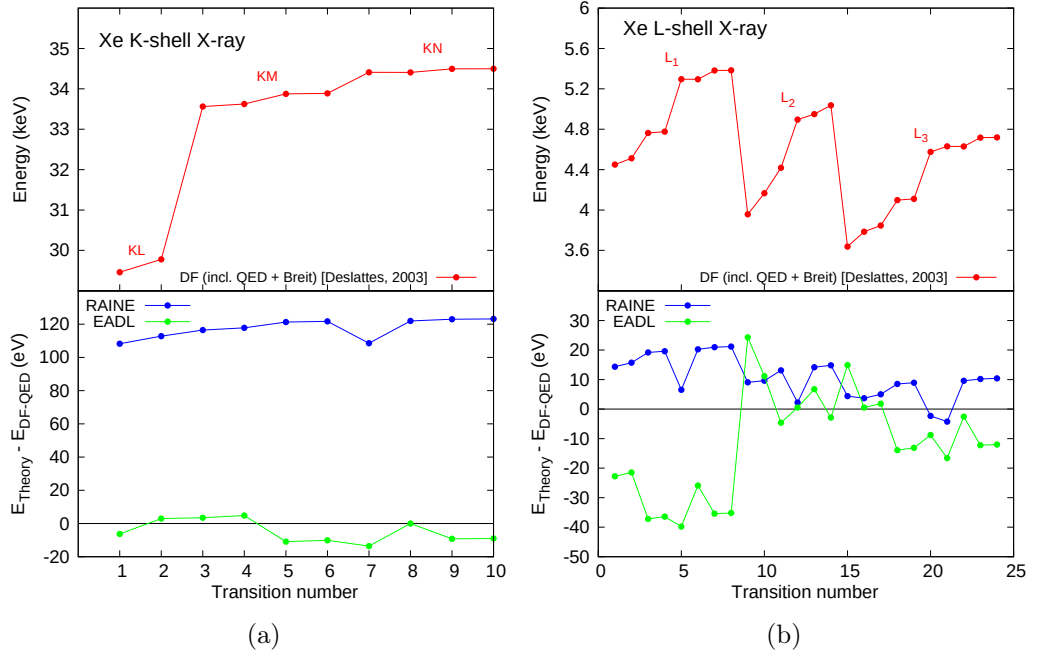


Figure 4.17: Comparison of the theoretical (a) K-shell and (b) L-shell X-ray transition energies from Deslattes et al. [172] with the theoretical energies from the **RAINE** code and **EADL** for xenon. The top panels show the Deslattes' values and the bottom panels show the differences between all three sources.

model like **BrIccEmis**. The bottom panel of Figure 4.18b shows the simulated correlation spectrum from **BrIccEmis** is in good agreement with the experiment. In the simulation, a vacancy cascade is initiated by a M_5 vacancy that is left behind by the ejection of a photoelectron with kinetic energy of 249.3 eV⁵. All vacancies are propagated based on the “random-vacancy” option described in Section 3.4.4⁶. Moreover, the shake-off process is only allowed to accompany the photoelectron and subsequent M_5 -subshell Auger electrons in the first step of the cascade. Auger and Coster-Kronig transitions in the second step of the cascade and after are generally too low in energies for the shake-off process to happen. All shake-off probabilities adopted in the simulation are shown in Table 5.1. In this investigation, the post-collision interaction (PCI) effect [174, 175, 176, 177], which can cause photoelectrons to lose energy and Auger electrons to gain energy, has been neglected since this effect is only important for threshold photoionisation. All sequences of radiations are tracked in the simulation to record the correlations among all emitted electrons which are then used to plot Figure 4.18b.

The simulation reproduces most major features of the experimental spectrum including the distinctive diagonal stripes at the total energies of ~ 415 and ~ 490 eV. Moreover, the simulated M_5 Auger-electron spectrum, which is shown in the top panel of Figure 4.18b

⁵The kinetic energy of the photoelectron is equivalent to the difference between the energy of the synchrotron radiation and the binding energy of the M_5 subshell in vapour xenon [124].

⁶The choice is made based on the results discussed in Chapter 5.

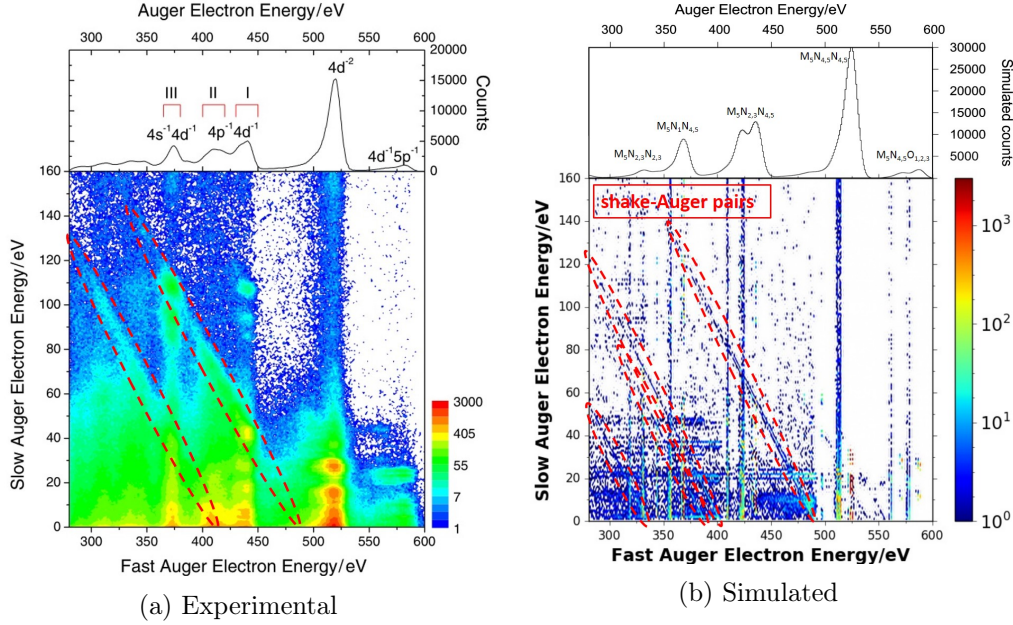


Figure 4.18: Experimental (a) and simulated (b) two-dimensional correlation spectrum between fast Auger electrons (280-600 eV) and slow Auger electrons (0-160 eV) emitted following the creation of an M_5 vacancy. The simulated spectrum is resulted from 100,000 simulated M_5 -photoionisation events and the range of its colorbar is from 1 to 3000. The upper panels show the M_5 Auger-electron spectra. The simulated M_5 Auger-electron spectrum has been folded with an approximate Voigt profile but the shown simulated energy-correlation spectrum is unfolded and binned with 1 eV bin size on both x and y axes. Figure (a) is extracted from Ref. [71].

agrees well with its experimental counterpart (see the top panel of Figure 4.18a). But it is also clear that the calculated energy separation between the two peaks within the $M_5N_{2,3}N_{4,5}$ Auger group is noticeably smaller than the measured one. The diagonal stripes can be sufficiently reproduced in the simulation by using the shake theory from Carlson and Nestor [14] together with the theory for the shake-off electron-energy distribution proposed by Levinger [156]. There are five noticeable diagonal stripes in the simulated correlation spectrum at the total energies of 333, 388, 396, 490 and 492 eV. These lines correspond to $M_5N_1N_{4,5}$, $M_5N_2N_{4,5}$, $M_5N_3N_{4,5}$, $M_5N_4N_5$, and $M_5N_5N_5$ shake-Auger pairs⁷, respectively, in which only the last three pairs are easily seen in the experiment. However, the first two pairs were seen in another experiment reported in Ref. [76]. It is important to note that the two diagonal lines due to the $M_5N_4N_5$, and $M_5N_5N_5$ shake-Auger pairs can only be seen as one line in the experimental spectrum.

The origin of the diagonal stripe at the total energy of 490 eV is depicted in Figure 4.19. Let's imagine a hypothetical scenario when there is no shake-off electron ejected together with the photoelectron. The xenon ion then remains at $1+$ and a subsequent $M_5N_4N_5$ Auger transition will be emitted at 524 eV. These transitions are responsible for the "hot

⁷A shake-Auger pair is defined as a pair of shake-off and Auger electrons that emit simultaneously. In the literature, the pair is also named as "double-Auger" decay.

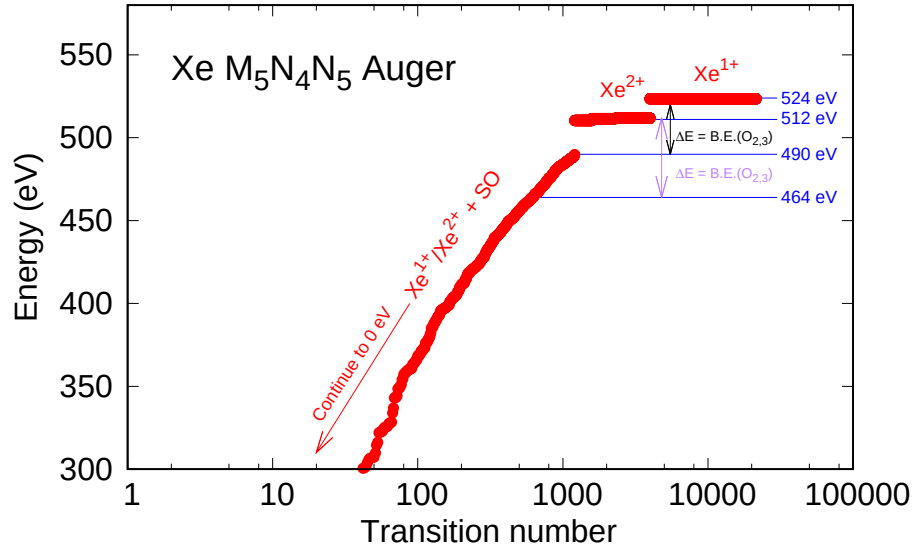


Figure 4.19: Calculated transition energies of $M_5N_4N_5$ Auger in xenon following photoionisation of M_5 -subshell from 100,000 simulation events. There were 21436 $M_5N_4N_5$ Auger electrons recorded from the simulation. The x-axis represents the index number for each $M_5N_4N_5$ Auger electron after they have been sorted in increasing energy order. The total available energy for a $M_5N_4N_5$ shake-Auger pair in Xe^{1+} is 490 eV, which is 34 eV lower than an Auger transition emitted in Xe^{1+} without the shake-off electron, for the liberation of the shake-off electron. See main text for more details.

spots" near 525 eV in Figure 4.18b. On the other hand, if a shake-photoelectron pair⁸, in which the shake-off electron is mostly ejected from the O_3 subshell, a pair of shake-off electron and photoelectron is emitted before the emission of a $M_5N_4N_5$ Auger transition, then the Auger electron will only have 512 eV. If the shake-off electron of the shake-photoelectron pair is ejected from the O_1 or O_2 subshell, then the total energy available for the $M_5N_4N_5$ Auger transition would be less than 512 eV. This is well represented in Figure 4.19 where there is a small dip in energy near the left end of the energy group labelled with Xe^{2+} . However, if a shake-off electron is ejected with the $M_5N_4N_5$ Auger transition in the singly-ionised xenon, i.e. there is no shake-photoelectron pair that precedes the shake-Auger pair, then the total available energy becomes only 490 eV as the excess energy (≥ 34 eV) is used to liberate the shake-off electron from its original subshell. The excess energy described here is also the threshold energy for electron shake-off, E_0 in the Eq. (3.25). The excess energy is equivalent to the binding energy of the subshell where the shake-off electron originates from Xe^{2+} ions, which is the intermediate state before the liberation of the shake-off electron (Xe^{3+} ion). Then the shake-Auger pairs share the total energy of 490 eV, and in the simulation, the Levinger's theory [156] is used to determine the shake-off energy of the pair, which can be any value from zero energy up to the total energy available (see 3.4.6). Therefore, the Auger electron of the pair can have only zero

⁸Similar to a shake-Auger pair, a shake-photoelectron pair consists of a shake-off electron and a photoelectron that are ejected from an atom simultaneously.

energy, i.e. the shake-off electron takes up all energy available, albeit with a very small probability. For example, Figure 4.19 shows that only 42 $M_5N_4N_5$ Auger electrons out of the 1206 recorded shake-Augger pairs have energies below 300 eV. Nonetheless, the two electrons are practically indistinguishable. Therefore, the one with higher energy is usually assigned as the Auger electron while the slower electron is named as the shake-off electron. In an extreme situation, if a shake-Augger pair is emitted after a shake-photoelectron pair, the total energy remaining for the shake-Augger pair to share will only be about 464 eV (diagonal lines labelled as " $M_5N_{4,5}N_5$ shake-Augger pairs in Xe^{2+} " in Figure 4.20a).

Another striking feature of the experimental correlation spectrum that the simulation has reproduced is the existence of multiple vertical lines that span the entire vertical range. The intensities of these vertical lines are mainly contributed by the shake-photoelectron pairs (see Figure 4.20b and 4.20c). After the ejection of a shake-photoelectron pair, the total energy available for the subsequent Auger transition is also reduced; therefore the shake-photoelectron pair will only be in coincidence with the Auger transitions emitted in the doubly-ionised xenon. Figures 4.20a and 4.20c demonstrate that the vertical lines represent the slow Auger electrons (0-160 eV) that are in coincidence with the fast Auger electrons emitted in the doubly-ionised xenon ions, which are created via the emission of the shake-photoelectron pairs.

The increase in the intensity of slow electrons near the top edge of the correlation spectrum (~ 160 eV) is due to contribution of the photoelectrons of the shake-photoelectron pairs. This is because the strong intensities near 160 eV diminish drastically when the photoelectrons are excluded from the simulated spectrum (see Figure 4.20c). If the shake-off process is not allowed to accompany the photoelectrons, then the photoelectrons should always have a kinetic energy that is close to 249.3 eV, which is the difference between the energy of the synchrotron radiation and the binding energy of the M_5 -subshell in atomic xenon. However, if the shake-off process is permitted and it occurs during the emission of a photoelectron, then the energy of the photoelectron is reduced by an amount that is equivalent to the binding energy of the subshells where the shake-off electrons originate from. Since the shake-off electrons are mostly coming from the O_2 or O_3 subshell, the photoelectrons need to "sacrifice" about 24 eV, which is equivalent to the $O_{2,3}$ -subshell binding energy in Xe^{1+} ion, to liberate the shake-off electrons from the xenon ions. Therefore, the shake-photoelectron pairs will share the remaining energy of ~ 226 eV, and the photoelectrons will tend to have the bigger portion according to Levinger's equation (see Section 3.4.6). Thus one should expect a satellite structure of M_5 photoelectrons from ~ 226 eV downwards, and this is consistent with the measured M_5 Auger-electron spectrum from Suzuki et al. [71] (see Figure 4.20e). The energy-sharing process for the shake-photoelectron pairs results in the vertical lines observed with the intensity decreases from 0 to 100 eV (slower electrons of the pairs) but increases afterwards (faster electrons of the pairs). Figure 4.20c shows that the intensities of slow electrons that are in coincidence with the $M_5N_{2,3}N_{4,5}$

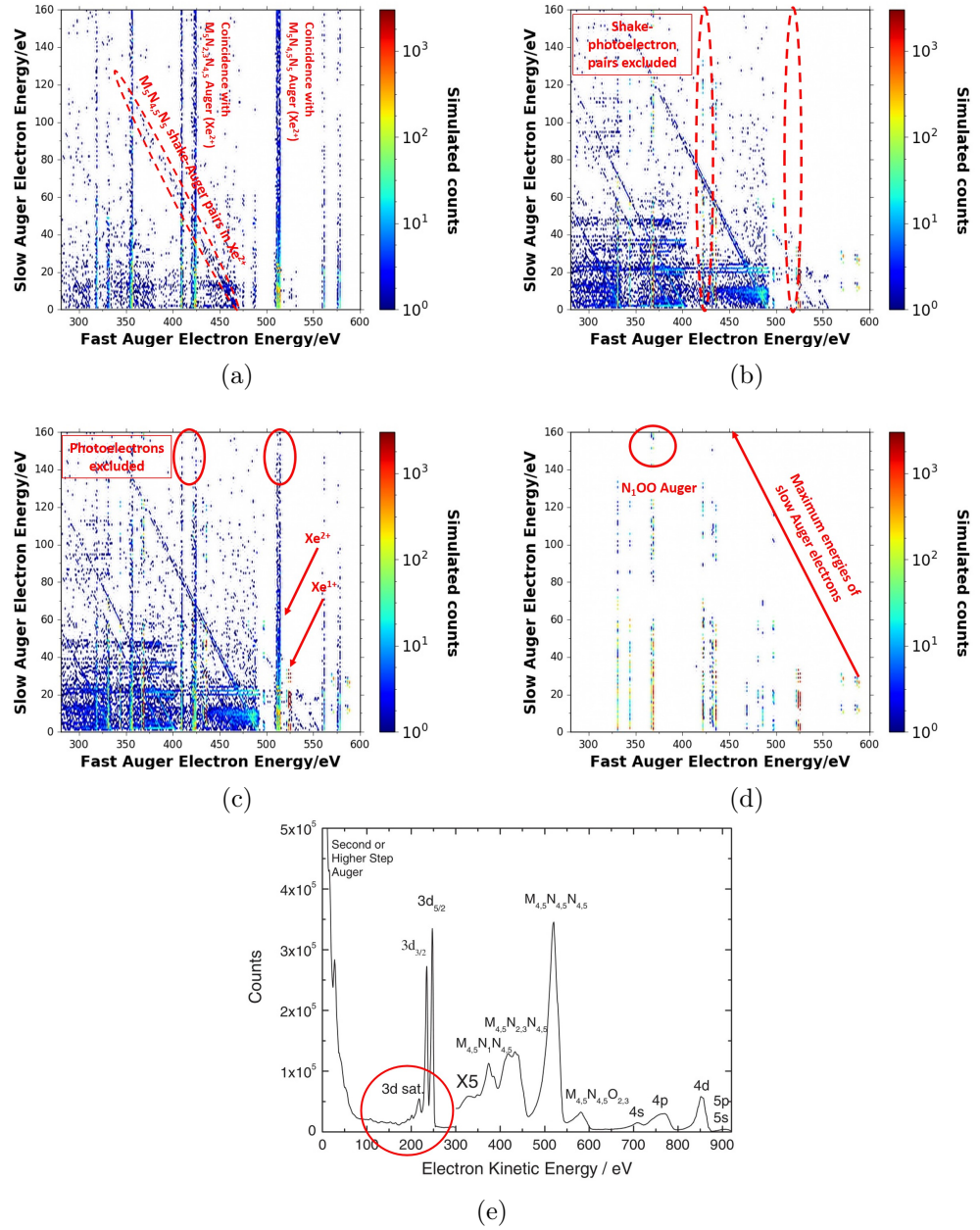


Figure 4.20: Simulated coincidence spectrum filtered with (a) only the cascades that begin with a shake-photoelectron pair, (b) only the cascades that start without the shake-photoelectron pair, or (c) only the Auger and shake-off electrons. (d) shows the coincidence spectrum simulated without the shake-off electrons, and (e) shows the experimental kinetic energy spectrum of electrons in the region of 0-910 eV from Suzuki et al. [71]. The settings of all simulated spectra are the same as those for Figure 4.18b.

(400-440 eV) and $M_5N_{4,5}N_5$ (510-530 eV) Auger electrons are very weak near the top edge of the figure after the photoelectrons are taken out of the spectrum. This strongly suggests that the contribution of the slow Auger electrons to the intensity of slow electrons near 160 eV is negligible.

Figures 4.20b and 4.20c exhibit many noticeable horizontal lines but these lines are

barely visible in Figure 4.20a. Figure 4.20c shows that the intensity of horizontal lines decreases from the high-energy end to the low-energy end. Similarly, the intensity of the diagonal lines decreases as the energy of the slower electrons of the shake-Auger pairs increases⁹. This indicates that the horizontal lines are representing electrons that are in coincidence with the fast Auger electrons that have lost some energy due to the shake-off process.

Figure 4.20d shows the correlation spectrum simulated excluding the shake-off electrons. It is clear from the figure that the maximum energies available after the emission of the fast Auger electrons are less than 160 eV, especially for the secondary transitions that follow $M_5N_{4,5}N_5$. Moreover, Figure 4.20d also indicates that the enhancement in the intensity of slow electrons near 160 eV observed in the experiment (see Figure 4.18a) is likely to be due to the N_5OO Auger transitions, which were neglected by Suzuki et al. [71] in their calculations of the branching ratios from the $4s^{-1}4d^{-1}$ two-hole state. However, it is also evident from Figures 4.18b and 4.20b that the branching ratios to the N_5OO Auger transitions from the $4s^{-1}4d^{-1}$ two-hole state used in the simulation¹⁰ could be higher than the measurement (see Figure 4.18a).

Overall, the simulation using the **BrIccEmis** code is in very good agreement with the experimental spectrum from Suzuki et al. [71]. All main features observed can be sufficiently explained using all the ingredients included in the vacancy-cascade model. Moreover, it is also clear that the shake-off process is extremely important in reproducing the measured spectrum but the shake-off probabilities calculated using the shake-theory from Carlson and Nestor [14] are only valid for near closed-shell atomic structure. It is important to note that **BrIccEmis** assumes the emission of the shake-Auger or shake-photoelectron pairs as a “two-step” process, i.e. the shake-off electron is assumed to be emitted from an intermediate state following the ejection of the Auger electron or photoelectron. For this reason, the agreement between the simulation and the experiment suggests that this “two-step” picture of the simultaneous ionisation of two electrons due to the shake-off process is still valid for Auger electrons and photoelectrons with kinetic energies in the range of 250 to 600 eV.

4.5 Average radiation spectra of medical radioisotopes

This section concerns the atomic radiations emitted in medical radioisotopes which are mostly studied in a molecular environment. The simulations used to produce the results are thus terminated when there are no more energetically allowed transition, or transitions that could happen within 10^{-12} s. The relationship between the atomic radiation yields

⁹Because the shake-off electrons, which are usually the slower electrons of the shake-Auger pairs, are less likely to have high energy, according to the Levinger’s equation (see Section 3.4.6).

¹⁰All branching ratios used **BrIccEmis** are adopted from the EADL database [27]. See Section 3.2.2 for more details.

and the lifetime of the vacancy cascade for the condensed-phase ^{125}I is illustrated in Figure 3.8. The mean lifetimes of vacancies calculated in the singly-ionised systems used for generating the figure are extracted from EADL [27], with the Krause and Carlson correction applied where necessary (see Section 3.4.3). The figure shows that all atomic transitions except low-energy N-shell X-rays, are released by about 10^{-12} s. In addition, some medical radioisotopes have intermediate nuclear states with lifetimes in the order of 10^{-11} s, for example ^{135}La . Therefore, it is assumed here that the vacancy cascade is terminated after 10^{-12} s. Any vacancies remaining after 10^{-12} s are assumed to be neutralised by the environment before the subsequent nuclear transition takes place.

In this section, the simulated average radiation spectra of ^{123}I and ^{125}I , which both have been studied extensively in *in vitro* and *in vivo* experiments in the past few decades [43, 178, 179, 180, 181], are compared with the values published in the literature.

Radioiodines

Tables 4.1 and 4.2 display the simulated average radiation spectra of ^{123}I and ^{125}I ¹¹ for the isolated-atom and condensed-phase approximations (see Section 3.4.7 for detailed explanation of the two approximations). Nuclear radiations with yields¹² of less than 10^{-3} have been excluded. The results of Pomplun [88], Howell [38], and Stepanek [86] are included for comparison. Since Pomplun included shake-off electrons while the others did not, the shake-off process is only included for the **BrIccEmis** simulation of ^{123}I decay.

Pomplun adopted the Dirac-Fock code from Desclaux¹³ [111] to calculate transition energies for both non-radiative and radiative transitions. In general, **BrIccEmis** and Pomplun are in good agreement in terms of average energies and yields, with the exception of N-shell non-radiative transitions. Moreover, Pomplun reported 0.9 shake-off electrons per decay compared to a yield of 0.217 calculated from **BrIccEmis**. The huge intensity of shake-off electrons calculated by Pomplun is associated with the decrease in the intensity of NXY Auger electrons in his simulations. However, both the **BrIccEmis** code and Pomplun's model agree on the total number of electrons released per decay (see Table 4.3).

Transition energies reported by Stepanek were calculated in a similar way to our method. However, his N-shell Auger yields in the decay of ^{125}I are significantly lower. In his calculations, the atom was assumed not to recover its neutral atomic configuration after the first vacancy cascade (following electron capture), thus the atom remained highly-ionised when the second vacancy cascade (following internal conversion) took place. In the

¹¹Decay schemes of ^{123}I and ^{125}I are shown in Figures 3.10a and 2.6, respectively. Distribution of initial vacancies created in these decays can be found in Tables A.5 and A.6 (in Appendix A.5).

¹²Yield is defined as the number of radiations released per decay.

¹³It is not clear that whether QED and Breit corrections were accounted for in the Pomplun's model. However, given that Deslattes [172] required other codes different from the original Desclaux code to account for QED and Breit corrections, therefore it is assumed that these corrections were not included in the Pomplun's model.

Table 4.1: Average spectrum per decay for isolated atom and condensed phase of ^{123}I . This table has been published in Ref. [122].

Process	Isolated atom				Condensed phase			
	BrIccEmis		Pomplun [88]		BrIccEmis		Howell [38]	
	Average energy (eV)	Yield	Average energy (eV)	Yield	Average energy (eV)	Yield	Average energy(eV)	Yield
γ_1	$1.59 \cdot 10^5$	$8.36 \cdot 10^{-1}$	$1.59 \cdot 10^5$	$8.45 \cdot 10^{-1}$	$1.59 \cdot 10^5$	$8.38 \cdot 10^{-1}$	$1.59 \cdot 10^5$	$8.39 \cdot 10^{-1}$
IC ₁ K	$1.27 \cdot 10^5$	$1.34 \cdot 10^{-1}$	$1.27 \cdot 10^5$	$1.28 \cdot 10^{-1}$	$1.27 \cdot 10^5$	$1.33 \cdot 10^{-1}$	$1.27 \cdot 10^5$	$1.30 \cdot 10^{-1}$
IC ₁ L	$1.54 \cdot 10^5$	$1.73 \cdot 10^{-2}$	$1.54 \cdot 10^5$	$1.71 \cdot 10^{-2}$	$1.54 \cdot 10^5$	$1.71 \cdot 10^{-2}$	$1.54 \cdot 10^5$	$1.79 \cdot 10^{-2}$
IC ₁ M ₊	$1.58 \cdot 10^5$	$4.11 \cdot 10^{-3}$	$1.58 \cdot 10^5$	$3.80 \cdot 10^{-3}$	$1.58 \cdot 10^5$	$4.42 \cdot 10^{-3}$	$1.58 \cdot 10^5$	$5.30 \cdot 10^{-3}$
γ_3	$4.40 \cdot 10^5$	$4.00 \cdot 10^{-3}$	$4.40 \cdot 10^5$	$3.00 \cdot 10^{-3}$	$4.40 \cdot 10^5$	$4.03 \cdot 10^{-3}$	$4.40 \cdot 10^5$	$3.90 \cdot 10^{-3}$
γ_5	$3.46 \cdot 10^5$	$1.25 \cdot 10^{-3}$	$3.46 \cdot 10^5$	$7.00 \cdot 10^{-4}$	$3.46 \cdot 10^5$	$1.06 \cdot 10^{-3}$	$3.46 \cdot 10^5$	$1.40 \cdot 10^{-3}$
γ_6	$5.05 \cdot 10^5$	$2.85 \cdot 10^{-3}$	$5.05 \cdot 10^5$	$2.50 \cdot 10^{-3}$	$5.05 \cdot 10^5$	$2.87 \cdot 10^{-3}$	$5.05 \cdot 10^5$	$2.80 \cdot 10^{-3}$
γ_{12}	$5.29 \cdot 10^5$	$1.38 \cdot 10^{-2}$	$5.29 \cdot 10^5$	$1.13 \cdot 10^{-2}$	$5.29 \cdot 10^5$	$1.38 \cdot 10^{-2}$	$5.29 \cdot 10^5$	$1.45 \cdot 10^{-2}$
γ_{17}	$5.39 \cdot 10^5$	$3.85 \cdot 10^{-3}$	$5.39 \cdot 10^5$	$2.80 \cdot 10^{-3}$	$5.39 \cdot 10^5$	$3.82 \cdot 10^{-3}$	$5.39 \cdot 10^5$	$4.50 \cdot 10^{-3}$
Auger KLL	$2.26 \cdot 10^4$	$8.27 \cdot 10^{-2}$	$2.25 \cdot 10^4$	$7.31 \cdot 10^{-2}$	$2.26 \cdot 10^4$	$8.14 \cdot 10^{-2}$	$2.24 \cdot 10^4$	$8.38 \cdot 10^{-2}$
Auger KLX	$2.65 \cdot 10^4$	$3.56 \cdot 10^{-2}$	$2.64 \cdot 10^4$	$3.28 \cdot 10^{-2}$	$2.65 \cdot 10^4$	$3.57 \cdot 10^{-2}$	$2.63 \cdot 10^4$	$3.84 \cdot 10^{-2}$
Auger KXY	$3.04 \cdot 10^4$	$3.96 \cdot 10^{-3}$	$3.03 \cdot 10^4$	$2.80 \cdot 10^{-3}$	$3.04 \cdot 10^4$	$4.15 \cdot 10^{-3}$	$3.02 \cdot 10^4$	$3.50 \cdot 10^{-3}$
CK LLX	$2.82 \cdot 10^2$	$1.54 \cdot 10^{-1}$	$2.80 \cdot 10^2$	$1.34 \cdot 10^{-1}$	$2.88 \cdot 10^2$	$1.55 \cdot 10^{-1}$	$2.13 \cdot 10^2$	$1.56 \cdot 10^{-1}$
Auger LMM	$3.04 \cdot 10^3$	$7.39 \cdot 10^{-1}$	$3.03 \cdot 10^3$	$7.11 \cdot 10^{-1}$	$3.05 \cdot 10^3$	$7.32 \cdot 10^{-1}$	$3.04 \cdot 10^3$	$7.51 \cdot 10^{-1}$
Auger LMX	$3.67 \cdot 10^3$	$2.06 \cdot 10^{-1}$	$3.66 \cdot 10^3$	$2.00 \cdot 10^{-1}$	$3.68 \cdot 10^3$	$2.08 \cdot 10^{-1}$	$3.66 \cdot 10^3$	$2.02 \cdot 10^{-1}$
Auger LXY	$4.31 \cdot 10^3$	$1.51 \cdot 10^{-2}$	$4.31 \cdot 10^3$	$1.22 \cdot 10^{-2}$	$4.32 \cdot 10^3$	$1.50 \cdot 10^{-2}$	$4.28 \cdot 10^3$	$1.30 \cdot 10^{-2}$
CK MMX	$9.42 \cdot 10^1$	$7.80 \cdot 10^{-1}$	$9.24 \cdot 10^1$	$8.70 \cdot 10^{-1}$	$1.03 \cdot 10^2$	$7.91 \cdot 10^{-1}$	$1.27 \cdot 10^2$	$8.69 \cdot 10^{-1}$
Auger MXY	$4.06 \cdot 10^2$	$1.95 \cdot 10^0$	$3.94 \cdot 10^2$	$1.93 \cdot 10^0$	$4.27 \cdot 10^2$	$1.94 \cdot 10^0$	$4.61 \cdot 10^2$	$1.97 \cdot 10^0$
CK NNX	$2.00 \cdot 10^1$	$1.32 \cdot 10^0$	$2.85 \cdot 10^1$	$1.27 \cdot 10^0$	$2.19 \cdot 10^1$	$2.00 \cdot 10^0$	$2.98 \cdot 10^1$	$2.10 \cdot 10^0$
Auger NXY	$2.19 \cdot 10^1$	$2.03 \cdot 10^0$	$3.02 \cdot 10^1$	$1.13 \cdot 10^0$	$1.49 \cdot 10^1$	$6.36 \cdot 10^0$	$3.25 \cdot 10^1$	$6.54 \cdot 10^0$
CK OOX	—	—	—	—	—	—	$6.00 \cdot 10^0$	$2.18 \cdot 10^0$
Shake-off	$3.09 \cdot 10^1$	$2.17 \cdot 10^{-1}$	^a	$9.00 \cdot 10^{-1}$	—	—	—	—
X-ray K _{α_1}	$2.76 \cdot 10^4$	$4.66 \cdot 10^{-1}$	$2.75 \cdot 10^4$	$4.47 \cdot 10^{-1}$	$2.76 \cdot 10^4$	$4.63 \cdot 10^{-1}$	$2.75 \cdot 10^4$	$4.62 \cdot 10^{-1}$
X-ray K _{α_2}	$2.73 \cdot 10^4$	$2.51 \cdot 10^{-1}$	$2.72 \cdot 10^4$	$2.48 \cdot 10^{-1}$	$2.73 \cdot 10^4$	$2.50 \cdot 10^{-1}$	$2.72 \cdot 10^4$	$2.37 \cdot 10^{-1}$
X-ray K _{β_1}	$3.11 \cdot 10^4$	$8.17 \cdot 10^{-2}$	$3.10 \cdot 10^4$	$8.19 \cdot 10^{-2}$	$3.11 \cdot 10^4$	$8.17 \cdot 10^{-2}$	$3.10 \cdot 10^4$	$8.13 \cdot 10^{-2}$
X-ray K _{β_2}	$3.18 \cdot 10^4$	$2.61 \cdot 10^{-2}$	$3.17 \cdot 10^4$	$2.24 \cdot 10^{-2}$	$3.18 \cdot 10^4$	$2.58 \cdot 10^{-2}$	$3.17 \cdot 10^4$	$2.27 \cdot 10^{-2}$
X-ray K _{β_3}	$3.10 \cdot 10^4$	$4.34 \cdot 10^{-2}$	$3.09 \cdot 10^4$	$3.97 \cdot 10^{-2}$	$3.10 \cdot 10^4$	$4.36 \cdot 10^{-2}$	$3.09 \cdot 10^4$	$4.45 \cdot 10^{-2}$
X-ray KM ₊	$3.17 \cdot 10^4$	$2.32 \cdot 10^{-3}$	$3.12 \cdot 10^4$	$8.00 \cdot 10^{-4}$	$3.17 \cdot 10^4$	$2.35 \cdot 10^{-3}$	$3.17 \cdot 10^4$	$1.30 \cdot 10^{-3}$
X-ray L	$3.93 \cdot 10^3$	$8.90 \cdot 10^{-2}$	$3.94 \cdot 10^3$	$8.22 \cdot 10^{-2}$	$3.93 \cdot 10^3$	$8.96 \cdot 10^{-2}$	$3.93 \cdot 10^3$	$7.90 \cdot 10^{-2}$
X-ray M	$5.41 \cdot 10^2$	$4.58 \cdot 10^{-3}$	—	—	$5.46 \cdot 10^2$	$4.61 \cdot 10^{-3}$	$5.43 \cdot 10^2$	$2.30 \cdot 10^{-3}$
X-ray N	$9.09 \cdot 10^1$	$1.08 \cdot 10^{-1}$	—	—	$1.02 \cdot 10^2$	$2.81 \cdot 10^{-3}$	—	—

^a Average shake-off energy is not reported by Pomplun [88].

Table 4.2: Average spectrum per decay for isolated atom and condensed phase of ^{125}I . This table has been published in Ref. [122].

Process	Isolated atom				Condensed phase			
	BrIccEmis		Stepanek [86]		BrIccEmis		Howell [38]	
	Average energy (eV)	Yield	Average energy (eV)	Yield	Average energy (eV)	Yield	Average energy(eV)	Yield
γ_1	$3.55 \cdot 10^4$	$6.65 \cdot 10^{-2}$	$3.55 \cdot 10^4$	$6.66 \cdot 10^{-2}$	$3.55 \cdot 10^4$	$6.52 \cdot 10^{-2}$	$3.55 \cdot 10^4$	$6.47 \cdot 10^{-2}$
IC ₁ K	$3.68 \cdot 10^3$	$7.75 \cdot 10^{-1}$	$3.69 \cdot 10^3$	$8.04 \cdot 10^{-1}$	$3.68 \cdot 10^3$	$7.76 \cdot 10^{-1}$	$3.65 \cdot 10^3$	$7.97 \cdot 10^{-1}$
IC ₁ L	$3.07 \cdot 10^4$	$1.26 \cdot 10^{-1}$	$3.06 \cdot 10^4$	$1.08 \cdot 10^{-1}$	$3.07 \cdot 10^4$	$1.28 \cdot 10^{-1}$	$3.06 \cdot 10^4$	$1.10 \cdot 10^{-1}$
IC ₁ M ₊	$3.47 \cdot 10^4$	$3.25 \cdot 10^{-2}$	$3.45 \cdot 10^4$	$2.15 \cdot 10^{-2}$	$3.47 \cdot 10^4$	$3.08 \cdot 10^{-2}$	$3.47 \cdot 10^4$	$2.84 \cdot 10^{-2}$
Auger KLL	$2.26 \cdot 10^4$	$1.28 \cdot 10^{-1}$	$2.26 \cdot 10^4$	$1.26 \cdot 10^{-1}$	$2.26 \cdot 10^4$	$1.30 \cdot 10^{-1}$	$2.24 \cdot 10^4$	$1.38 \cdot 10^{-1}$
Auger KLX	$2.65 \cdot 10^4$	$5.67 \cdot 10^{-2}$	$2.65 \cdot 10^4$	$5.80 \cdot 10^{-2}$	$2.65 \cdot 10^4$	$5.65 \cdot 10^{-2}$	$2.64 \cdot 10^4$	$5.90 \cdot 10^{-2}$
Auger KXY	$3.04 \cdot 10^4$	$5.97 \cdot 10^{-3}$	$3.03 \cdot 10^4$	$5.50 \cdot 10^{-3}$	$3.04 \cdot 10^4$	$5.96 \cdot 10^{-3}$	$3.02 \cdot 10^4$	$6.50 \cdot 10^{-3}$
CK LLX	$2.87 \cdot 10^2$	$2.69 \cdot 10^{-1}$	$2.56 \cdot 10^2$	$2.57 \cdot 10^{-1}$	$2.90 \cdot 10^2$	$2.68 \cdot 10^{-1}$	$2.19 \cdot 10^2$	$2.64 \cdot 10^{-1}$
Auger LMM	$3.05 \cdot 10^3$	$1.22 \cdot 10^0$	$3.01 \cdot 10^3$	$1.22 \cdot 10^0$	$3.05 \cdot 10^3$	$1.22 \cdot 10^0$	$3.05 \cdot 10^3$	$1.25 \cdot 10^0$
Auger LMX	$3.68 \cdot 10^3$	$3.42 \cdot 10^{-1}$	$3.63 \cdot 10^3$	$3.39 \cdot 10^{-1}$	$3.68 \cdot 10^3$	$3.44 \cdot 10^{-1}$	$3.67 \cdot 10^3$	$3.40 \cdot 10^{-1}$
Auger LXY	$4.30 \cdot 10^3$	$2.33 \cdot 10^{-2}$	$4.28 \cdot 10^3$	$2.24 \cdot 10^{-2}$	$4.31 \cdot 10^3$	$2.39 \cdot 10^{-2}$	$4.34 \cdot 10^3$	$2.11 \cdot 10^{-2}$
CK MMX	$9.39 \cdot 10^1$	$1.30 \cdot 10^0$	$7.58 \cdot 10^1$	$1.04 \cdot 10^0$	$1.00 \cdot 10^2$	$1.32 \cdot 10^0$	$1.27 \cdot 10^2$	$1.44 \cdot 10^0$
Auger MXY	$4.08 \cdot 10^2$	$3.22 \cdot 10^0$	$3.80 \cdot 10^2$	$3.24 \cdot 10^0$	$4.22 \cdot 10^2$	$3.23 \cdot 10^0$	$4.61 \cdot 10^2$	$3.28 \cdot 10^0$
CK NNX	$1.99 \cdot 10^1$	$2.18 \cdot 10^0$	$2.74 \cdot 10^1$	$1.21 \cdot 10^0$	$2.19 \cdot 10^1$	$3.20 \cdot 10^0$	$2.99 \cdot 10^1$	$3.51 \cdot 10^0$
Auger NXY	$2.23 \cdot 10^1$	$3.15 \cdot 10^0$	$4.53 \cdot 10^1$	$1.40 \cdot 10^0$	$1.41 \cdot 10^1$	$1.02 \cdot 10^1$	$3.24 \cdot 10^1$	$1.09 \cdot 10^1$
CK OOX	—	—	—	—	—	—	$6.00 \cdot 10^0$	$3.66 \cdot 10^0$
X-ray K _{α_1}	$2.76 \cdot 10^4$	$7.41 \cdot 10^{-1}$	$2.76 \cdot 10^4$	$7.62 \cdot 10^{-1}$	$2.76 \cdot 10^4$	$7.41 \cdot 10^{-1}$	$2.75 \cdot 10^4$	$7.51 \cdot 10^{-1}$
X-ray K _{α_2}	$2.73 \cdot 10^4$	$3.98 \cdot 10^{-1}$	$2.73 \cdot 10^4$	$4.03 \cdot 10^{-1}$	$2.73 \cdot 10^4$	$4.01 \cdot 10^{-1}$	$2.72 \cdot 10^4$	$3.94 \cdot 10^{-1}$
X-ray K _{β_1}	$3.11 \cdot 10^4$	$1.31 \cdot 10^{-1}$	$3.11 \cdot 10^4$	$1.35 \cdot 10^{-1}$	$3.11 \cdot 10^4$	$1.34 \cdot 10^{-1}$	$3.10 \cdot 10^4$	$1.38 \cdot 10^{-1}$
X-ray K _{β_2}	$3.18 \cdot 10^4$	$4.15 \cdot 10^{-2}$	$3.18 \cdot 10^4$	$3.96 \cdot 10^{-2}$	$3.18 \cdot 10^4$	$4.04 \cdot 10^{-2}$	$3.17 \cdot 10^4$	$4.03 \cdot 10^{-2}$
X-ray K _{β_3}	$3.10 \cdot 10^4$	$6.84 \cdot 10^{-2}$	$3.11 \cdot 10^4$	$6.71 \cdot 10^{-2}$	$3.10 \cdot 10^4$	$6.86 \cdot 10^{-2}$	$3.09 \cdot 10^4$	$6.85 \cdot 10^{-2}$
X-ray KM ₊	$3.17 \cdot 10^4$	$3.86 \cdot 10^{-3}$	$3.14 \cdot 10^4$	$1.50 \cdot 10^{-3}$	$3.17 \cdot 10^4$	$3.65 \cdot 10^{-3}$	$3.16 \cdot 10^4$	$4.20 \cdot 10^{-3}$
X-ray L	$3.93 \cdot 10^3$	$1.50 \cdot 10^{-1}$	$3.91 \cdot 10^3$	$1.61 \cdot 10^{-1}$	$3.93 \cdot 10^3$	$1.49 \cdot 10^{-1}$	$2.93 \cdot 10^3$	$1.32 \cdot 10^{-1}$
X-ray M	$5.46 \cdot 10^2$	$8.38 \cdot 10^{-3}$	—	—	$5.56 \cdot 10^2$	$7.75 \cdot 10^{-3}$	$5.42 \cdot 10^2$	$4.00 \cdot 10^{-3}$
X-ray N	$9.16 \cdot 10^1$	$1.95 \cdot 10^{-1}$	—	—	$9.91 \cdot 10^1$	$6.40 \cdot 10^{-2}$	—	—

case of ^{125}I , the excited state of the daughter nucleus ^{125}Te has a half-life of 1.48 ns, which means that no interaction between the atom and the environment for at least 10^{-9} s was

assumed in Stepanek’s model.

For ^{123}I and ^{125}I in the condensed-phase approximations, there is a good agreement in terms of radiation yields between the present spectra and those of Howell up to N-shell transitions. Calculated average energies of outer-shell transitions in this work are consistently lower than Howell’s values. This can be attributed to the fact that Howell’s model adopted the $(Z+1)/Z$ rule [116] for the calculations of Auger-electron energies and did not account for the effect of spectator vacancies. The effect of spectator vacancies can reduce the transition energies resulting in lower average transition energies, especially for those emitted in the later stages of the vacancy cascade. It is important to note that Howell also included O-shell non-radiative transitions in his calculations, resulting in an extra 2.18 and 3.66 very low-energy OOX transitions for ^{123}I and ^{125}I , respectively. These transitions are not present in the EADL [27] and it is highly debatable to include them in the simulations. O-shell non-radiative transitions of singly-ionised xenon, which has two more electrons than singly-ionised tellurium, are determined to be energetically forbidden in the relativistic Dirac-Fock method by using the atomic structure packages GRASP2K [150] and RATIP [151]¹⁴. Moreover, the energy of the $\text{O}_1\text{O}_3\text{O}_3$ super-Coster-Kronig transition, which should have the largest energies among all O-shell non-radiative transitions, is calculated to -6.2 eV by the RAINE code. This indicates that the OOX transitions in tellurium are unphysical, and therefore they are not considered in BrIccEmis.

Table 4.3: Comparison of calculated Auger and Coster-Kronig yields with the literature for ^{123}I , ^{124}I and ^{125}I . All values shown below are calculated from simulations excluding the shake-off electrons unless specified. This table has been published in Ref. [122].

Radio-nuclide	Isolated atom			Condensed phase				
	BrIccEmis	Stepanek [86]	Pomplun [87, 88]	BrIccEmis	Nikjoo et al. [118]	Howell [38]	Stepanek [117]	Pomplun [87]
^{123}I	7.38/7.54 ^a		7.30 ^a	12.3		14.9		
^{124}I	5.04			8.38	8.2			
^{125}I	11.9	8.92	12.2	20.0	20.2	24.9	14.5	18.3

^a Shake-off electrons included.

A comparison of the simulated Auger and Coster-Kronig yields with the literature values for ^{123}I , ^{124}I and ^{125}I is shown in Table 4.3. Excellent agreement with the results from Nikjoo et al. [118], in which the $Z/Z+1$ rule [116] was used, is obtained even though two very different methods of calculating transition energies are used. This agreement shows that the $Z/Z+1$ rule, which was used by Nikjoo et al. as well as Howell [38], is a good approximation for Auger electron energies in the condensed-phase condition, where the effect of spectator vacancies is diminished, even though it does not account for a realistic vacancy distribution. Due to the presence of multiple vacancies during the vacancy cascade, one Auger electron transition can carry away many different kinetic energies, but this is ignored by the $Z/Z+1$ rule. The changes in transition energies need to be carefully

¹⁴The calculations were performed by Dr. Jörgen Ekman of Malmö University.

determined and accounted for since the interaction of low-energy electrons with water (≤ 1000 eV) is highly dependent of their incident energy [118]. Howell [38] agreed that each Auger electron energy should be calculated based on the exact electron configuration for that particular event but he chose the $Z/Z + 1$ rule due to the extensive CPU time required for the strict energy bookkeeping.

4.6 Discussion

Comparisons shown in Section 4.2.1 demonstrate that the Dirac-Fock method in the implementation of the **RAINE** code [112] is in better agreement with the widely accepted semi-empirical values from the Larkins tabulations [28] than the EADL database [27]. It is important to note that EADL-derived non-radiative transition energies based on neutral binding energies and Eq. (4.1). This method tends to overestimate the non-radiative transition energies and therefore several existing programs [38, 86, 118] choose to adopt the $Z/Z + 1$ rule [116] for transitions emitted from an element Z :

$$E_{\text{WXY}} = \text{B.E.}(\text{W})^Z - 0.5(\text{B.E.}(\text{X})^Z + \text{B.E.}(\text{Y})^Z) + 0.5(\text{B.E.}(\text{X})^{Z+1} + \text{B.E.}(\text{Y})^{Z+1}), \quad (4.8)$$

where E_{WXY} is the energy of the non-radiative transition WXY and $\text{B.E.}(\text{W})^Z$, $\text{B.E.}(\text{X})^Z$ and $\text{B.E.}(\text{Y})^Z$ are the neutral binding energies of W, X and Y subshells, respectively; $\text{B.E.}(\text{X})^{Z+1}$ and $\text{B.E.}(\text{Y})^{Z+1}$ are the neutral binding energies of X and Y subshells in the element $Z + 1$, respectively.

Although **RAINE** tends to overestimate K-shell transitions for heavy elements due to the lack of QED and Breit corrections (see Sections 4.2 and 4.3), it has also been shown to agree well with experimental data for transitions from the L shell and above in both the singly-ionised and multiply-ionised systems (see Section 4.2 and 4.4). Therefore, it is still a valuable tool for vacancy-cascade simulations that can involve highly-ionised systems, where the $Z/Z + 1$ rule and Eq. (4.1) becomes invalid. Future upgrades of **RAINE** via the inclusions of QED and Breit corrections are crucial to improve the agreement with the experimental data and semi-empirical values.

Comparison of the simulated L-Auger spectrum with experiment for ^{131}Cs , which decays to ^{131}Xe by electron capture, in Section 4.2.2 demonstrates that users of **BrIccEmis** must be aware of the energy shift from the vapour phase to the solid system, i.e the vapour-metal shift. Transition energies calculated by the **RAINE** code are referenced to the vacuum level but experimental transition energies are referenced to the Fermi level when solid radiation sources are used. Figure 4.11 shows that there is a near-perfect agreement between the **BrIccEmis** simulation and the experimental data from Kovalík et al. [144]. However, the agreement is likely to be due to the cancellation of two opposite effects. The **RAINE** code tends to overestimate the L-shell Auger transition energies in the singly-ionised

xenon by about 15 eV compared to Larkins' semi-empirical values [28] (see Figure 4.6b), but the vapour-metal shift can increase the energies referenced to the Fermi level by 10-15 eV [167, 168]. The overestimation in the transition energies by RAINE is fairly consistent over a wide range of subshells and elements (see Section 4.2.1).

Recently, there is an increased interest in the enhancement of secondary electrons from the gold substrate as the medical applications of low-energy electrons emitted from gold nanoparticles via external ionisation have been studied extensively in the last few years [8, 10, 182]. Therefore, detailed calculations of the low-energy electron spectrum from ^{125}I that is deposited on a gold substrate will be important in quantifying the enhancement of secondary electrons from the substrate. Figure 4.13 shows that Auger-energy spectra simulated using BrIccEmis are in fairly good agreement with the recent measurement of electrons emitted from ^{125}I at ANU. The K-shell conversion line at 3678 eV provides the reference level for absolute intensity since the number of K-shell conversion electrons emitted per decay of ^{125}I is well-known. Intensities of all Auger energy lines can be calibrated to the intensity of the K-shell conversion line. Therefore, the differences between the simulation and the experiment could be assigned to sources of low-energy electrons other than the ^{125}I source. These low-energy electrons are likely to originate from the ionisations of gold atoms in the substrate via inelastic scattering of primary radiations from the ^{125}I source. Hence, the number of low-energy electrons emitted from the gold substrate per decay of ^{125}I could be directly measured. However, the differences between the simulation and the experiment could also be due to the incorrect Auger transition probabilities from EADL [27]. Thus the L-shell Auger-energy spectrum (2000-4000 eV) needs to be fully understood before one could attempt to quantify the enhancement of low-energy electrons from gold.

Comparisons of residual charge-state distributions with experimental data in Chapter 5 have shown that BrIccEmis has captured most physics of the vacancy cascade in terms of the multiplicity distributions of emitted electrons. But the energy distribution of electrons is lost in the charge-state measurements. Therefore, the recent energy spectrum of low-energy electrons (< 900 eV) emitted following M_5 -subshell photoionisation reported by Suzuki et al. [71] provides invaluable data for the benchmarking of BrIccEmis. Moreover, the experimental spectrum also revealed the sequences of emitted Auger electrons since it was measured using a coincidence technique. Figure 4.18b shows that the BrIccEmis simulation can reproduce most features of the experimental spectrum when the shake-off process is included. This means that BrIccEmis has the necessary ingredients to simulate the true events that occur after the creation of a vacancy in the M_5 subshell fairly well. Moreover, the calculated energy distributions in multiply-ionised systems are also in good agreement with the experiment. This agreement indicates that the validity of the "two-step" picture for the emission of the shake-Auger or shake-photoelectron pairs is still established for the Auger electrons or photoelectrons with kinetic energies around 250 eV.

However, the validity of this assumption for electrons with lower kinetic energies should be investigated in future studies. Furthermore, detailed comparisons of simulated energy-correlation spectra and experimental data in future studies can pave the way to improve the branching ratios reported in EADL [27], which are adopted by **BrIccEmis** as well as many other Monte Carlo codes such as **PENELOPE** [183] and **Geant4** [184].

Comparisons in Section 4.5 have shown that the simulated average radiation spectra from **BrIccEmis** are generally in good agreement with the literature values apart from the N-shell non-radiative transitions. It is important to note that the average radiation spectra are frequently used by researchers in medical physics for dosimetric calculations of Auger therapy [159, 185, 186]. However, many fine details about the emitted Auger electrons and X-rays are lost in the average radiation spectra. The impact of averaging a radiation spectrum on the dosimetric calculations will be discussed in Chapter 7.

4.7 Summary

The **RAINE** code [112], which is adopted by **BrIccEmis** for the energy calculation, has been shown to agree well the semi-empirical Auger-electron energies from Larkins [28] for the singly-ionised systems, apart from the K-shell transitions in heavy elements where QED and Breit corrections, which are not accounted by **RAINE**, become important. A similar trend is seen for the X-ray energies when the calculation of **RAINE** is compared to the latest theoretical values from Deslattes et al. [172]. For X-ray transitions, EADL [27], which derives energies based on neutral binding energies, is in better agreement with Deslattes than **RAINE** for singly-ionised systems. **BrIccEmis** simulation has also been demonstrated to be in excellent agreement with experiments for Auger electrons emitted from the L-shell and above.

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Residual charge-state distribution following inner-shell ionisation

Overview

In this chapter, the charge-state distributions simulated by `BrIccEmis` are benchmarked against the experimental data for the M_5 -subshell photoionisation of atomic xenon and the decay of ^{131m}Xe . The impact of a new set of atomic transition probabilities and the effect of the inaccurate calculation of energies by the `RAINE` code are also presented. All simulated results shown in this chapter are based on 100,000 simulated events unless specified otherwise.

5.1 Background

Due to the emission of Auger, Coster-Kronig and shake-off electrons, the vacancy cascade will yield highly ionised states. During the 1950s, measurements of residual charge distribution were primarily performed for nuclear decays of noble gases [187, 188, 189, 190, 191]. From the 1960s up to early 1990s, the focus shifted to studying residual charge distributions of rare gases in vacuum following photoionisation [39, 40, 41, 85, 192, 193, 194]. It was during this time that Krause and Carlson [85] first used the concept of a vacancy cascade to explain the observed charge distributions of krypton ions following inner-shell photoionisation. Since the residual charge-state distribution is a signature of the vacancy cascade, it has been used by Mukoyama, Pomplun and others, to validate their calculations in the isolated-atom approximation [54, 87, 88, 153].

In recent years, new measurements of residual charge-state distribution following inner-shell photoionisation of xenon have been performed using time-of-flight (TOF) mass spectrometers [195, 196, 197]. These new measurements allow comparison between theory and experiment with smaller uncertainty inherited from the distribution of initial atomic holes. Moreover, these charge-state distributions can be used to study the sequences of transitions emitted in the vacancy cascade.

In this chapter, a comparison of the simulated charge-state distribution with experi-

mental data for the initial vacancies created via the M_5 -subshell photoionisation of atomic xenon and the decay of ^{131m}Xe will be presented. The impact of a new set of atomic transition probabilities, which are calculated based on the Dirac-Fock method in replacement of those from the Evaluated Atomic Data Library (EADL) [27], on the simulation of charge-state distributions will be discussed. Moreover, a qualitative study on the effect of the inaccurate calculation of transition energies by the RAINE code [112] will be described.

5.2 Xe M_5 photoionisation

Tamenori et al. [195], Eland et al. [197] and Partanen et al. [196] recently measured the residual charge-state distribution of Xe ions following photoionisation of the M_5 subshell using TOF mass spectrometers. Apart from the work in Partanen et al., the charge-state distributions reported in the other two publications are measured using coincidence techniques.

5.2.1 Calculation of shake-off rates for atomic xenon

Due to energy restrictions, shake-off is only important for the first Auger transition in the vacancy cascade following the creation of an M_5 atomic hole. The total energy available for the subsequent transitions is smaller than 120 eV while the ionisation energy of Xe^{3+} is at least 42 eV [198]. It is important to note that this is the energy range where the sudden approximation becomes invalid (see Eq. (3.16)).

Carlson and Nestor [14] did not provide the probability for shake-off accompanying Auger processes and the approximation for these shaking processes described in Section 3.4.5 is not very good. Instead of using the standard protocol described in Chapter 3, the shake-off probabilities due to photoionisation of the M_5 subshell and the M_5 -Auger processes have been recalculated based on the methodology described by Carlson and Nestor (see Eq. (2.33)). The quantity P_F in Eq. (2.34), which represents the transition probability to occupied bound states that are forbidden due to Pauli principle, is neglected but this only incurs a small error in the rates as the term $\int \psi_{n'l}^{\prime*} \psi_{nl} d\tau$ in Eq. (2.34) is usually smaller than the term $\int \psi_{n'l}^{\prime*} \psi_{nl} d\tau$ in Eq. (2.33) by at least two orders of magnitude. The single-electron wave functions of the neutral atom (initial state for photoionisation), ion with a single-hole configuration (final state for photoionisation or initial state for Auger process) and ion with a two-hole configuration (final state for Auger process) can be calculated using the self-consistent-field Dirac-Fock method. The Dirac-Fock single-electron wave functions were calculated using the **Grasp2K** package [150] and the **xToolbox** program of the **Ratip** distribution [151] was used to extract the wave-function overlaps. Electron-electron correlation was not accounted for in these calculations. Table 5.1 shows the calculated shake-off probabilities for photoionisation of M_5 subshells and several subsequent Auger processes.

Table 5.1: Probabilities (in percent) for electron shake-off from the various subshell, n_s , in xenon as a result of a sudden vacancy in orbital, n_0 , due to photoionisation of the $M_{4,5}$ subshell and several subsequent Auger processes calculated using the methodology proposed by Krause and Carlson [85]. For Auger processes, the new vacancies are denoted by the last two subshells involved. See main text for more details.

$n_s \backslash n_0$	M_5	$M_5\text{-}$ $N_1N_{2,3}$	$M_5\text{-}$ $N_1N_{4,5}$	$M_5\text{-}$ $N_{2,3}N_{2,3}$	$M_5\text{-}$ $N_{2,3}N_{4,5}$	$M_5\text{-}$ $N_{4,5}N_{4,5}$	$M_5\text{-}$ $N_{4,5}O_1$	$M_5\text{-}$ $N_{4,5}O_{2,3}$
M_1	0.012	0.016	0.016	0.012	0.012	0.012	0.012	0.012
M_2	0.020	0.020	0.016	0.020	0.020	0.016	0.016	0.084
M_3	0.040	0.040	0.024	0.040	0.040	0.032	0.032	0.032
M_4	0.080	0.056	0.080	0.056	0.080	0.080	0.088	0.088
M_5	0.060	0.070	0.050	0.070	0.050	0.050	0.050	0.050
N_1	0.176	0.088	0.096	0.024	0.028	0.032	0.100	0.084
N_2	0.252	0.120	0.036	0.112	0.128	0.044	0.108	0.124
N_3	0.511	0.240	0.064	0.216	0.256	0.072	0.200	0.248
N_4	1.249	0.080	0.112	0.088	0.128	0.040	0.407	0.447
N_5	1.679	0.108	0.096	0.108	0.096	0.108	0.419	0.479
O_1	1.808	0.814	0.758	0.817	0.758	0.698	0.260	0.144
O_2	3.579	1.705	1.567	1.630	1.579	1.436	0.399	0.407
O_3	8.031	3.892	3.614	3.737	3.652	3.373	1.011	1.067
Total	17.496	7.248	6.528	6.930	6.826	5.993	3.103	3.266

The Auger processes included in the table account for more than 98% of the branching ratios from the M_5 subshell.

5.2.2 Comparison with experiments

Table 5.2 compares the simulated results using four different options of choosing the propagating vacancy in BrIccEmis (see Section 3.4.4) with experimental data from recent measurements on Xe ions following the M_5 photoionisation [195, 196, 197]. In this short vacancy cascade, the simulations are rather independent of the propagating-vacancy option as there is no significant difference among all simulated distributions. In the simulations, it is assumed that the incident-photon energy is just above the binding energy of the M_5 subshell ($E_b = 674.7$ eV [124]), thus it is not enough to satisfy the sudden approximation limit (Eq. (3.16)). In general, all simulated results are in reasonable agreement with experiments. All simulated distributions correctly predict the peak at Xe^{4+} but fail to produce the right intensities for Xe^{5+} and Xe^{6+} ; simulated intensities of Xe^{5+} are always lower than the experimental values while there are fewer Xe^{6+} produced in the simulations than the measurements. Moreover, all simulations record negligible intensity of Xe^{7+} whereas experiments report that the species has around 2-4% of the total intensity.

The Hartree-Fock calculation by Kochur et al. [54] is in better agreement with the experiments; their calculated intensities of Xe^{5+} to Xe^{8+} are very close to the experimental

Table 5.2: Experimental, simulated and calculated intensities of different charge states of Xe ions following M_5 -subshell ionisation.

Charge state	Experiment			Theory				
	Exp.1 ^a	Exp.2 ^b	Exp.3 ^c	Random	Lifetime	Lowest	Highest	HF ^d
2			<0.003	<0.001	<0.001	<0.001	<0.001	
3	0.06(1) ^e	0.047(2)	0.042	0.044	0.043	0.045	0.045	0.050
4	0.42(2)	0.537(10)	0.39	0.464	0.470	0.467	0.461	0.460
5	0.31(2)	0.256(10)	0.32	0.382	0.393	0.403	0.386	0.318
6	0.17(1)	0.130(5)	0.20	0.109	0.093	0.085	0.108	0.150
7	0.04(1)	0.024(3)	0.041	0.001	0.000	0.000	0.000	0.021
8	0.01(1)	0.004(2)	0.004	0.000	0.000	0.000	0.000	0.001

^a Exp.1: Partanen et al. [196]

^b Exp.2: Eland et al. [197]

^c Exp.3: Tamenori et al. [195]

^d HF: Hartree-Fock calculation from Kochur et al. [54]

^e 0.06(1) means 0.06 ± 0.01

values apart from Xe^{5+} of Exp.2 [197] (see Table 5.2 and Figure 5.1).

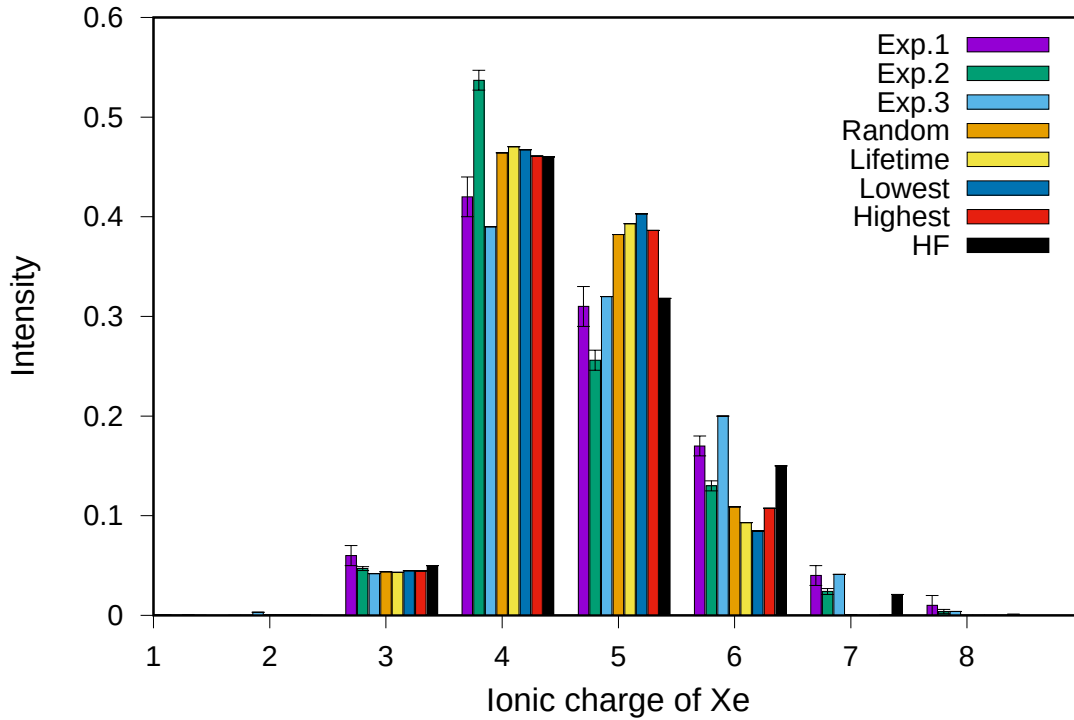


Figure 5.1: Comparison of charge-state distributions of xenon following photoionisation of M_5 subshell obtained from experiments (Exp.1 [196], Exp.2 [197] and Exp.3 [195]) with the simulated distributions based on four different vacancy-propagating options of BrIccEmis and a Hartree-Fock calculation (HF) from Kochur et al. [54]. The vertical lines in experimental bars indicate the errors.

Table 5.3 compares the branching ratios from EADL and Kochur et al. [54] for subshells

involved in the vacancy cascade following M_5 ionisation. EADL has higher Auger yield than Kochur et al. for an N_1 vacancy while the latter has zero intensity for $N_{2,3}N_{2,3}O_{2,3}$ and $N_{2,3}N_{4,5}N_{4,5}$ transitions compared to 93.92% for those transitions from EADL. This discrepancy is likely due to the fact that EADL calculated the transition rates using the so-called “*frozen-orbital*” approximation. However, if both the aforementioned transitions are removed from the list of possible final states, both EADL and Kochur et al. are in good agreement for the branching ratios following $N_{2,3}$ ionisation. For $N_{4,5}$ subshells, HF values from Kochur et al. tend to favour slightly the $O_{2,3}O_{2,3}$ final states over EADL data.

Table 5.3: Comparison of branching ratios (in percent) from EADL[27] and Hartree-Fock calculation of Kochur et al. [54] for subshells that involve in the vacancy cascade following M_5 ionisation.

Final vacancies	Initial vacancies								
	EADL					Kochur et al. (HF)			
	$M_{4,5}$	N_1	$N_{2,3}$	$N_{2,3}^a$	$N_{4,5}$	$M_{4,5}$	N_1	$N_{2,3}$	$N_{4,5}$
$M_{4,5}O_{2,3}$	0.85								
N_1N_1	0.01					0.01			
$N_1N_{2,3}$	1.11					0.88			
$N_1N_{4,5}$	13.0					10.91			
N_1O_1	0					0			
$N_1O_{2,3}$	0.08					0.07			
$N_{2,3}N_{2,3}$	2.81					2.75			
$N_{2,3}N_{4,5}$	30.48					29.31			
$N_{2,3}O_1$	0.09	20.8				0.1	14.89		
$N_{2,3}O_{2,3}$	0.41	42.3	0.39			0.51	54.46		
$N_{4,5}N_{4,5}$	46.48	12.63	93.53			50.19	11.52		
$N_{4,5}O_1$	1.55	7.92	1.72	28.34		1.47	9.24	27.36	
$N_{4,5}O_{2,3}$	2.91	6.14	4.2	69.14		3.76	8.61	68.91	
O_1O_1		1.4	0	0.02	34.66		0.22	0.06	29.35
$O_1O_{2,3}$	0.01	4.05	0.06	1.06	14.6	0.01	0.82	1.19	10.85
$O_{2,3}O_{2,3}$	0.03	4.74	0.09	1.44	50.75	0.02	0.23	2.47	59.78
CK yield	0.85	89.79	99.84	97.48	0	0	98.73	96.27	0
Auger yield	98.97	10.19	0.15	2.52	100.01	99.96	1.27	3.72	99.98

^aDistribution without consideration of $N_{2,3}O_{2,3}$ and $N_{4,5}N_{4,5}$ final states.

Since the Hartree-Fock (HF) calculation of Kochur et al. [54] achieved a better agreement with experiments, one should check if the disagreement between the experiments and simulated results from BrIccEmis is due to the input relative transition rates from EADL [27]. Since Kochur et al. did not distinguish the distributions of final vacancies for initial holes in subshells with the same orbital angular momentum, it is assumed that they are the same. Moreover, if a branching ratio is assigned to a group with multiple final states, for example, the $N_{4,5}O_{2,3}$ group, which has four different final states according to the jj coupling scheme, it is assumed that the reported branching ratio is distributed to

all final states equally.

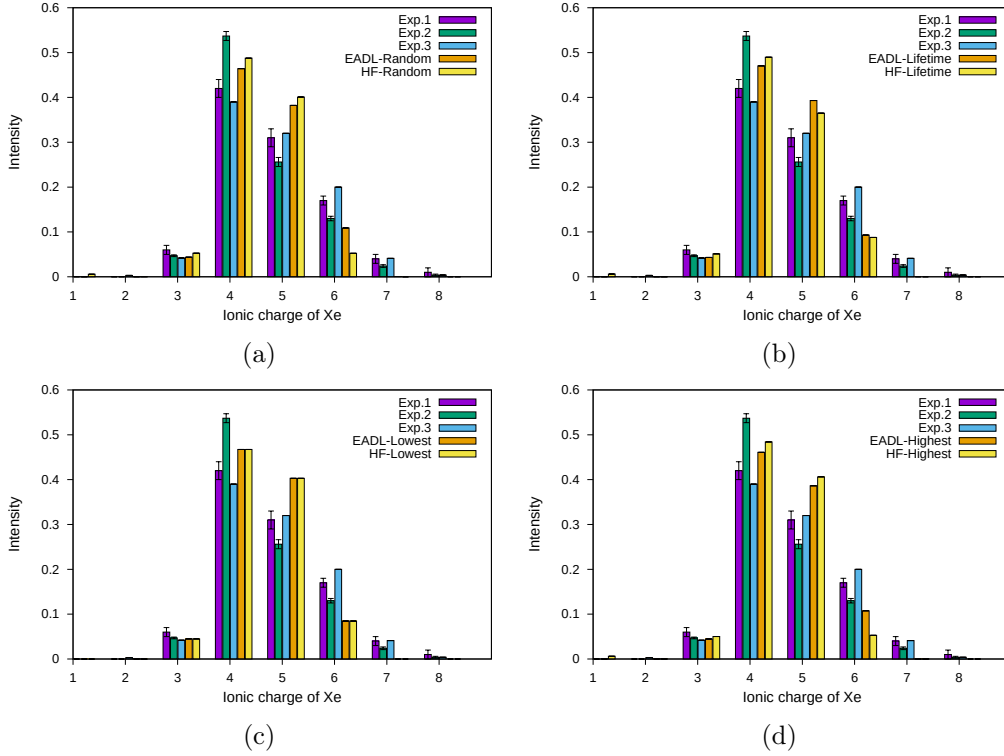


Figure 5.2: Comparison of charge-state distributions of xenon following the photoionisation of M_5 subshell obtained from experiments with the simulated distributions based on the four different vacancy-propagation options of **BrIccEmis**, i.e. (a) random, (b) lifetime, (c) lowest, and (d) highest, using the relative transition rates from EADL [27] and Kochur et al. (HF) [54]. The vertical lines in experimental bars indicate the errors.

Figure 5.2 compares the simulated results from **BrIccEmis** using both the EADL and HF relative transition rates with experiments as well as calculation from Kochur et al. (labelled as HF). In general, the simulated charge-state distributions do not change drastically. For the lowest-vacancy (or deepest-hole) approximation, both input data yield exactly the same residual charge-state distribution. For other approximations, there is a redistribution of intensities towards the lower charge states. It is important to note that there is still a petty amount of Xe^{7+} generated from the simulations. It seems that the simulation always hits a *bottleneck* at Xe^{6+} and the model does not permit further non-radiative transitions to be released.

Contribution of shake-off electrons

All results presented in Table 5.2 are derived from simulations assuming the incident-photon energy is very close to the binding energy of the M_5 subshell, therefore the shake-off accompanying the photoelectron is not considered because the sudden-approximation limit is not satisfied (see Eq. (3.16)). However, Table 5.1 shows that there is a 6-7% probability for a $M_5\text{NN}$ Auger transition to shake an orbital electron off and this Auger transition group

is the dominant channel for an M_5 initial vacancy (see Table 5.3). Table 5.4 compares the contribution from electrons of three different origins, i.e. photoelectrons, Auger and CK electrons, and shake-off electrons, to the simulated charge-state distributions presented in Table 5.2. Although shake-off electrons only contribute, at most, 1.3% to the total number of ejected electrons (or mean charge of Xe ions) following the creation of the M_5 vacancy, the intensity of an individual charge state could change by up to 3%.

Table 5.4: Breakdown of the total number of emitted electrons in the vacancy cascade following the creation of an M_5 vacancy into electrons of three different origins for four different vacancy-propagation options of BrIccEmis.

	Random	Lifetime weighted	Lowest vacancy	Highest vacancy
Photoelectrons	1.00	1.00	1.00	1.00
Auger and CK electrons	3.50	3.48	3.47	3.50
Shake-off electrons	0.06	0.05	0.06	0.06
Total	4.56	4.53	4.53	4.56

Figure 5.3 compares the charge-state distributions simulated with and without shake-off processes. Exclusion of shake-off from the vacancy cascade leads to higher intensity of Xe^{4+} but lower intensities of Xe^{5+} and Xe^{6+} . The intensity of Xe^{3+} is rather independent of shake-off due to smaller shake-off probability for $M_5N_{4,5}O_1$ and $M_5N_{4,5}O_1$ Auger transitions, since the formation of Xe^{3+} ions is mainly through the chain of $M_5N_{4,5}O_{1,2,3}$ and $N_{4,5}OO$ transitions (see Tables 5.1 and 5.3).

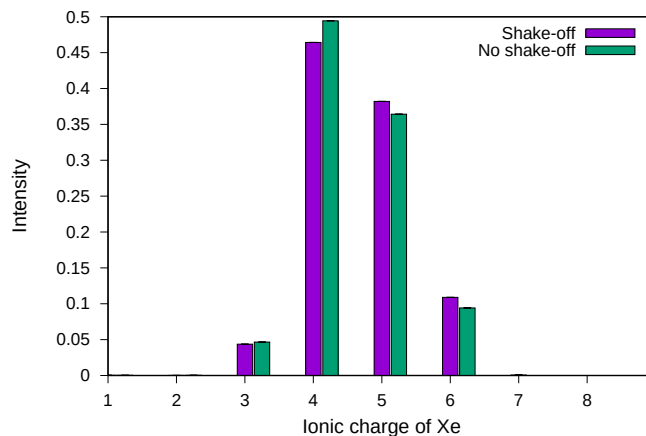


Figure 5.3: Comparison of charge-state distributions of xenon simulated with and without shake-off processes following the photoionisation of the M_5 subshell. Only the simulation based on the random-vacancy approximation is shown as other approximations produce almost the same distribution.

5.3 $^{131\text{m}}\text{Xe}$ IT

The measurement of the charge-state distribution following the isomeric decay of $^{131\text{m}}\text{Xe}$ ($T_{1/2} = 11.84$ d) from Pleasonton and Snell [189] is one of the very few experimental data in the literature that studies the vacancy cascade initiated by nuclear decay; in this case, the vacancy cascade is started by the internal conversion of an $M4$ γ -transition from the isomeric state of ^{131}Xe ($E_\gamma = 163.930$ keV). The experimental data have also been used by Mukoyama [53] and Pomplun [87] for benchmarking their vacancy-cascade models.

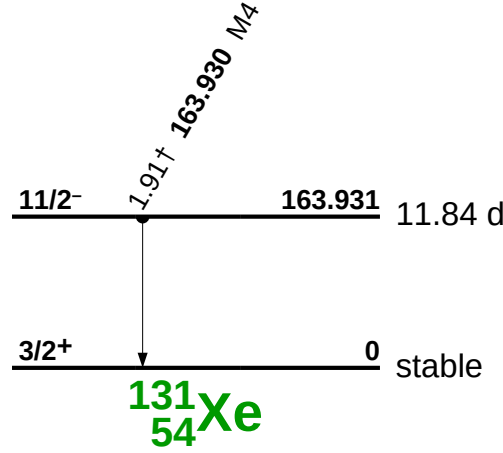


Figure 5.4: Decay scheme of $^{131\text{m}}\text{Xe}$. Extracted from Firestone et al. [99].

Table 5.5: Initial vacancy distribution calculated for the $M4$ γ -transition ($E_\gamma = 163.930$ keV) in $^{131\text{m}}\text{Xe}$.

Atomic Subshell	Vacancies per decay	Atomic Subshell	Vacancy per decay
K	0.616	N_1	0.006
L_1	0.139	N_2	0.001
L_2	0.032	N_3	0.005
L_3	0.116	N_4	<0.001
M_1	0.031	N_5	<0.001
M_2	0.007	O_1	0.001
M_3	0.028	O_2	<0.001
M_4	<0.001	O_3	<0.001
M_5	<0.001		

Figure 5.4 shows the decay scheme of $^{131\text{m}}\text{Xe}$ and Table 5.5 displays the calculated initial-vacancy distribution of $^{131\text{m}}\text{Xe}$ decay using BrIcc [18]. Figure 5.5 compares the residual charge-state distributions of xenon following the isomeric decay of $^{131\text{m}}\text{Xe}$ with the experimental data from Pleasonton and Snell [189]. Four different options of selecting the propagating vacancy described in Section 3.4.4 have been considered. All simulated distributions are able to reproduce the intensities of several highly ionised states (i.e. Xe^{12+}

to Xe^{15+}) and achieve a fair agreement with the experiment in terms of the width of the distribution. Nevertheless, discrepancies can be seen for several charge states, especially those around the most intense ionic charge. The strongest charge state in the simulations based on the “randomly-selected” and “highest-vacancy” approximations is calculated to be $7+$ compared to $8+$ in the experiment. However, both the lifetime-weighted and lowest-vacancy approximations produce $6+$ as the peak of the distribution. All simulated distributions have a significantly lower intensity for Xe^{3+} ions compared to the experiment. Meanwhile simulations from Mukoyama [153] and Pomplun [87] agree well with the experiment for the intensity of Xe^{3+} ions. Mukoyama did not use the strict-energy restriction like `BrIccEmis` in eliminating energetically-forbidden transitions and neglected the internal conversion process from the M shell and above, so the agreement between Mukoyama and the experiment could be fortuitous. Pomplun adopted a Dirac-Fock method from Desciaux [111] to calculate transition energies during the vacancy cascade which is similar to `BrIccEmis`, but his work used the radiative transition probabilities from Storm [109] while `BrIccEmis` adopted the values from EADL [27]. One of the main channels to produce Xe^{3+} ions is through two consecutive radiative transitions after a K-shell internal conversion and two further non-radiative transitions when the initial vacancy has been transported to higher subshells, which means the discrepancy between `BrIccEmis` and Pomplun could be due to the differences in radiative transition probabilities. With that said, Xe^{3+} ions could also be produced via an M-shell internal conversion followed by two non-radiative transitions. It is important to note that Pia et al. [133] recently claimed that EADL [27] does not agree well with experimental data for some K and L-shell radiative transitions and they concluded that probabilities calculated using the Hartree-Fock approach are better.

Unlike the charge-state distribution resulting from the M_5 -shell ionisation (see Figure 5.1), the simulated distribution varies significantly from one vacancy-propagation assumption to another for the isomeric decay of ^{131m}Xe . Overall, the lifetime-weighted assumption is similar to the lowest-vacancy approximation, as it turns out that the vacancy with largest subshell width also tends to be the lowest during the vacancy cascade in xenon. Both the random-vacancy and highest-vacancy approximations yield similar distributions, although the highest-vacancy approximation appears to give the best agreement with the experiment. Table 5.7 details the values used to plot Figure 5.5. It can be seen from the table that the lowest-vacancy approximation yields the worst agreement with the experiment as many outer-shell non-radiative transitions, XYZ, simply become energetically forbidden by the time the vacancy in subshell X is picked to propagate. This is in stark contrast to Pomplun’s model [87] in which the lowest-vacancy approximation gives the best agreement with the same experimental data. An interesting observation is that the lifetime-weighted assumption gives the highest intensity among all approximations for Xe^{14+} onwards, even though it causes the peak of the distribution to shift towards lower charges. In addition, it is worth noting that the very high-charge states, though weak in experimental intensities,

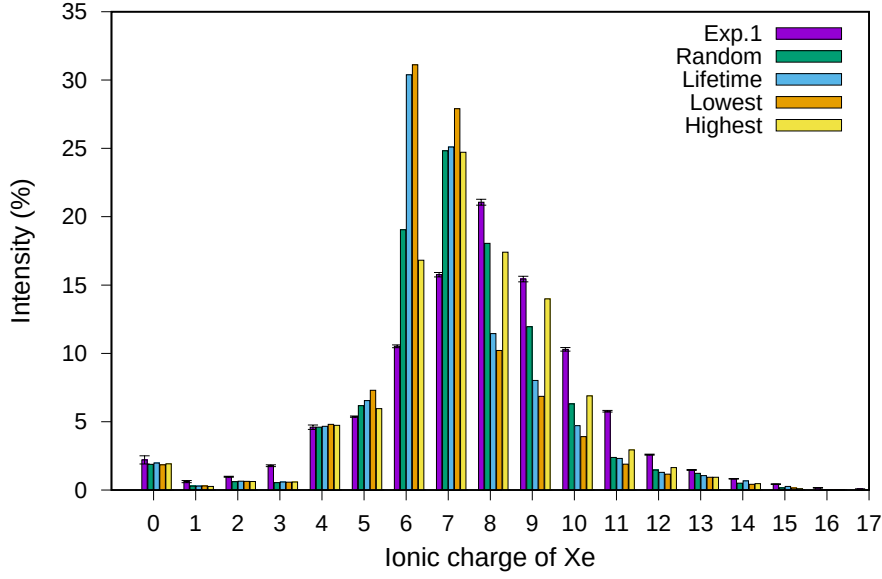


Figure 5.5: Comparison of simulated charge-state distributions of xenon following the isomeric decay of $^{131\text{m}}\text{Xe}$ based on four different vacancy-propagating options of **BrIccEmis** with experimental data from Pleasonton and Snell [189] (Exp.1). The vertical lines in experimental bars indicate the errors.

are missing completely from the simulation. This is similar to the case of M_5 -subshell ionisation where the intensity of Xe^{7+} could not be reproduced in the simulation.

Contribution of shake-off electrons

Table 5.7 compares the contribution from electrons of three different origins, i.e. conversion electrons, Auger and CK electrons, and shake-off electrons, to the simulated charge-state distributions presented in Table 5.6. In the isomeric decay of $^{131\text{m}}\text{Xe}$, most shake-off electrons are emitted together with conversion electrons. Since the nuclear transition energy ($E_\gamma = 163.930$ keV) is much higher than the orbital binding energies of xenon, which is to say that the sudden approximation is always valid regardless of which subshell is ionised, the total number of shake-off electrons ejected with conversion electrons can be easily calculated using the initial vacancy distribution in Table 5.5 and subshell shake-off probabilities from Carlson and Nestor [14]. This gives approximately 0.19 shake-off electrons being ejected together with conversion electrons per isomeric decay of $^{131\text{m}}\text{Xe}$. Other shake-off electrons reported in Table 5.6 are due to non-radiative transitions, especially Auger transitions which have much higher energies than Coster-Kronig transitions, as the shake-off process accompanying radiative transitions (i.e. Radiative Auger effect) are negligible and it is neglected in **BrIccEmis**.

It is important to note that the probability of shake-off that accompanies Auger transitions could be underestimated by a few percent for the first few step of the vacancy cascade using the approximation described in Section 3.4.5, but the error is unlikely to significantly

Table 5.6: Experimental and simulated residual charge-state distributions of Xe ions following the decay of ^{131m}Xe .

Charge state	Experiment	Calculation			
	Exp.1 ^a	Random	Lifetime	Lowest	Highest
0	2.20±0.30	1.88	1.98	1.85	1.92
1	0.62±0.08	0.29	0.29	0.30	0.26
2	0.97±0.04	0.61	0.65	0.64	0.62
3	1.78±0.07	0.54	0.59	0.57	0.59
4	4.60±0.16	4.60	4.66	4.81	4.74
5	5.37±0.05	6.17	6.54	7.30	5.96
6	10.52±0.10	19.05	30.39	31.12	16.82
7	15.76±0.16	24.82	25.10	27.90	24.71
8	21.06±0.21	18.05	11.45	10.21	17.41
9	15.44±0.20	11.95	8.02	6.86	13.99
10	10.30±0.13	6.30	4.71	3.91	6.89
11	5.77±0.06	2.38	2.31	1.89	2.94
12	2.59±0.04	1.46	1.30	1.16	1.64
13	1.47±0.02	1.22	1.06	0.93	0.94
14	0.82±0.01	0.50	0.67	0.42	0.47
15	0.43±0.01	0.16	0.26	0.14	0.10
16	0.165±0.004	0.02	0.04	0.01	0.02
17	0.071±0.004	0.00	0.00	0.00	0.00
18	0.02±0.002	0.00	0.00	0.00	0.00
19	0.015±0.002	0.00	0.00	0.00	0.00
20	0.003±0.002	0.00	0.00	0.00	0.00
21	0.010±0.002	0.00	0.00	0.00	0.00
22	-0.0016±0.0023	0.00	0.00	0.00	0.00
Mean charge	7.73±0.05	7.25	6.92	6.78	7.33

^a Exp.1: Pleasonton and Snell [189]Table 5.7: Similar to Table 5.4 but for vacancy cascade following the isomeric decay of ^{131m}Xe .

	Random	Lifetime weighted	Lowest vacancy	Highest vacancy
Conversion electrons	0.98	0.98	0.98	0.98
Auger and CK electrons	5.96	5.64	5.48	6.07
Shake-off electrons	0.31	0.30	0.32	0.28
Total	7.25	6.92	6.78	7.33

change the distribution. However, the inclusion of shake-off electrons is crucial in achieving a reasonable agreement with experiment. Figure 5.6 compares the charge-state distributions simulated with and without shake-off. The exclusion of shake-off electrons leads to lower mean charge and shifts the distribution towards lower charges, which is moving away from the experimentally observed distribution.

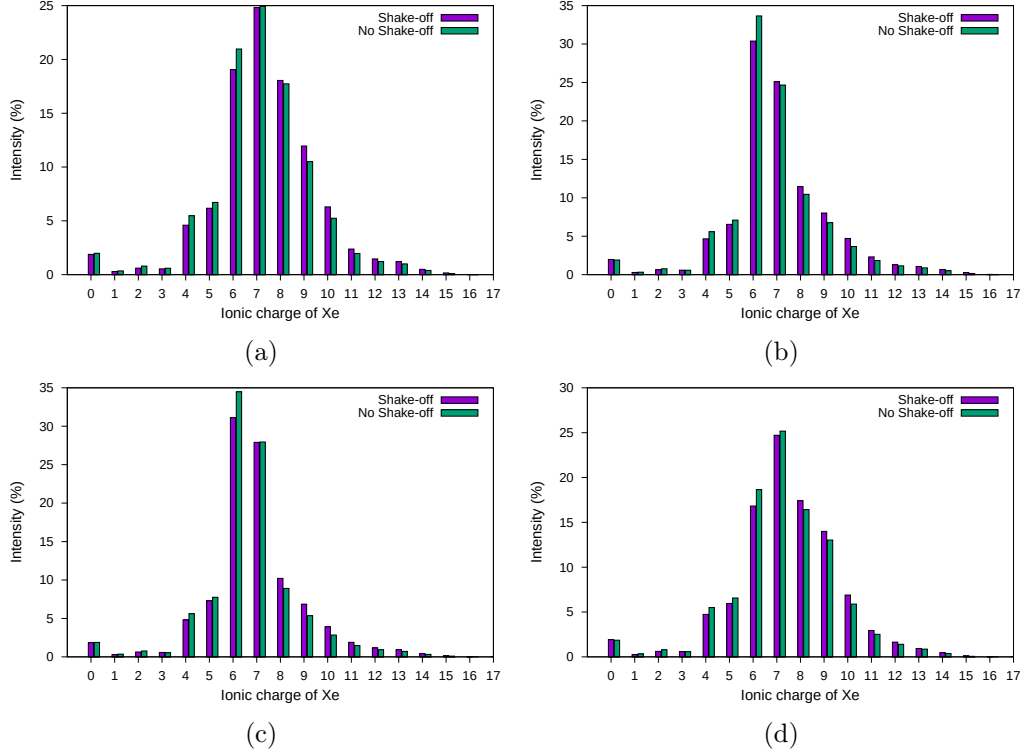


Figure 5.6: Comparison of simulated charge-state distributions of xenon with and without shake-off processes for the decay of $^{131\text{m}}\text{Xe}$. Four different vacancy-propagation options of **BrIccEmis** have been considered: (a) random, (b) lifetime, (c) lowest, and (d) highest.

5.4 Alternative input data

A recent review of K- and L-shell radiative transition probabilities by Pia et al. [133] has indicated that EADL [27] may not represent the state-of-the-art and further work could be done to improve the accuracy of the library. Chen, who is one of the coauthors of the EADL library, has stated in his paper with Crasemann and Mark [134] that transition probabilities calculated within the physics framework used for EADL are inaccurate for atomic numbers $Z \leq 60$. A collaboration with the development team of the **Grasp2K** code at the Malmö University, which is led by Prof. Per Jönsson and Dr. Jörgen Ekman, and the ANU group developing **BrIccEmis**, has been formed to explore the possibility of providing updated atomic transition probabilities based on the state-of-the-art atomic structure packages. Wave functions describing the initial and final states of each atomic transition for xenon in a singly-ionised system are calculated using the **Grasp2K** package based on a Dirac-Fock (DF) approach. Transition energies are given by the energy difference of the independently-optimised initial and final states. Radiative and non-radiative transition rates are computed using the **Ratip** package. The latter are calculated based on the frozen-orbital approximation, i.e. a common orthonormal basis with one-electron Dirac orbitals describing the initial state, in order to ensure orthogonality. A more detailed description of the computation can be found in Appendix A.6. Radiative and non-radiative rates are

normalised for each subshell and the relative probabilities are then used in BrIccEmis for simulation of the vacancy cascade.

5.4.1 $^{131\text{m}}\text{Xe}$

Figure 5.7 compares the simulated charge-state distributions following the isomeric decay of $^{131\text{m}}\text{Xe}$ with the experimental data from Pleasonton and Snell [189]. Although the simulation still could not reproduce the peak at Xe^{8+} , it is now in better agreement with the experiment. Apart from the lowest-vacancy approximations, the simulated mean charge of xenon ions is in reasonable agreement with the experimental value (see Table 5.8). Although the highest-vacancy approximation gives the best agreement with the experiment in terms of the mean charge of xenon ions, it also produces a trough at Xe^{8+} where the experimental peak is located. Similar to the EADL-based simulation, the lifetime-weighted approximation agrees well with the experiment for highly-ionised states, i.e. Xe^{10+} onwards, even though it gives the peak of the distribution at Xe^{6+} . Both the EADL-based and DF-based simulations are unable to reproduce the intensity of Xe^{3+} .

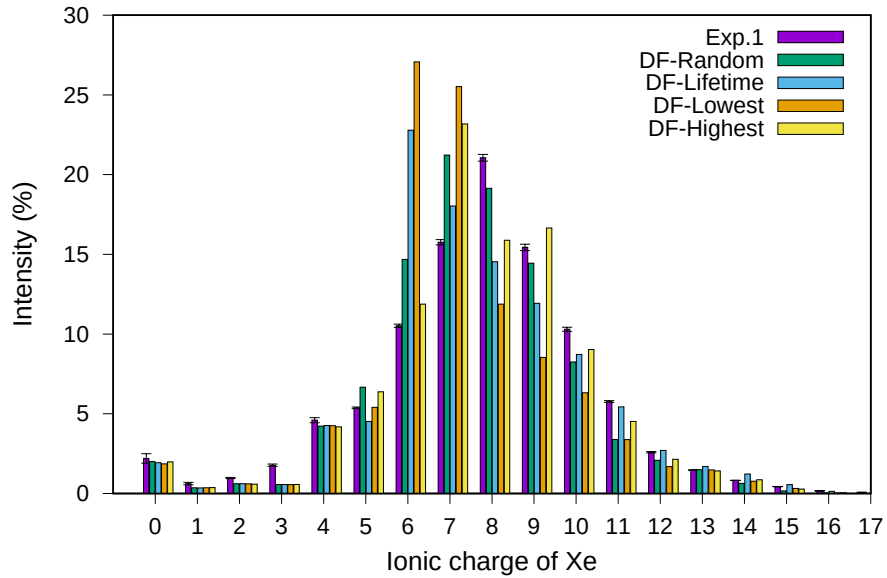


Figure 5.7: Similar to Figure 5.5 but the transition probabilities from EADL have been replaced by values calculated using the Dirac-Fock (DF) method.

The increase in the mean charge and the shift of the distribution towards higher charges are mainly due to the increase in the yield of low-energy Coster-Kronig (CK) transitions. Figure 5.8 shows the difference in yield of different atomic transition groups between EADL-based and DF-based simulations; only values derived from the random-vacancy approximation are presented but the same can be applied to other approximations. Apart from the NXY Auger group, the yield of other Auger groups is similar for both EADL-based and DF-based simulations. The decrease in the yield of the NXY Auger group is a re-

Table 5.8: Similar to Table 5.6 but the input transition rates for the simulation are calculated based on the Dirac-Fock (DF) method.

Charge state	Experiment	Calculation-DF			
	Exp.1 ^a	Random	Lifetime	Lowest	Highest
0	2.20±0.30	2.00	1.93	1.86	1.98
1	0.62±0.08	0.36	0.35	0.36	0.37
2	0.97±0.04	0.62	0.62	0.61	0.59
3	1.78±0.07	0.56	0.57	0.55	0.57
4	4.60±0.16	4.22	4.26	4.25	4.18
5	5.37±0.05	6.67	4.52	5.40	6.38
6	10.52±0.10	14.68	22.79	27.07	11.88
7	15.76±0.16	21.23	18.03	25.52	23.18
8	21.06±0.21	19.14	14.54	11.88	15.88
9	15.44±0.20	14.45	11.93	8.54	16.65
10	10.30±0.13	8.25	8.72	6.32	9.03
11	5.77±0.06	3.39	5.43	3.38	4.52
12	2.59±0.04	2.09	2.70	1.68	2.15
13	1.47±0.02	1.49	1.70	1.48	1.42
14	0.82±0.01	0.64	1.22	0.77	0.86
15	0.43±0.01	0.16	0.55	0.32	0.28
16	0.165±0.004	0.04	0.13	0.03	0.05
17	0.071±0.004	0.00	0.02	0.00	0.01
18	0.02±0.002	0.00	0.00	0.00	0.00
19	0.015±0.002	0.00	0.00	0.00	0.00
20	0.003±0.002	0.00	0.00	0.00	0.00
21	0.010±0.002	0.00	0.00	0.00	0.00
22	-0.0016±0.0023	0.00	0.00	0.00	0.00
Mean charge	7.73±0.05	7.50	7.59	7.16	7.64

^a Exp.1: Pleasonton and Snell [189]

sult of the enhancement in the yield of the NNX CK group. Additionally, O-shell X-rays are neglected in EADL thus the DF-based simulation produces significantly more of these low-energy X-rays per decay. It is important to note that most of the low-energy X-rays proceed very slowly ($\geq 10^{-10}$ s)

Figure 5.9 shows that the energies calculated using the DF method are quite different for some CK transitions while the Auger energies are rather similar for the two approaches. This is the main reason behind the increase in CK relative probabilities as the CK rates are extremely sensitive to the transition energy [199].

5.4.2 Xe M₅ photoionisation

Figure 5.10 compares the simulated charge-state distributions with data from several experiments. Due to the increase in the yield of the CK NNX group, DF-based simulations shift the peak of distribution from Xe⁴⁺ (see Figure 5.1) to Xe⁵⁺ and produces a worse

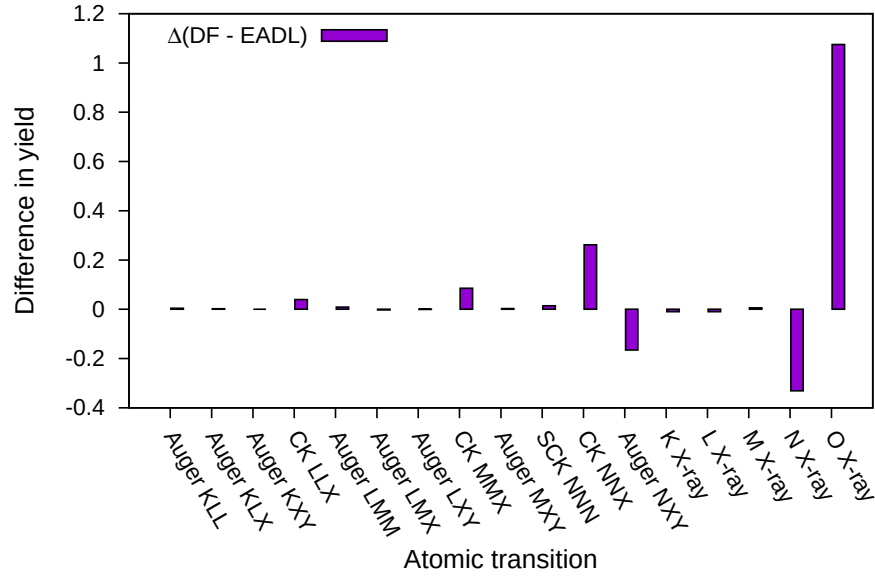


Figure 5.8: Difference in yield of atomic transitions following the isomeric decay of $^{131\text{m}}\text{Xe}$ between EADL-based and DF-based simulations.

agreement with the experiments than EADL-based simulations. This seems to suggest that the rates of CK transitions calculated using the frozen-orbital approximation may be overestimated. It is important to note that EADL transition rates are also calculated using the same approximation but the DF-based CK transition energies are generally lower and this could change the CK rates significantly. In the case of xenon, lower transition energy causes enhancement of the rate for CK transitions.

5.5 Effect of inaccurate calculation of transition energies

Although the **RAINE** code [112] is able to calculate the electron binding energies for any given electron configurations, it is unfortunately not perfect. Section 3.3 has detailed the flaw where calculated neutral binding energies are almost always higher than the experimental values (see Figure 3.3). Inaccurate calculation of electron binding energies results in the overestimation (K and L shells) or underestimation (M shell and above) of Auger and Coster-Kronig (CK) transition energies (see Section 4.2). This could significantly affect the residual charge-state distribution as many M-shell and N-shell CK transitions could have near-zero energies in a highly-ionised system. If the **RAINE** code underestimates a near-threshold non-radiative transition by several eV, then **BrIccEmis** will treat the transition as energetically forbidden when it actually has positive energy, thus the simulation will generate a charge-state distribution with a smaller mean than it should be. In fact, it has been demonstrated in Figure 4.7b that **RAINE** tends to underestimate the energy of low-energy N-shell non-radiative transitions in xenon. Figure 5.11 compares with experiments the results of an EADL-based simulation assuming that all transitions are underestimated

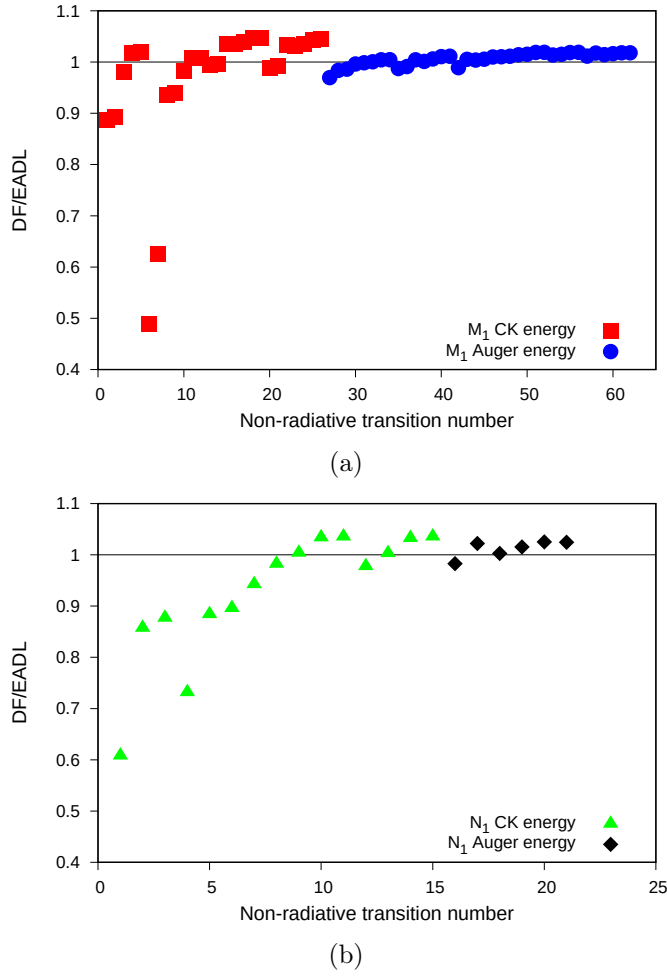


Figure 5.9: Ratios of non-radiative transition energies calculated using the DF method and extracted from EADL for (a) M₁ and (b) N₁ subshells.

by 2 eV, this comparison should only be taken as a qualitative analysis since the degree of overestimation or underestimation of energies by the **RAINE** code varies for different transitions. The comparison shows that the simulations are now in better agreement with experiments compared to the simulations not assuming the transition energies are underestimated. Therefore, this qualitative comparison suggests that improving the accuracy of energy calculation could be an important step in order to achieve better agreement with experiments.

5.6 Discussion

Several assumptions on the propagating of atomic vacancies have been considered, i.e. random, lowest, highest and lifetime-weighted, but the simulated charge-state distribution of xenon following photoionisation of M₅ is rather independent of the selection of the propagating vacancy, which is probably due to the fact that the cascade that follows the creation of an M₅ vacancy is short (see Figure 5.1). Nevertheless, the difference between

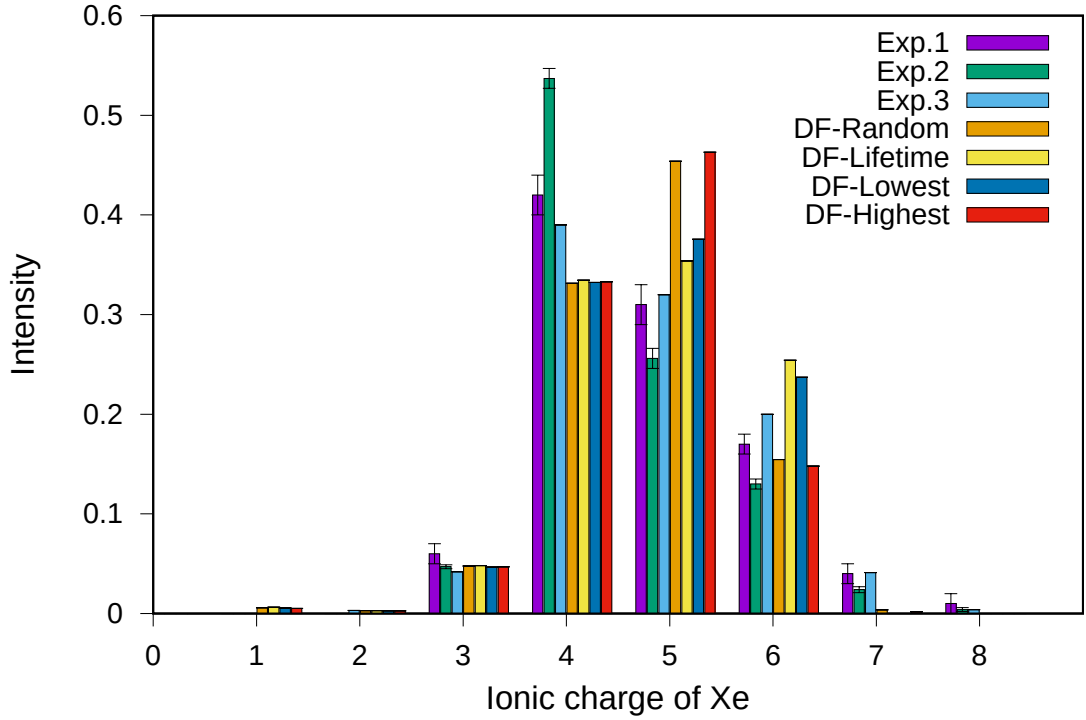


Figure 5.10: Similar to Figure 5.1 but the transition probabilities calculated using the Dirac-Fock (DF) method are used as the input instead of EADL probabilities. The distribution calculated using the Hartree-Fock method (HF) [54] has been excluded.

each assumption is demonstrated in the simulated charge distributions following the isomeric decay of $^{131\text{m}}\text{Xe}$ (see Figure 5.5). Both the random-vacancy and highest-vacancy approximations yield better agreement with experiment than the other two assumptions in the current version of **BrIccEmis**. However, the latter yields a strange trough at Xe^{8+} when the transition probabilities from EADL are replaced with values derived from a Dirac-Fock method; this seems to indicate that the highest-vacancy approximation maybe too extreme. Meanwhile the lowest-vacancy approximation yields the worst agreement with experimental data which is in stark contrast to Pomplun’s model [87]. The latter achieves the best agreement with the same experimental data from Pleasonton and Snell [189] using the lowest-vacancy approximation and it almost reproduces the experimentally observed distribution perfectly. This could be due to overestimation in the yield of shake-off electrons. Although Pomplun did not specify the calculated number of shake-off electrons for the decay of $^{131\text{m}}\text{Xe}$, the same author reported a huge 0.9 shake-off electrons from the electron capture decay of ^{123}I in a later publication [88]. Since the shake-off accompanying electron capture is usually negligible (see Section 3.2.3), only Auger and Coster-Kronig (CK) transitions, as well as the internal conversion process, could shake off outer-shell electrons at a significant probability. Based on the decay spectrum of ^{123}I from Pomplun (see Table 4.1), one could provide a crude estimate of the yield of shake-off electrons. According to the spectrum of ^{123}I decay from Pomplun’s model, there are 0.15

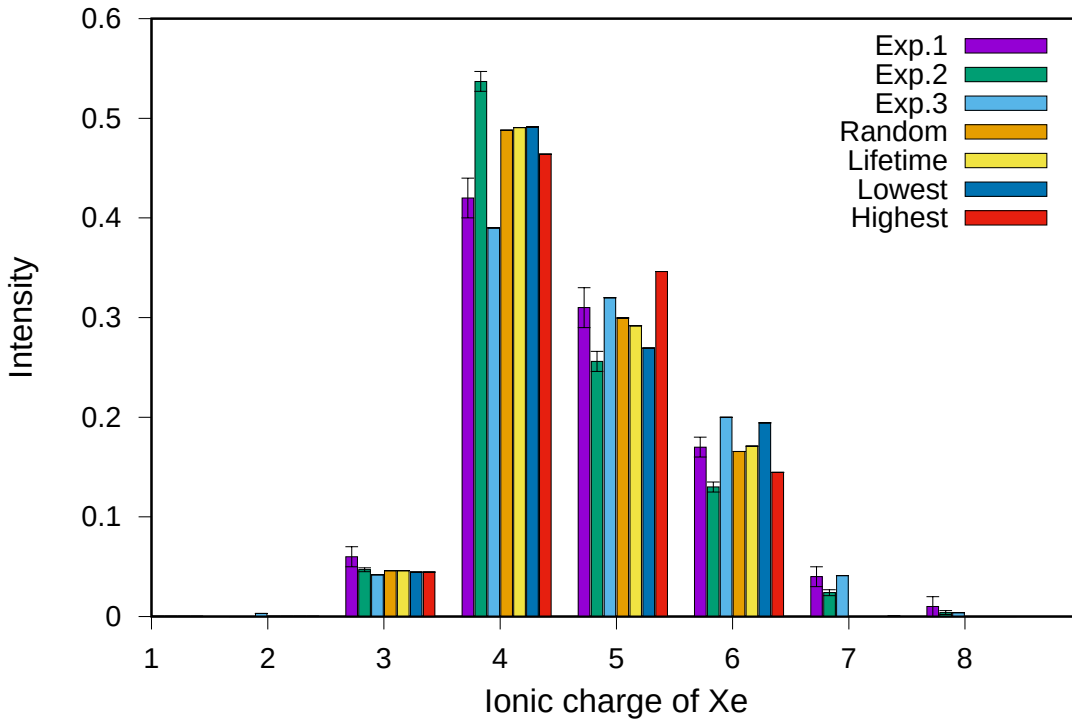


Figure 5.11: Similar to Figure 5.1 but the energy threshold has been lowered from 0 to -2 eV. The distribution calculated using the Hartree-Fock method (HF) [54] has been excluded.

conversion electrons, 0.11 K-shell Auger electrons, 1.06 L-shell Auger and CK electrons, and 2.80 M-shell Auger and CK electrons; although the yield of N-shell Auger and CK electrons is significant (2.40), they are too low in energy to allow the shake-off process and the shake-off probability diminishes with increasing charges. Therefore, the shake-off yield in the ^{123}I decay is estimated to be 0.29 (see Table 5.9) which is significantly smaller than the value published by Pomplun [87]. It is worth noting that Pomplun's model reproduced the peak of the distribution at Xe^{8+} when shake-off was excluded. This suggests that the Dirac-Fock code from Desclaux [111], which is adopted by Pomplun, could be better in estimating the transition energies in highly-ionised systems. Overall, the discrepancy between charge-state distributions simulated using BrIccEmis and Pomplun's model is unlikely to be caused by the different initial vacancy distributions applied¹.

If the propagating vacancy is chosen based on its lifetime, the simulation is able to retain the high intensity of highly-ionised states. This observation suggests that this assumption could be the best of all options if the accuracy of the energy calculation could be improved in the future. In fact, it should be the best option when all other ingredients of the vacancy-cascade simulation are improved. Given that the random-vacancy approximation

¹Pomplun used decay data from International Commission on Radiological Protection (ICRP) Publication 38 [114] which has been superseded by ICRP Publication 107 [200]. The author only has access to the latter and it gives similar initial vacancy distribution to the one shown in Table 5.5.

Table 5.9: Estimation of shake-off yield for the decay of ^{123}I based on the decay spectrum from Pomplun’s model [87].

Transition	Yield	Approximate	Estimated
		shake-off probability ^a	shake-off yield
K-shell conversion	0.13	0.20	0.03
L-shell conversion	0.02	0.18	<0.01
K-shell Auger	0.11	0.10	0.01
L-shell Auger and CK	1.06	0.08	0.08
M-shell Auger and CK	2.80	0.06	0.17
Total			0.29

^aShake-off probability is estimated based on values published for xenon [14] and the approximation for shake-off accompanying non-radiative transitions (see Section 3.4.5), and the effect of spectator vacancies is neglected.

gives results that are in between the two extremes, i.e. lowest-vacancy and highest-vacancy approximations, and that it also provides a better agreement with experiments than the lifetime-weighted assumption within the current setting of **BrIccEmis**, it is set as the default option for simulating the vacancy cascade and will be used to generate the energy spectrum or charge-state distribution in the following chapters, unless specified otherwise.

All experimental charge-state distributions following M_5 photoionisation that have been used for comparisons in this chapter were detected at slightly different incident-photon energies. Eland et al. [197] used the incident-photon energy of 710 eV while Partanen et al. [196] used photon energies in the range of 664-694 eV. Since the neutral binding energy of the M_5 subshell of xenon is 676 eV [124], the energy of photoelectrons ejected from the M_5 subshell in these experiments would be ~ 46 eV. Shake-off probability calculated using the shake theory from Carlson and Nestor [14] is only valid when the sudden-approximation limit is satisfied. The sudden-approximation limit requires that the energy of the photoelectron to be at least 3 times the threshold energy of the shake-off electron. Since the theoretical binding energy of the O_3 subshell in Xe^{1+} ions is 22 eV, the minimum photoelectron energy needed for the sudden-approximation limit to be valid is 742 eV. It is important to note that the shake-off electron is most likely to come from the outermost subshell, which is the O_3 shell in the case of Xe (see Table 5.1). Therefore, the shake theory of Carlson and Nestor is not applicable for the measurement reported by Partanen et al. [196] and Eland et al. [197]. The shake-off probability was found to be smaller when the sudden-approximation limit is not met [35], therefore it is expected that the emission of shake-off electrons will be highly suppressed in the experiment. The distribution of Xe ions following M_5 ionisation in Tamenori et al. [195] was measured with photons of higher energy at 800 eV. In this case, the photoelectron energy is about 124 eV and there is a finite probability for the shake-off process (see Table 5.1).

Figure 5.12 compares the simulated charge-state distributions for three different incident-

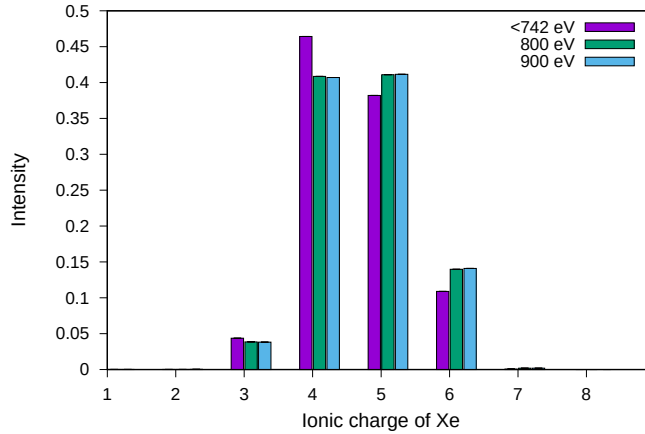


Figure 5.12: Comparison of simulated charge-state distributions following M_5 ionisation for three different incident-photon energies. The minimum energy of the incident photon required to satisfy the sudden-approximation limit, i.e. Eq. (3.16), is 742 eV (see main text for more details). Only the simulation based on the random-vacancy approximation is shown as other approximations produce almost the same distribution.

photon energies at < 742 , 800 and 900 eV. In general, the distribution calculated for incident-photon energies of < 742 eV have lower mean charge than the distributions derived for the other two energies due to the lack of shake-off electrons. The simulated charge-state distributions for incident-photon energies of 800 and 900 eV are very similar, because the shake-off probability is constant once the sudden-approximation limit is met. Therefore, an apparent shift in charge-state distribution towards higher charges observed by Tamenori et al. [195] compared to other experiments, could be explained theoretically by the emission of shake-off electrons. Nevertheless, the shift could also be due to experimental artefacts.

The residual charge-state distributions reported in Refs. [196, 197] were measured following threshold photoionisation. Therefore, the distributions could be subject to the post-collision interaction (PCI) effect [174, 175, 176, 177]. In the vacancy cascade following threshold photoionisation, an initially ejected M_5 photoelectron can be overtaken by the Auger electrons ejected in the subsequent steps of the cascade, which have higher energy. This overtaking causes sudden changes in the Coulomb field for the photoelectron and the Auger electrons, which causes the photoelectron to lose energy and the Auger electrons to gain energy. The impact of PCI on the energy distribution of the photoelectrons has been investigated extensively (see Ref. [176] and therein). But the impact of PCI on the residual charge-state distributions resulting from the threshold photoionisation remains to be established, and any future studies on this topic should be encouraged. In this study, **BrIccEmis** does not account for PCI in the simulation of charge-state distribution.

Figure 5.9 shows that the Auger energies from EADL agree well with transition energies calculated using the Dirac-Fock method discussed earlier but the Coster-Kronig energies can differ significantly. The discrepancy in Coster-Kronig energy can lead to drastic changes

in transition rates because the Coster-Kronig rate is sensitive to the transition energy [105]. It is worth noting that non-radiative transition rates in EADL are also calculated using the frozen-orbital approximation. In addition, non-radiative transition rates derived from the frozen-orbital approximation have been shown to be larger than transition rates obtained using the fully relaxed wave functions² [205, 206]. The agreement with experimental distributions could potentially be improved with the use of relaxed wave functions in future studies³. The agreement between the theoretical distribution calculated by Kochur et al. [54] and experiments for xenon M₅ photoionisation could be attributed to the partial inclusion of the relaxation effect.

Another important aspect for improvement in future studies is the calculation of transition energies. The **RAINE** code usually overestimates the binding energies and this could result in overestimation or underestimation of transition energies (see Chapter 4). Apart from K-shell transitions, the deviations from accepted values are usually in the order of several eV to tens of eV and for the most part, can be considered as good agreement. However, the charge-state distribution is highly sensitive to the energies of low-energy transitions such as the Coster-Kronig transitions. Therefore, the accuracy of the charge-state distribution simulated by **BrIccEmis** is currently limited by the accuracy of the transition energies calculated from **RAINE**. Hence, any possible ways to improve the accuracy of transition energies, including energy calculation using the **Grasp2K** package, should be explored.

5.7 Summary

The **BrIccEmis** simulation agrees well with the experimental charge-state distributions following the M₅-subshell photoionisation apart from the strong intensity of Xe⁷⁺ ions observed in all experiments. For the decay of ^{131m}Xe, the **BrIccEmis** simulation agrees well with experiment in terms of the distribution width but produces the peak at a slightly lower charge state. The agreement could be improved with a more accurate calculation of transition energies. Moreover, the comparison of the charge-state distribution simulated based on the Dirac-Fock atomic transition probabilities suggests that the use of the "frozen-orbital approximation" in the calculation of non-radiative transition rates is a more critical

²Calculation of transition rates using independently-optimised initial- (single-hole) and final-state (double-hole) wave functions. The two wave functions are inherently non-orthogonal to each other. The seminar paper by Löwdin [201] has provided a set of equations for cases where different orbitals are used for the initial and final states and they are exploited for the calculation of radiative transition rates in the REOS program of the **Ratip** package. However, the implementation for non-radiative transition rates, which is governed by the Coulomb interaction between electrons (a two-body operator), is far from trivial. Although a lot of effort has been devoted to implement the Löwdin equations in the **AUGER** program of **Ratip** and the MCDFGME code [202, 203], the resulting non-radiative rates are disputable and have hardly been used. Nevertheless, another method based on the biorthogonal transformation [204] for the computation of Auger rates is being implemented by Dr. Jörgen Ekman in the **Grasp2K** package which means the frozen-orbital approximation may not be needed for much longer.

³On the other hand, experimental distributions could be used for validation of the calculation of non-radiative transition rates using the biorthogonal transformation.

aspect than the use of accurate energies, especially for those very low-energy Coster-Kronig transitions.

Acknowledgements

The author thanks Dr. Jörgen Ekman for providing Dirac-Fock atomic transition energies and rates as well as valuable comments.

Towards understanding the charge-state distribution of fission fragments

Overview

In this chapter, the perturbed charge-state distributions of three barium isotopes produced in the $^{233}\text{U}(\text{n}_{\text{th}},\text{f})$ reaction are studied using the theory of the vacancy cascade in order to understand the underlying principles behind the observed shifts in the charge state distributions compared with empirical expectations. This is also the first time, to the author's knowledge, that such an ionic-charge distribution is being studied theoretically. Here, a novel technique, which is developed in this thesis, that combines the vacancy-cascade simulation and a Bayesian analysis is used to extract useful parameters which can be compared to widely accepted semi-empirical calculations of charge state distributions. Several future directions that could improve the agreement with the experiment are also discussed.

6.1 Background

A recent 2013 campaign of $^{233}\text{U}(\text{n}_{\text{th}},\text{f})$ fission-yield measurements using the LOHENGRIN mass separator [12] at the Institut Laue-Langevin (ILL) has been performed [207]. Figure 6.1 shows a schematic diagram of the LOHENGRIN mass separator. A fissile actinide target is placed close to the reactor core at ILL, which operates at a thermal neutron flux reaching $5 \times 10^{14} \text{ neutrons.cm}^{-2}.\text{s}^{-1}$. The target is prepared in the form of a thin layer of UO_2 (typically between 50 and 150 $\mu\text{g}/\text{cm}^2$) on a Ti backing, covered with a 0.25 μm nickel foil. The nickel foil is usually fixed in a 0.3 mm thick titanium frame that is placed directly on the target backing [208]. Fission fragments emerge from the target along the beam tube axis and are analysed by LOHENGRIN according to their A/q and E/q ratios (where A , q and E are the mass, charge state and kinetic energy of the ions, respectively).

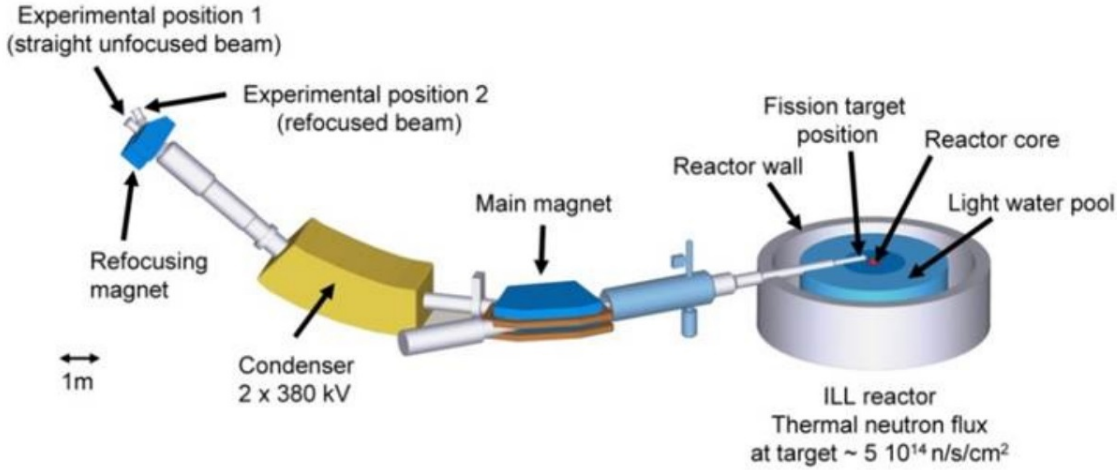


Figure 6.1: Schematic diagram of the mass separator LOHENGRIN at ILL.

The primary goal of the campaign was to assess precisely the fission yields in the heavy mass region, which becomes increasingly important as the interest in the Th- ^{233}U cycle grows [207]. However, isotopic studies¹ using the same data set reveal that ionic charge distributions of many fission fragments are affected by the presence of nanosecond isomers (see Ref.[207] for details of isotopic yield analysis). The experimental data allows a detailed study of perturbed ionic charge distributions to extract nuclear physics parameters such as isomeric ratio, etc.

An equilibrium charge distribution is usually established during the fragment's passage through the nickel foil. The equilibrium distribution is approximately a Gaussian [210] centred at $\bar{q} \approx 20\text{--}21$ for fragments with mass around $A = 140$ ². Rzaca-Urban et al. [209] have recently shown that a shift in the mean charge, $\bar{q}$ is an indication of the presence of ns and/or μs nuclear isomer(s). Figure 6.2 (also the Fig. 10 of Ref. [209]) compares the unperturbed ionic charge distribution of ^{137}Te and shifted distributions of ^{142}Cs and ^{144}Cs . This effect is explained as a direct consequence of internal conversion processes following isomeric decays in fission fragments after they pass the Ni foil, i.e. after the equilibrium charge distribution has been established [12]. If such a transition is strongly converted and bearing a significant fraction of the total decay intensity, the internal conversion triggers atomic relaxation via inner-shell ionisation which further increases the charge state of the already highly-ionised fragments, through the vacancy cascade. If the transition happens before the main magnet, which is $\approx 8\text{ m}$ from the target³, then at the exit of the separator one observes the shifted ionic charge distribution.

¹An isotopic study is an investigation of the yield of a specific mass and Z .

²Exact $\bar{q}$, also depends on the kinetic energy of a fragment before the Ni foil.

³Time of flight from the exit of the Ni foil and the main magnet is approximately 880 ns for a fragment with mass, $A = 140$ and kinetic energy, $K_E = 60\text{ MeV}$

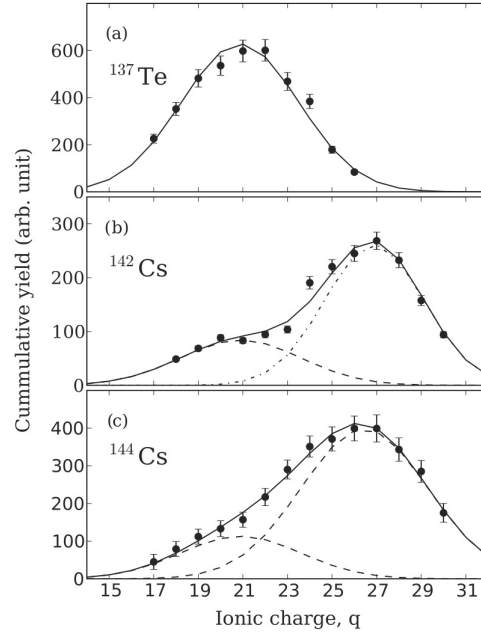


Figure 6.2: Ionic charge distribution for (a) ^{137}Te , (b) ^{142}Cs , and (c) ^{144}Cs . An unperturbed distribution is observed for ^{137}Te while the distributions of ^{142}Cs and ^{144}Cs are shifted by the presence of nuclear isomers. Image extracted from Ref.[209].

This chapter presents results from a detailed study of the ionic charge distributions of several barium isotopes, which have low-lying nanosecond isomers, based on the evaluated nuclear structures from ENSDF [113] and a novel technique to extract nuclear physics parameters from the distribution. Experimental data used for the analysis in this chapter were provided by Dr. Gregoiré Kessedjian and his coworkers at the Laboratory of Subatomic Physics & Cosmology, Grenoble, France.

6.2 Simulation of residual charge distribution for highly-ionised ions

Since the mean equilibrium charge state of fission fragments exiting the Ni foil is usually about 20+ for mass around $A = 140$, the vacancy cascade accompanying internal conversion occurs in highly-ionised ions. This means that most outer-shell electrons are stripped away from the atoms and the vacancy cascade only involves the remaining inner-shell electrons; therefore the number of electrons that could be ejected in a cascade process is significantly reduced. As a first approximation, it is assumed that an ion bears the *ground-state* electron configuration for the given charge state before a vacancy-cascade simulation begins. For example, the starting electron configuration of a Ba ($Z = 56$) ion of 20+ for simulation is assumed to be $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6$, i.e. the ground state of a Kr-like Ba ion, before the internal conversion and subsequent vacancy cascade proceed.

General protocol

For the vacancy-cascade simulation in highly-ionised ions, the following steps are followed:

1. Nuclear structure data is extracted from the Evaluated Nuclear Structure Data File (ENSDF) [113].
2. **BrIcc** is then used to calculate the internal conversion coefficient (ICC) of each subshell that is available in the ground-state electron configuration of a given initial charge state, q_i for all nuclear isomers. These ICC values are used to determine the initial vacancy distribution for the vacancy-cascade simulation.
3. The **INC-VC** mode of **BrIccEmis** is used to simulate the nuclear-level cascade from a selected nuclear isomer, if there is more than one, as well as the subsequent vacancy cascade. It is assumed that only atomic transitions with mean lifetime shorter than 1 ns are allowed to occur during the vacancy cascade⁴.
4. The total number of electrons ejected, including conversion, Auger and Coster-Kronig electrons, is recorded when the nuclear ground state is reached in the simulation. The final ionic charge of an ion, q_f is represented as the sum of the initial charge state, q_i and the total number of electrons emitted.
5. The final charge-state distribution for each initial charge state is calculated based on 100,000 simulated events.

The shake-off process in highly-ionised ions

Unlike the case of neutral atoms, the shake-off probability is highly suppressed in heavily-ionised systems [54]. The shake-off probabilities due to K-shell internal conversion in the Kr-like Ba ion have been derived based on the methodology described by Carlson and Nestor (see Eq. (2.33)). Table 6.1 shows the calculated shake-off probability for subshells up to N_3 , that is the highest subshell in the ground state of Kr-like Ba. The total shake-off probability is 4.8% per K-shell internal conversion. In comparison, the total shake-off probability for a K-shell vacancy is usually about 20-25% [14]. Since the shake-off probability is always the biggest for K-shell vacancies, the significance of shake-off accompanying internal conversion from other subshells or non-radiative transitions is expected to be even smaller. Therefore, shake-off is neglected in the vacancy-cascade simulation for highly-ionised systems.

⁴Since this chapter concerns nanosecond isomers, the atomic rearrangement due to the vacancy cascade should be terminated before the next nuclear transition. All non-radiative transitions proceed very quickly within the first 1 ps after the creation of an atomic vacancy, but the mean lifetime of some outer-shell radiative transitions is in the order of 100 ns. If these slow radiative transitions are allowed to proceed, the charge of an ion would not be modified but the electron configuration for the vacancy cascade associated with the next nuclear transition could be different.

Table 6.1: Probability (in percent) for shake-off from the various subshells, n_s in Kr-like barium ions as a result of a sudden vacancy in the orbital n_0 due to internal conversion calculated using the methodology proposed by Krause and Carlson [85]. Only K-shell internal conversion is considered and the P_F term in Eq. (2.33) is neglected.

$n_0 \backslash n_s$	K	L ₁	L ₂	L ₃	M ₁	M ₂	M ₃	M ₄	M ₅	N ₁	N ₂	N ₃	Total
K	<0.01	0.05	0.08	0.14	0.18	0.27	0.52	0.40	0.59	0.50	0.70	1.37	4.80

Choice of propagating vacancy

Figure 5.5 shows that the simulated charge-state distribution in the isomeric decay of $^{131\text{m}}\text{Xe}$ for the neutral atom is dependent on the choice of the propagating vacancy at each step of the vacancy cascade. On the other hand, the simulated ionic charge distribution following M₅-subshell photoionisation in xenon is insensitive to the same parameter (see Figure 5.1), and this could be attributed to the rather short vacancy cascade that follows the creation of an M₅ vacancy. Therefore, the simulation of the vacancy cascade in a heavily-stripped atom, which is short in length due to the reduced number of bound electrons, is expected to be rather independent of the choice of the propagating vacancy. Figure 6.3 compares the charge-state distributions following K-shell internal conversion in highly-ionised barium simulated using different choices of the propagating vacancy (see Section 3.4.4 for more details). The comparison shows that, apart from Ba¹⁸⁺, the simulated distributions are largely constant across the four different options. Nonetheless, the discrepancy between the highest-vacancy option and the rest for Ba¹⁸⁺ is not significant. Therefore, all simulated distributions presented from here onwards are based on the **random-vacancy** option.

6.3 Analysis of perturbed charge-state distribution

The equilibrium charge-state distribution, which is also referred as the unperturbed distribution⁵, may be sufficiently described by a Gaussian distribution [210]. However, the perturbed charge-state distributions due to isomeric decays and their associated vacancy cascades can no longer be represented by a Gaussian distribution. Nonetheless, perturbed charge distributions measured at ILL could be explained well by multiple Gaussian distributions [211]. Figure 6.4 shows the fitting of the perturbed charge distribution of ^{141}Ba using several different Gaussian distributions. However, the physical basis of the use of these Gaussian distributions is not established.

BrIccEmis offers a new theoretical method to analyse the perturbed charge-state distribution using the vacancy cascade. The final charge-state distribution can be simulated for different initial ionic charges. Then one can convolute the unperturbed charge-state

⁵Because it is not perturbed by isomeric decays.

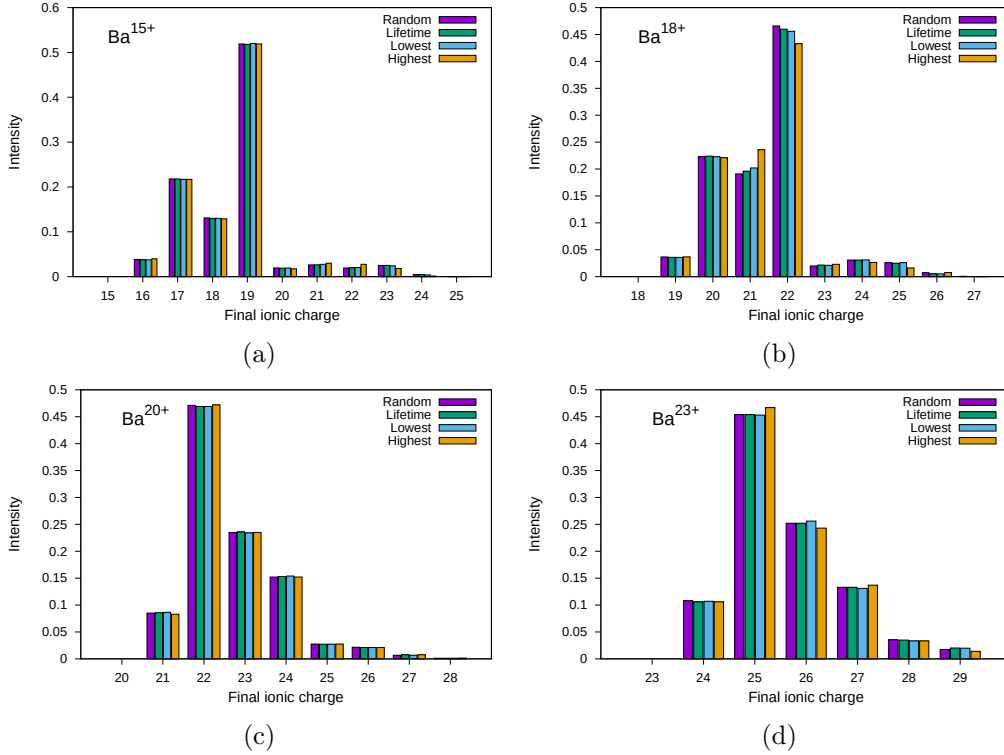


Figure 6.3: Simulated charge-state distributions following K-shell internal conversion in barium based on four different choices of the propagating vacancy for four initial charge states at (a) $15+$, (b) $18+$, (c) $20+$, and (d) $23+$.

distribution, which is assumed Gaussian, with the simulated final ionic-charge distributions to produce a perturbed one. However, for the measurements performed at the ILL, the only observable is the perturbed charge-state distribution and the information about the unperturbed one is lost in the process. Therefore, one could derive the mean and standard deviation of the unperturbed charge-state distribution for useful comparisons⁶. For this chapter, a Bayesian approach is adopted for the derivation of the mean and standard deviation of the unperturbed charge-state distribution. The inferred results were then compared with the expected values calculated using semi-empirical methods.

6.3.1 Bayesian approach

An experimental charge-state distribution can be utilised to infer the population of the nuclear ground state and excited states, and the mean and standard deviation of the unperturbed ionic-charge distribution. The inference is based on a Bayesian approach and all aforementioned quantities are treated as the free parameters that need to be derived:

$$P(\theta|data;\eta) = \frac{P(data|\theta;\eta)P(\theta)}{P(data)}, \quad (6.1)$$

⁶With that said, one could, in principle, analyse a number of charge-state distributions for fission fragments that do not have isomers and establish a set of parameters for the unperturbed distributions.

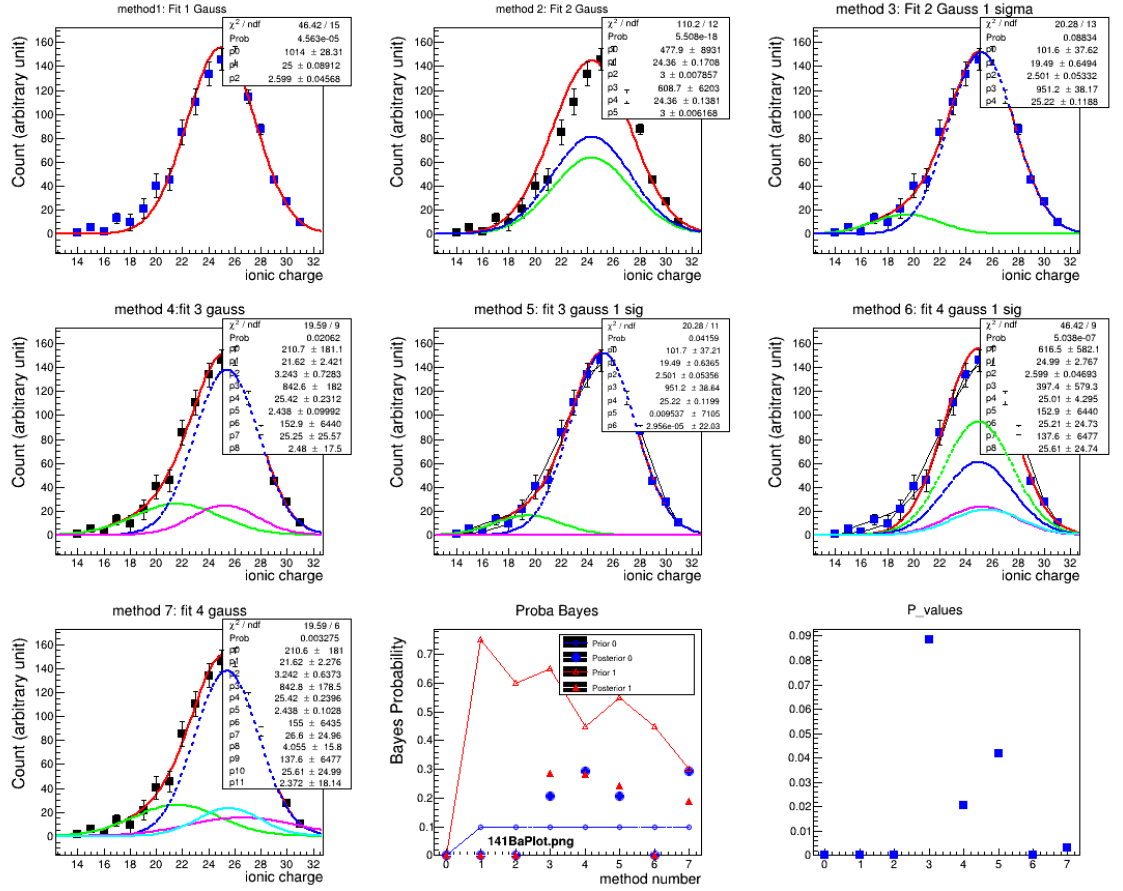


Figure 6.4: Multiple-Gaussian fitting of the perturbed charge distribution of ^{141}Ba . Image credit: Grégoire Kessedjian and Abdelaziz Chebboubi (LPSC, France).

where the “*data*” is the experimental ionic-charge distribution, “ θ ” is the assumption on one of the free parameters, and “ η ” are the other free parameters at fixed values. $P(\theta|\text{data};\eta)$ is the probability of hypothesis θ given data and η and $P(\theta)$ represents the *prior* knowledge on the parameter of interest which we do not have, and therefore it is assumed to follow a uniform distribution, i.e. $P(\theta) = 1$. Since $P(\text{data})$ is the probability of the data averaged over all parameters and independent of θ , Eq. (6.1) becomes:

$$P(\theta|\text{data};\eta) \propto P(\text{data}|\theta;\eta) = \mathcal{L}(\text{data}|\theta;\eta) \propto \exp(-\chi^2/2), \quad (6.2)$$

where $\mathcal{L}$ is the likelihood function for a Gaussian propagation of uncertainties with χ^2 being the least square function. For example, if θ is the population of the ground state, then the other free parameters η , which include the population of the excited states and the mean and standard deviation of the unperturbed ionic-charge distribution, are set at certain values, so $P(\theta|\text{data};\eta)$ can be determined for all possible values of θ . After all possible values of θ have been considered, the other free parameters η will be changed to another set of values. This repeats until all possible sets of values for η have been accounted for.

The following list details the range of values for each free parameters that are considered in this thesis:

1. The population of a nuclear state⁷: 0 - 120%.

Bayesian step size: 1%.

Since the theoretically derived perturbed charge-state distribution will be normalised to the area of the experimental distribution, but the latter is usually missing some charge states with very low intensity, the sum of the derived population of nuclear states is therefore allowed to go over 100%.

2. The mean charge of the unperturbed distribution: 15 - 25.

Bayesian step size: 0.1.

Most of the experimentally-observed unperturbed distributions have the mean charge around 20 for mass region $A = 140$.

3. The standard deviation of the unperturbed distribution: 1 - 5.

Bayesian step size: 0.1.

The width of the experimental distributions measured at ILL are around 3.

A probability distribution can be derived for each parameter using such a Bayesian analysis. The probability distribution can then be used to estimate the uncertainty of a derived parameter.

6.3.2 Semi-empirical calculation of the mean and width of the equilibrium charge distribution

The Bayes-derived mean charge of unperturbed distribution, which is established during the passage of fission fragments (projectile) in the nickel foil (target), can be compared to semi-empirical values for validation. Among the semi-empirical formulas derived by different authors, the frequently used ones include one by Nikolaev and Dmitriev [212]:

$$\begin{aligned}\bar{q}(Z_p, v_p)_{\text{N\&D}} &= Z_p \cdot (1 + 2.29 \cdot \left(Z_p^{-0.45} \left(\frac{v_p}{v_0} \right) \right)^{-5/3})^{-3/5} \quad \text{with} \\ d_{\text{N\&D}} &= \frac{1}{2} \sqrt{\bar{q} \left(1 - \left(\frac{\bar{q}}{Z_p} \right)^{5/3} \right)},\end{aligned}\tag{6.3}$$

and one by Betz [213]:

$$\bar{q}(Z_p, v_p)_{\text{Betz}} = Z_p \cdot (1 - 1.041 \cdot \exp \left(-0.851 \cdot Z_p^{-0.432} \left(\frac{v_p}{v_0} \right)^{0.847} \right)) \quad \text{with} \quad d_{\text{Betz}} = 0.27 \cdot Z_p^{1/2},\tag{6.4}$$

⁷A nuclear state here can be the ground state or an excited state.

where $\bar{q}$ and d ⁸ are the mean charge and width of the equilibrium (unperturbed) distribution, respectively, Z_p and v_p are the nuclear charge and velocity of the projectile ions, respectively, and v_0 is the Bohr velocity of 2.188×10^6 m/s. Recently, Schiwietz and Grande [214] provided an updated formula that includes the dependency of the nuclear charge of a solid target Z_t . Their formula for the mean charge of the equilibrium distribution is

$$\bar{q}(Z_p, Z_t, x)_{\text{S\&G}} = Z_p \frac{12x + x^4}{0.07/x + 6 + 0.3x^{0.5} + 10.37x + x^4} \quad \text{with} \quad (6.5)$$

$$x = \left(\left(\frac{v_p}{v_0} \right) Z_p^{-0.52} \frac{Z_t^{-0.019Z^{-0.52}v_p/v_0}}{1.68} \right)^{1+1.8/Z_p},$$

and the width

$$d(Z_p, Z_t, q)_{\text{S\&G}} = w \cdot \frac{Z_p^{0.27} \cdot Z_t^{-0.035+0.0009 \cdot Z_p}}{f(\bar{q})f(Z_p - \bar{q})} \quad \text{with} \quad f(x) = \sqrt{\frac{(x + 0.37 \cdot Z_p^{0.6})}{x}}. \quad (6.6)$$

where w is the reduced width⁹.

All three formulae show a smooth trend and come from averages which do not include the effects of closed-shell configurations of the projectile ions. The projectile-shell effects, which have been extensively shown in Refs. [214, 215], can have a large effect on both the mean charge and distribution width. Attempts have been made to calculate correction factors for the projectile-shell effects, but these calculations are dependent on both the nuclear charge and energy of the projectile ion. Therefore extrapolation is difficult and the correction factors could only be calculated on a case-by-case basis [216].

Estimation of the velocity of projectile ions

This section describes the procedure for estimating the velocity of projectile ions before they impinge on the nickel target. This velocity is required for the calculation of the mean and standard deviation of the equilibrium charge distribution established during the ions' passage in the target. The LOHENGRIN mass separator can be set to allow only fission fragments with specific kinetic energies¹⁰ to reach the detectors at experimental positions 1 and 2 (see Figure 6.1). Table 6.2 shows the projectile-kinetic energies of several masses around $A = 140$ used for determining the charge-state distributions during the $^{233}\text{U}(\text{n}_{\text{th}}, \text{f})$ fission-yield measurements campaign [207]. Since these values are the energies of fission fragments when they *exit* the target nickel foil, they can be used to estimate the velocity of projectile ions before they *enter* the target, if the target thickness and the nuclear charge of

⁸Here $d = [\sum (q - \bar{q})^2 P_q]^{1/2}$ and P_q is the normalised charge-state distribution.

⁹ w is close to 0.7 empirically [214].

¹⁰For measurements of charge-state distributions, which require the gamma spectrometry to isolate the individual isotopes of the same mass, the maximum energy acceptance is usually around 4.4%, i.e. $\pm 2.2\%$.

the target, which is $Z = 28$ (Ni), and projectile are known. The projectile-fission fragments concerned in this chapter are mainly the barium isotopes, i.e. $Z = 56$, and the thickness of the nickel foil used in the measurements campaign was $0.25 \mu\text{m}$ with a 30% uncertainty¹¹.

Table 6.2: Projectile-kinetic energy of mass, $A = 139 - 145$ used for charge-state measurements during the $^{233}\text{U}(\text{n}_{\text{th}}, \text{f})$ fission-yield measurements campaign [207].

Mass, A	Kinetic energy, E_K (MeV)
139	51.0
140	60.0
141	51.0
142	48.0
143	48.0
144	46.0
145	46.0

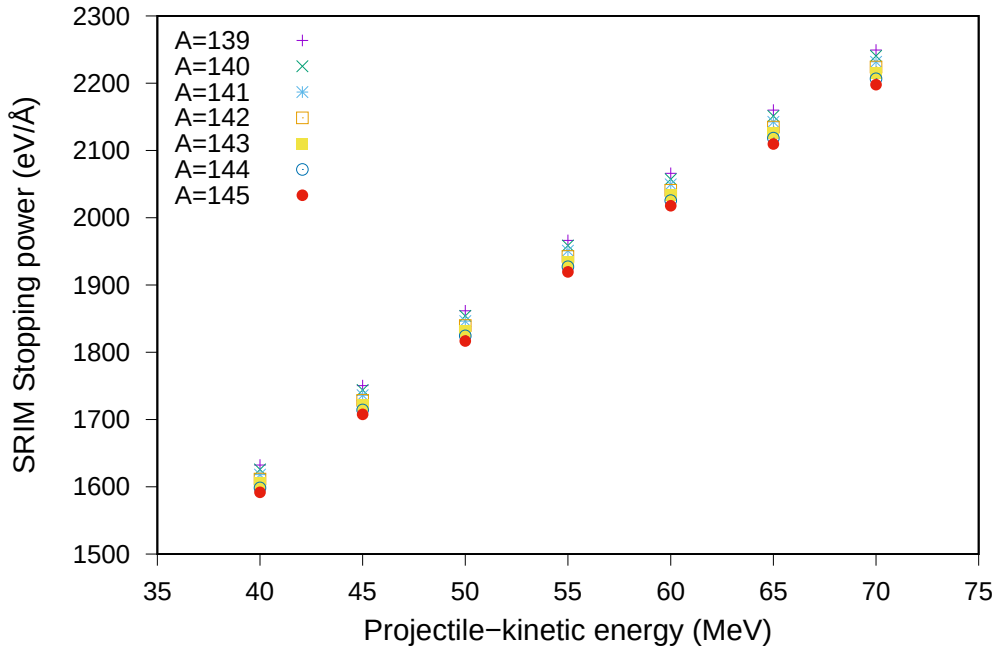


Figure 6.5: SRIM stopping powers ($\text{eV}/\text{\AA}$) of barium ions ($Z = 56$) in nickel target for projectile-kinetic energies between 40 and 70 MeV.

The stopping powers for barium ions in a nickel solid target were extracted from the SRIM-2013 software package [217]. Figure 6.5 shows the SRIM stopping powers for barium ions with different mass within the energy range of interest. The stopping powers exhibit a linear relationship with the projectile-kinetic energies and a small dependence on the mass. A linear least-square regression was therefore used to estimate the stopping-power function

¹¹It is worth noting that the nickel foil was depleting during the measurements campaign, so the exact thickness at the time of measurement for each barium isotope is difficult to be determined. The thickness reported here is the known quantity before the campaign.

and Table 6.3 shows the slope and y -intercept of the derived stopping-power function for several isotopes.

Table 6.3: The slope and y -intercept of the estimated stopping-power function for different barium isotopes. Projectile-kinetic energy is in the unit of eV while the stopping power is in eV/Å.

Isotope	Parameters of linear regression	
	slope	y -intercept
^{139}Ba	2.0530×10^{-5}	825.96
^{140}Ba	2.0446×10^{-5}	822.73
^{141}Ba	2.0406×10^{-5}	816.98
^{142}Ba	2.0336×10^{-5}	811.51
^{143}Ba	2.0290×10^{-5}	808.02
^{144}Ba	2.0243×10^{-5}	802.96
^{145}Ba	2.0167×10^{-5}	799.45

One can calculate the *entering* projectile-kinetic energy, E_0 by solving the following differential equation:

$$\frac{dE(x)}{dx} = -(aE + b), \quad (6.7)$$

where x is the total distance travelled in the target by the projectile, which is the thickness of the nickel target, $\frac{dE(x)}{dx}$ is the stopping-power function, and a and b are the slope and y -intercept of the stopping-power function estimated from the linear regression. Therefore, the projectile-kinetic energy can be expressed as the following:

$$E_0 = \frac{E_f + b/a}{\exp(-ax)} - \frac{b}{a}, \quad (6.8)$$

where E_f is the *exiting* projectile-kinetic energy, which is known. Table 6.4 shows the calculated entering projectile-kinetic energy, E_0 , and its corresponding velocity, v_0 , for the concerned barium isotopes. If the uncertainties of a and b are neglected, then the uncertainty of E_0 can be propagated from the uncertainties of E_f and the thickness of nickel target based on Eq. (6.8) as follows:

$$\begin{aligned} \Delta E_0 &= \frac{dE_0}{dE_f} \cdot \Delta E_f + \frac{dE_0}{dx} \Delta x \\ \Rightarrow \Delta E_0 &= e^{ax} \cdot \Delta E_f + ae^{ax} \cdot \Delta x, \end{aligned} \quad (6.9)$$

where $\Delta E_f = 0.022E_f$ and $\Delta x = 0.3x$, and E_f and x are assumed to be uncorrelated. The uncertainty of the velocity presented in Table 6.4 is propagated from E_0 ¹². It is also important to note that for the Rb-like and Kr-like configurations, i.e. Ba^{19+} and Ba^{20+}

¹²It is assumed that the rest mass of an isotope is $u \times A$, in which $u = 931.4940954 \text{ MeV}/c^2$ is the unified atomic mass unit and A is the rest mass of the isotope in the unit of u . The latter is extracted from Ref. [218].

ions, the semi-empirical equilibrium mean charge is always lower than the experimental one by around 0.5 due to the projectile-shell effects.

Table 6.4: Estimated E_0 and v_0 for several barium fission fragments based on their corresponding E_f . Semi-empirical equilibrium mean charges are calculated based on the estimated v_0 and its uncertainty is propagated. Velocity is given as a ratio v_0/c (in percent) where c is the speed of light. $\bar{q}_{\text{N\&D}}$, $\bar{q}_{\text{Betz}}$ and $\bar{q}_{\text{S\&G}}$ are calculated based on Eqs. (6.3), (6.4) and (6.5).

Isotope	E_f (MeV) ^a	E_0 (MeV)	v_0/c (%)	$\bar{q}_{\text{N\&D}}$	$\bar{q}_{\text{Betz}}$	$\bar{q}_{\text{S\&G}}$
¹³⁹ Ba	51.0	55.8(12) ^b	2.94(3)	19.9(2)	20.2(2)	20.0(1)
¹⁴⁰ Ba	60.0	65.3(14)	3.16(3)	21.1(2)	21.3(2)	21.0(1)
¹⁴¹ Ba	51.0	55.8(12)	2.92(3)	19.8(2)	20.1(2)	19.9(1)
¹⁴² Ba	48.0	52.6(11)	2.82(3)	19.3(2)	19.6(2)	19.4(1)
¹⁴³ Ba	48.0	52.6(11)	2.81(3)	19.2(2)	19.5(2)	19.3(1)
¹⁴⁴ Ba	46.0	50.5(11)	2.74(3)	18.8(2)	19.2(2)	19.0(1)
¹⁴⁵ Ba	46.0	50.4(11)	2.73(3)	18.8(2)	19.1(2)	18.9(1)

^aThe uncertainty of E_f is $\pm 2.2\%$.

^b55.8(12) means 55.8 ± 1.2 .

6.4 Case studies

In this section, the analysis of ¹⁴³Ba and ¹⁴⁵Ba decays, both of which have one known nanosecond isomer, are presented before the more complicated case of ¹⁴¹Ba.

6.4.1 ¹⁴³Ba

In ¹⁴³Ba, there is a known nanosecond isomer at 117.368 keV above the nuclear ground state and its nuclear half-life is 3.5 ns [113]. Figure 6.6 shows the experimental ionic charge distribution of ¹⁴³Ba and it can be fitted fairly well with a Gaussian function. However, the Bayes-derived equilibrium mean charge is $\bar{q} = 21.36 \pm 0.05$ ¹³, which is significantly higher than the semi-empirical values (see Table 6.4). This is expected given that the theoretical total conversion coefficient of the γ -transition from the nanosecond isomer to the ground state is $\alpha_T = 1.091$ [18]. Figure 6.7 shows the residual charge-state distributions of barium ions simulated using the properties (from ENSDF [113]) of the nuclear isomer in ¹⁴³Ba for different initial charge states. In general, the mean number of electrons emitted following an isomeric decay decreases with the increase in initial charge state.

With the simulated final charge-state distributions in hand, one can perform a Bayesian analysis to infer the most likely mean charge and width of the equilibrium distribution based on the experimental data. Figure 6.8 compares the experimental ionic charge dis-

¹³The stated uncertainty in the Bayes-derived mean charge is defined as the standard deviation calculated based on the inferred probability distribution, which could be not Gaussian, over the pre-defined range (see Section 6.3.1).

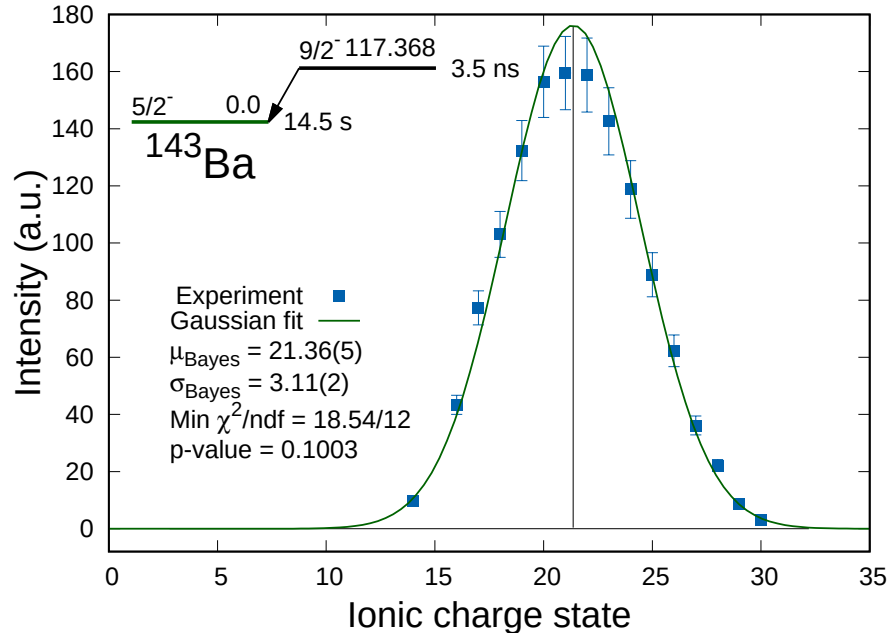


Figure 6.6: Experimental ionic charge distribution of ^{143}Ba fitted with a Bayes-derived Gaussian distribution. Nuclear structure information is extracted from the latest ENSDF [113].

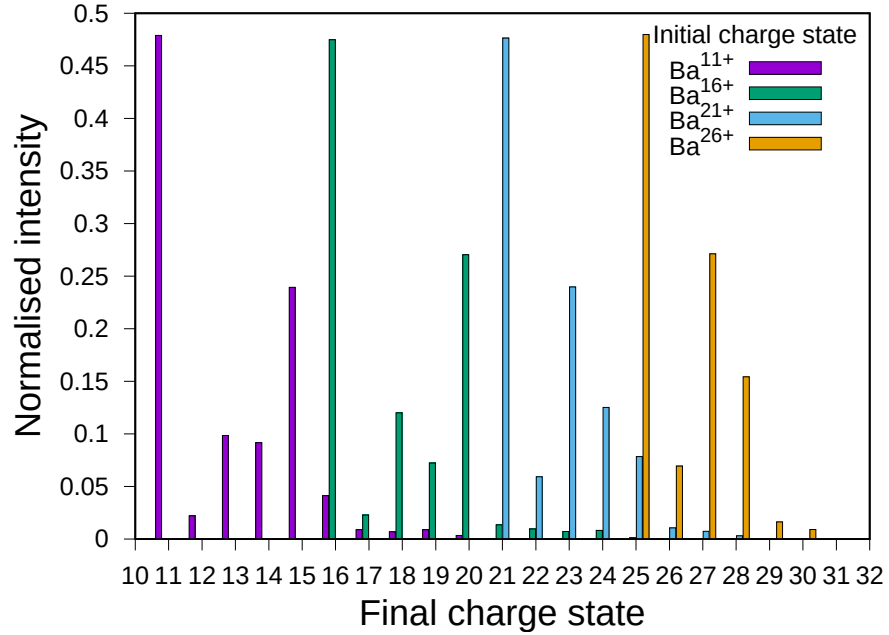


Figure 6.7: Simulated residual charge-state distributions of barium ions following the isomeric decay ($E_\gamma = 117.368$ keV) in ^{143}Ba for different initial charge states.

tribution of ^{143}Ba with a Bayes-derived perturbed charge distribution. The Bayes-derived distribution consists of an equilibrium Gaussian distribution and a perturbed charge distribution. The latter is a modified distribution based on the former due to the emission of conversion, Auger and Coster-Kronig electrons. In this case, the Bayesian analysis indicates

that the mean charge of the equilibrium Gaussian distribution is $\bar{q}_{Bayes}^{gs} = 20.76 \pm 0.23$ and the Bayes-derived width is 2.96 ± 0.06 . These values are calculated based on the posterior probability distributions derived from the Bayesian analysis (see Figure 6.9). The derived $\bar{q}_{Bayes}^{gs}$ is now in better agreement with the semi-empirical value (see Table 6.4). The broad width of the posterior probability distribution for the mean charge can be attributed to the rather large uncertainties in the experimental data points. When the individual data point has a relatively big uncertainty, any mean value within the range 20 - 21.5 will result in a relatively small χ^2 , and hence a high likelihood $\mathcal{L}$.

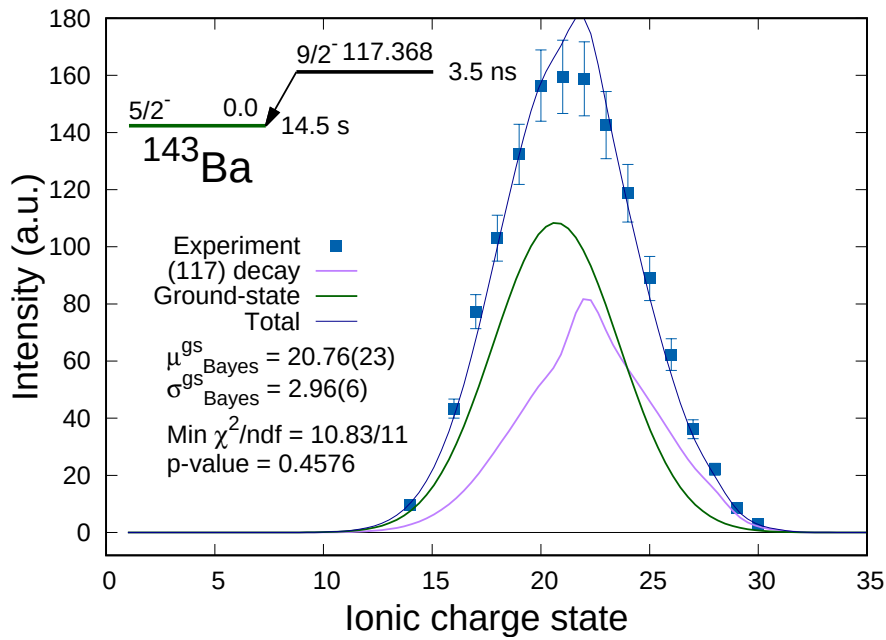


Figure 6.8: Comparison of experimental ionic charge distribution of ^{143}Ba with a Bayes-derived perturbed charge distribution.

Figure 6.10 shows the Bayes-derived correlation density plot for the ground state and the first excited state of ^{143}Ba . The density plot indicates that a bigger population of the ground state is favoured in the Bayesian analysis.

Impact of smaller uncertainties

If the uncertainty of each data point were smaller, then the Bayesian analysis could produce a more precise solution. Figure 6.11 shows the Bayes-derived correlation density plot assuming that the uncertainty of each experimental data point is 5%. The figure provides a more focused density plot than Figure 6.10 and it indicates that the population of the first excited state is comparable to that of the ground state.

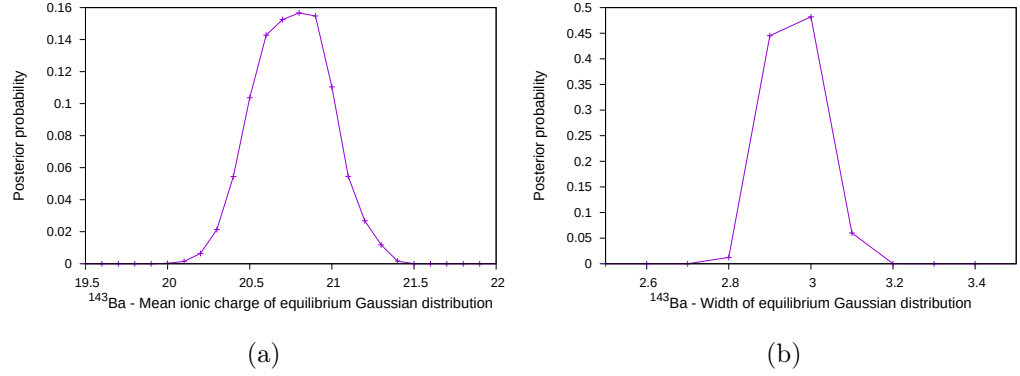


Figure 6.9: Posterior probability distributions of the (a) mean and (b) width of the equilibrium Gaussian charge-state distribution of ^{143}Ba derived from the Bayesian analysis.

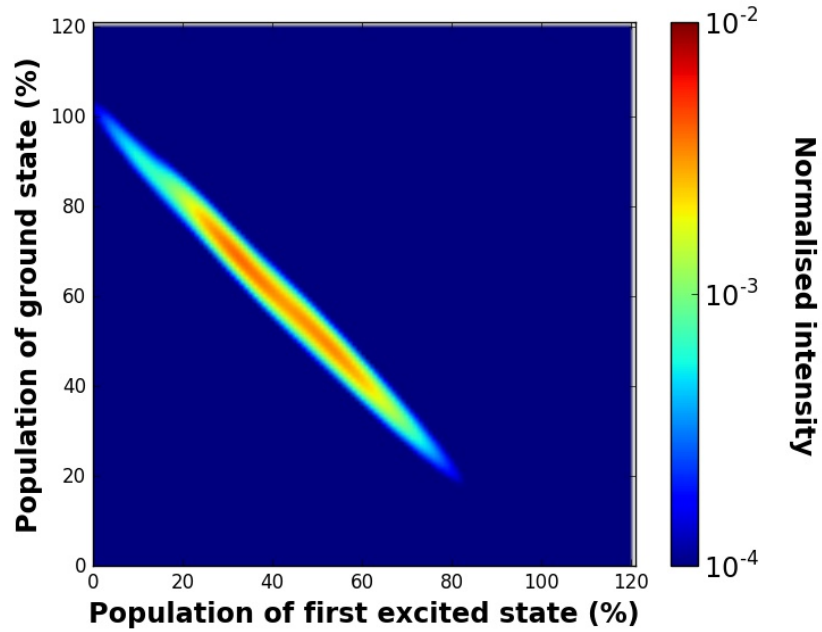


Figure 6.10: Bayes-derived correlation density plot for the ground state and the first excited state of ^{143}Ba .

6.4.2 ^{145}Ba

Figure 6.12 shows the experimental charge-state distribution of ^{145}Ba produced from the $^{233}\text{U}(\text{n}_{\text{th}},\text{f})$ reaction. A unique feature of the experimental distribution is the flat peak between 19+ and 22+, which could not be explained by a Bayes-derived Gaussian distribution. This could however be described using a combination of an equilibrium Gaussian distribution and a perturbed one resulting from the internal conversion of the γ -transition ($E\gamma = 112.64$ keV) from the isomer, which is also the first excited state, to the ground state of ^{145}Ba . The total conversion coefficient of the γ -transition, which is a mixed $M1 + E2$ transition with a mixing ratio of 0.13_{-6}^{+7} [113], is $\alpha_T = 0.716 \pm 0.16$. Figure 6.13 shows the Bayes-derived charge-state distribution for ^{145}Ba . The Bayesian analysis indicates that

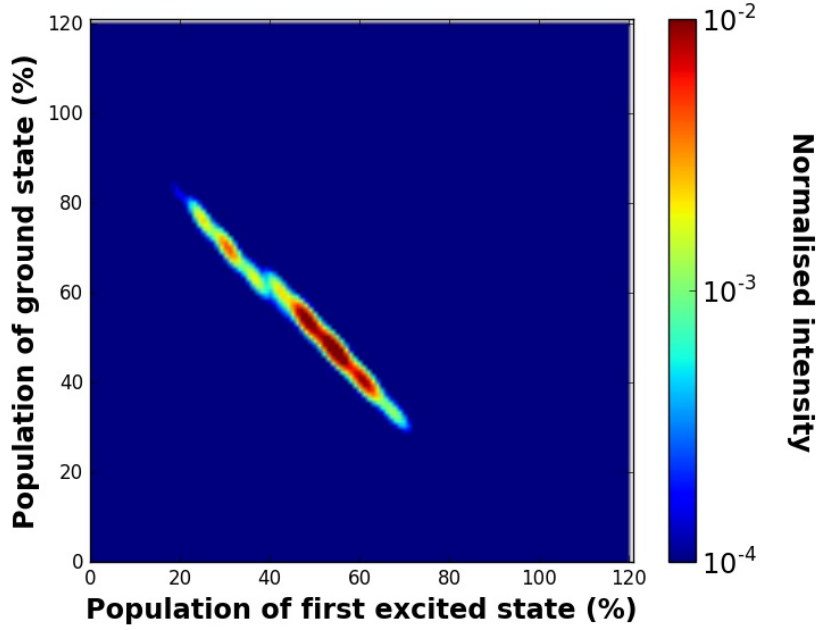


Figure 6.11: Bayes-derived correlation density plot for the ground state and the first excited state of ^{143}Ba assuming 5% uncertainty for each data point.

the mean charge of the equilibrium Gaussian distribution is $\bar{q}_{Bayes}^{gs} = 20.38 \pm 0.20$ and the Bayes-derived width is 3.40 ± 0.08 (see Figure 6.14 for the derived probability density functions for these two parameters). The perturbed charge-state distribution, which has a mean charge of 21.52 ± 0.19 , increases the intensities of 21+ and 22+ charge states, and thus flattens the peak of the overall distribution resulting in better agreement with the experiment.

Figure 6.15a displays the derived correlation density plot for the ground state and the first excited state of ^{145}Ba . It shows that there is a slight favour towards a higher population of the ground state than the first excited state, but it also shows that there are many possible combinations of the populations of the ground state and the first excited state that can lead to a small χ^2 in the analysis. Figure 6.15b demonstrates that a smaller uncertainty in the experimental data could lead to a more precise solution.

6.4.3 ^{141}Ba

Figure 6.16 shows the experimental ionic charge distribution of ^{141}Ba produced from the $^{233}\text{U}(\text{n}_{\text{th}}, \text{f})$ reaction. Due to the decays of nanosecond isomers, the observed distribution is severely perturbed and peaks at 26+ instead of 20+. The γ -ray transitions from the two known nanosecond isomers at 48.5 keV and 55 keV above the ground state, which are also the first and second excited states in ^{141}Ba , have large internal conversion coefficients. These highly converted transitions can cause several electrons to be ejected through the inner-shell ionisation by internal conversion and the subsequent vacancy cascade, and thus

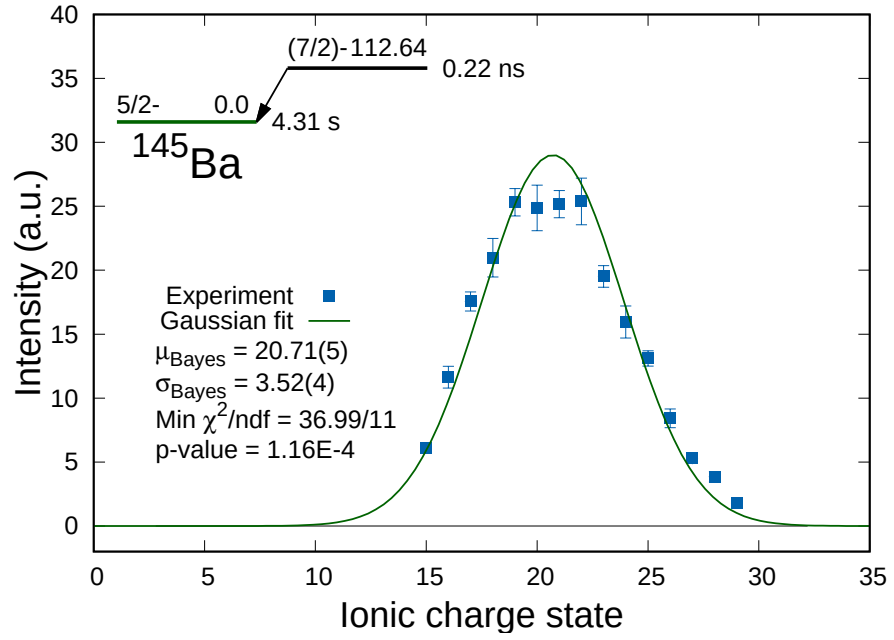


Figure 6.12: Experimental ionic charge distribution of ^{145}Ba and it is fitted with a Bayes-derived Gaussian distribution. Nuclear structure information is extracted from the latest ENSDF [113].

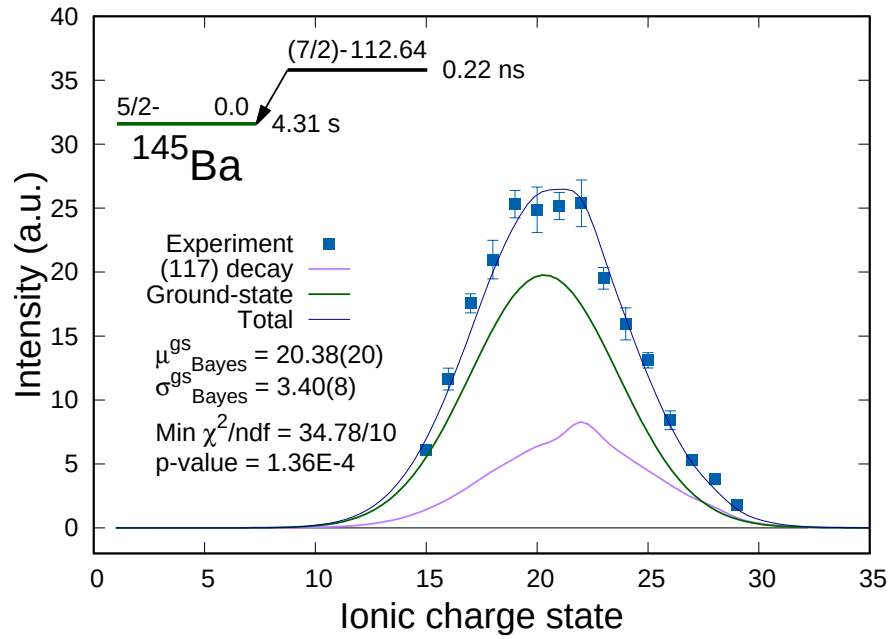


Figure 6.13: Comparison of experimental ionic charge distribution of ^{145}Ba with a Bayes-derived perturbed charge distribution.

are the main reasons behind the significant shift in the centroid of the experimental charge-state distribution.

Electromagnetic transitions between the first two excited states and the ground state are listed in Table 6.5. The internal conversion of the 6.469 keV transition between the

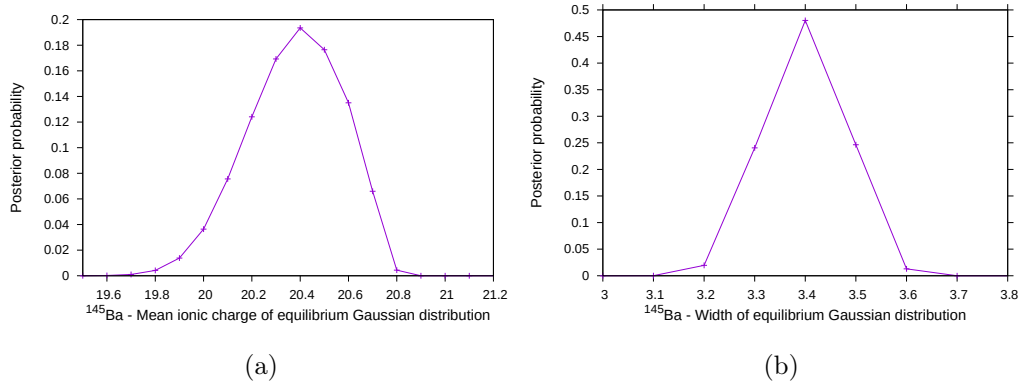


Figure 6.14: Posterior probability distributions of the (a) mean and (b) width of the equilibrium Gaussian charge-state distribution of ^{145}Ba derived from the Bayesian analysis.

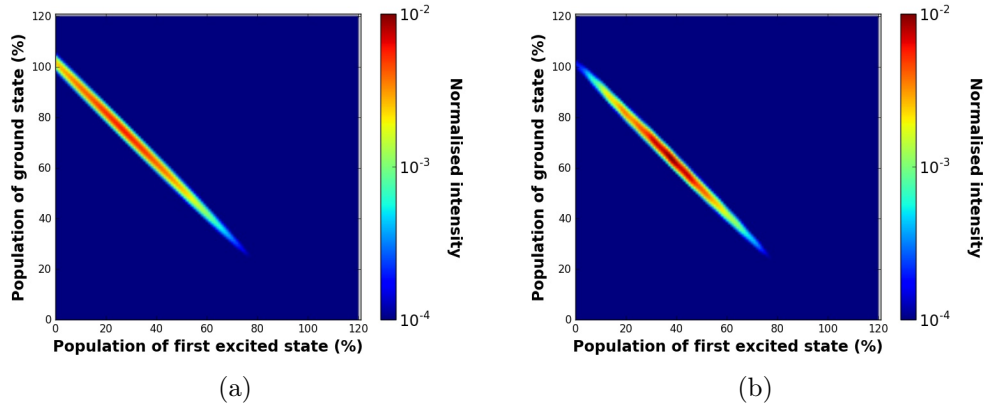


Figure 6.15: Bayes-derived correlation density plot for the ground state and the first excited state of ^{145}Ba using (a) actual experimental uncertainties and (b) assuming 5% uncertainty for each data point.

first and second excited states is, however, hindered at high-charge states. The conversion process of the other two transitions listed in the table is not considerably hindered, even though the decay pathways to outer-shells are closed in the highly-ionised states, since these shells have relatively small conversion coefficients.

Table 6.5: Electromagnetic transitions between the first two excited states and the ground state in ^{141}Ba extracted from the ENSDF [113]. E_i and E_f represent the initial and final nuclear states of a transition while E_γ is the γ energy; I_γ is the γ intensity and δ is the mixing ratio of a mixed transition. The final column shows the theoretical total internal conversion coefficients in the neutral atom, $\alpha_{T,N}$ calculated using BrIcc [18].

E_i (keV)	E_f (keV)	E_γ (keV)	I_γ	Multipolarity	δ	$\alpha_{T,N}$
55	48.53	6.469(50)	100 (9)	$M1$		456
55	0	54.997(23)	91(7)	$E2$		17.0
48.53	0	48.53(2)	100	$M1 + E2$	0.36	10.2

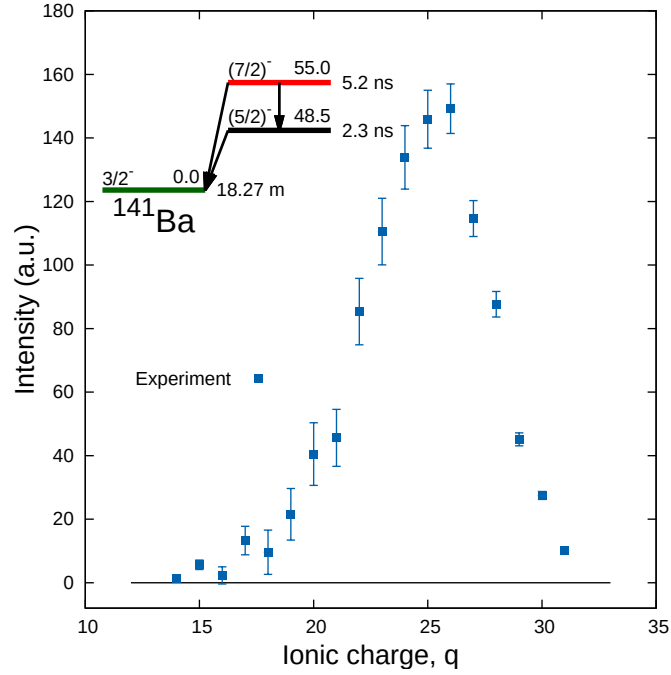


Figure 6.16: Experimental ionic charge distribution of ^{141}Ba produced from $^{233}\text{U}(\text{n}_{\text{th}},\text{f})$ fission. Nuclear structure information is extracted from the latest ENSDF [113].

Internal conversion coefficients for highly-ionised ions

Atomic binding energies increase as the charge state of an ion rises. Internal conversion coefficients of a low-energy electromagnetic transitions could be significantly reduced at high-charge state. For example, the transition between the first and second excited states of ^{141}Ba could be hindered eventually at some high-charge state because the energy of the γ -transition between the two states is only higher than the neutral L_1 subshell binding energy of barium by about 480 eV. The binding energy of the L_1 subshell increases with the charge state, and it eventually passes the transition energy, $E_\gamma = 6.469$ keV. Figure 6.17 shows the changes in the theoretical binding energies, which are calculated using the RAINE code [112], as a function of ionic charge. Binding energies of L_1 , L_2 and L_3 subshells exceed E_γ at ionic charge, $q = 18, 25$ and 31 , respectively.

This has significant impact on the internal conversion coefficients of this $M1$ transition as the L_1 conversion coefficient is the most dominant component to the total. Figure 6.18 shows the changes in the theoretical subshell conversion coefficients of the 6.469 keV $M1$ transition between the first and second excited states of ^{141}Ba , which are calculated using the BrIcc code [18]. The total internal conversion coefficient is significantly reduced at high-charge states when the L-shell binding energies exceed the transition energy. This also leads to an increase in isomeric lifetime of the second excited state.

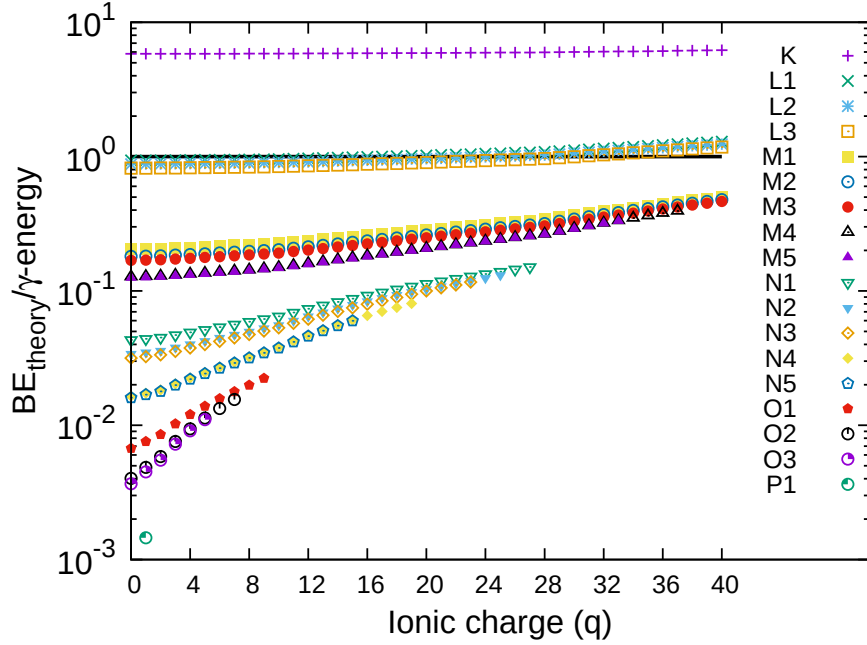


Figure 6.17: Theoretical atomic binding energies of ^{141}Ba relative to E_γ of the 6.469 keV transition as a function of charge state. Atomic binding energies increase with the charge state and this leads to the suppression of L-shell internal conversion at high-charge states.

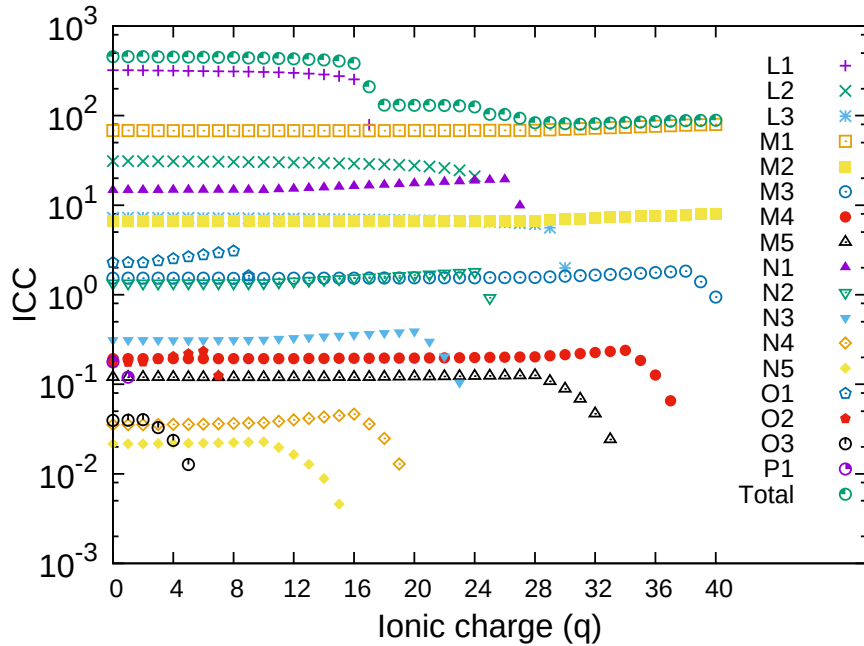


Figure 6.18: Theoretical sub-shell internal conversion coefficients of the 6.469 keV transition between the second and first excited states in ^{141}Ba as a function of charge state. Due to the increase in binding energies, conversion processes from L_1 , L_2 and L_3 sub-shells become hindered at ionic charges, $q = 18, 25$ and 31 , respectively. This leads to significant decrease in the total conversion coefficients.

Prolonged lifetime of isomeric states in ions

Fission fragments are in highly-ionised states after travelling through the nickel foil. Akber et al. [219] has recently shown that the lifetime of an isomeric state could be significantly

prolonged when the electron configuration is hydrogen-like, i.e. the atom is almost fully stripped. In that paper, **BrIcc** [18] is shown to be very reliable in the calculations of the internal conversion coefficients at high-charge states for the prediction of the prolonged lifetime of a nuclear isomeric state.

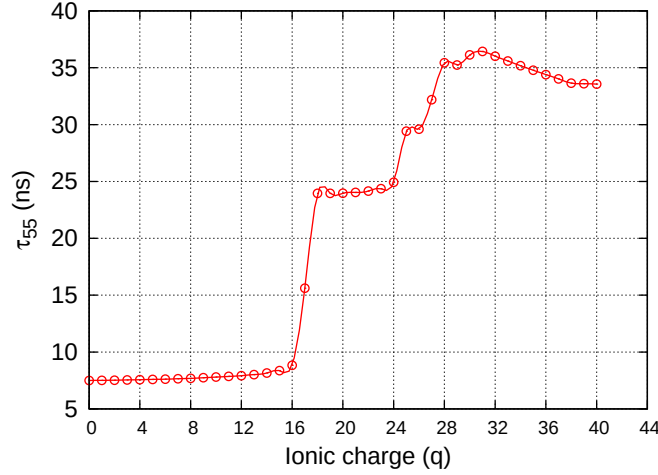


Figure 6.19: Theoretical lifetime of the second excited state in ^{141}Ba (55 keV) as a function of the ionic charge, q .

One could calculate the lifetime of an isomeric state at highly-ionised states using the equation below:

$$\tau_I = \frac{\sum_i I_{\gamma N,i} (1 + \alpha_{N,i})}{\sum_j I_{\gamma I,j} (1 + \alpha_{I,j})} \tau_N \quad (6.10)$$

where τ_I and τ_N are the lifetimes of an isomeric state in the ionic and neutral states, respectively, and α_I and α_N are the total conversion coefficients of the isomer in the ionic and neutral states, respectively. The summations are over all possible decay branches from the isomer.

Figure 6.19 shows the theoretical lifetime of the second excited state in ^{141}Ba at different ionic charges, calculated based on Eq. (6.10). Both α_I and α_N in the equations are calculated using the **BrIcc** code [18]. The increase in the lifetime is mainly due to the reduced total internal conversion coefficient of the $M1$ transition from the second excited state to the first excited state ($E_\gamma = 6.469$ keV). Conversion of the $E2$ transition ($E_\gamma = 54.997$ keV) to the ground state is not hindered due to its high transition energy relative to the atomic binding energies. At extremely high-charge states ($q > 30$), the lifetime of the second excited state decreases when the M_1 -subshell internal conversion coefficient becomes the dominant contributor to the total, and it increases with ionic charge (see Fig. 6.18).

The average time of flight from the target to the entrance of dipole magnet, which is approximately 8 m from the target, of ^{141}Ba fission fragments with the mean energy of 51 MeV is ~ 868 ns. This is equivalent to about 38 times the calculated maximum half-life

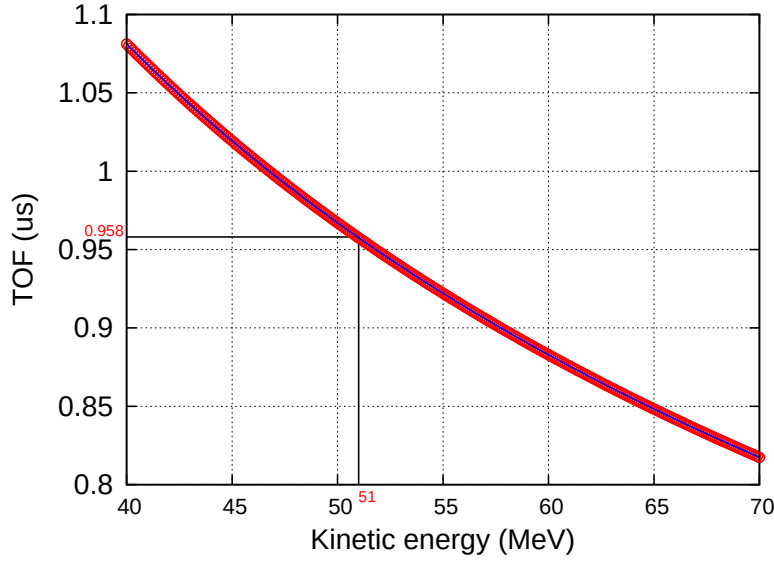


Figure 6.20: Time of flight (TOF) from the target to the entrance of dipole magnet of ^{141}Ba fission fragments as a function of kinetic energy. Fragments with mean energy of 51 MeV take approximately 868 ns to reach the magnet.

($T_{1/2}^{\max} \approx 25.2$ ns) of the 55 keV isomeric state at $31+$. Therefore, it is safe to assume that all isomeric states of ^{141}Ba must have decayed before the fission fragments enter the magnet, even though the lifetime of the second excited state has been significantly increased.

Results from Bayesian analysis

Figure 6.21 shows the fitting of the experimental ionic-charge distribution using a Bayes-derived distribution. The Bayes-derived distribution is made up of three separate components: a ground state-associated Gaussian distribution, a perturbed one caused by the decay of the first excited state and another distorted one due to the decay of the second excited state. The derived distribution is in a fairly good agreement with the experimental data given the complexity of the problem. It is also determined from the Bayesian analysis that the first excited state is the most dominant component to the total. However, the derived mean of the equilibrium Gaussian distribution, $\bar{q}_{\text{Bayes}}^{gs}$, is 23.12 ± 0.73 which is significantly higher than values calculated by the semi-empirical methods (see Table 6.4). Figure 6.22 shows the correlation density plot and posterior probability density functions (pdfs) for the derived mean and width of the equilibrium Gaussian distribution. There are two local maxima in the posterior pdf for both the calculated mean and width. The local maximum in the posterior pdf of the mean at $\bar{q}_{\text{Bayes}}^{gs} \approx 22.6$ corresponds to the local maximum in the width at $d_{\text{Bayes}}^{gs} \approx 2.75$. Another local maximum at higher mean is associated with the local maximum at a smaller width.

Figure 6.23a shows that the local maximum at lower mean is associated with a smaller population of the ground state ($\approx 20\%$) while the other local maximum in the posterior

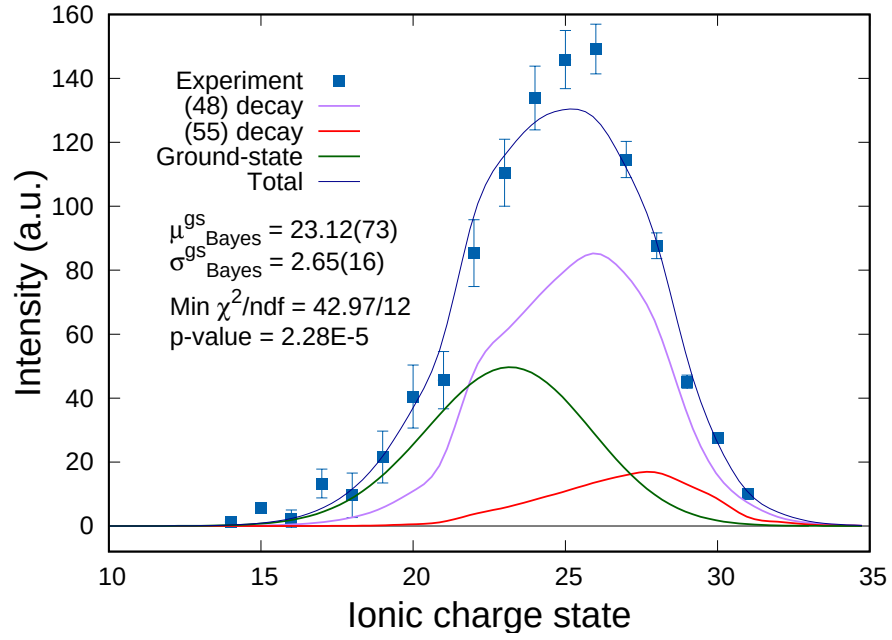


Figure 6.21: Experimental ionic-charge distribution of ^{141}Ba is fitted with a Bayes-derived distribution that consists of a Gaussian distribution associated with the ground state (green), and two perturbed distributions created due to the two nanosecond isomers (purple and red).

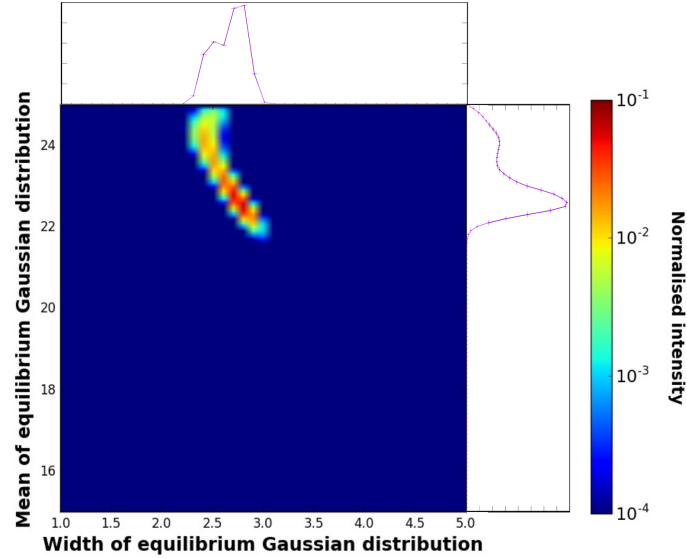


Figure 6.22: Bayes-derived correlation density plot for ^{141}Ba for the mean and width of the equilibrium Gaussian distributions. The probability density distributions for both free parameters are also shown.

pdf of the mean corresponds to a much larger population of the ground state ($\geq 60\%$). Figures 6.23b - 6.23d indicate that the most dominant species is the first excited state (up to 85%) when the mean of the equilibrium Gaussian distribution is close to 22. The second excited state of ^{141}Ba only contributes up to about 20% to the full distribution. This scenario is well represented in Figure 6.21. Another highly likely scenario derived from

the analysis is that the experimental distribution could be explained well by a Gaussian distribution with dominant contribution and its mean is ≈ 24.1 . However, this is unlikely to be physical because the derived mean is too high and a larger contribution from the two isomeric states is expected given that many excited states above them strongly populate these two isomeric states rather than the ground state.

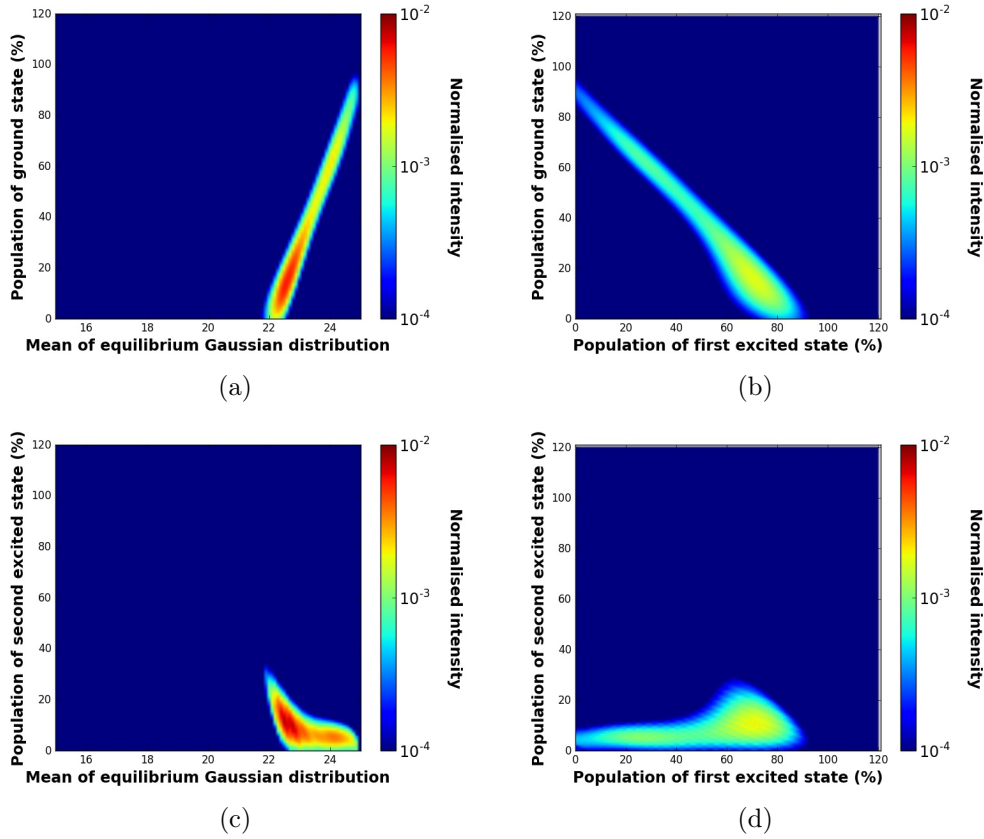


Figure 6.23: Bayes-derived correlation density plots for ^{141}Ba for (a) the mean of the equilibrium Gaussian distribution and the population of the ground state, (b) the populations of the ground state and the first excited state, (c) the mean of the equilibrium Gaussian distribution and the population of the second excited state, and (d) the populations of the ground state and the first excited state.

In the cases of ^{143}Ba and ^{145}Ba , smaller uncertainties lead to a more precise solution which is still within the solution space derived using the original experimental uncertainties. However, in this case, smaller uncertainties lead to a drastically different solution space. Figure 6.24 shows the Bayes-derived perturbed charge distribution if the uncertainty of each experimental data point is assumed to be 5%. The large minimum χ^2 achieved is mainly due to the very small absolute uncertainties for the experimental data points at small charge states but this does not undermine the fact that the experimental distribution is still well described by the derived one in terms of width and general structure. The second excited state of ^{141}Ba is now calculated to be the most dominant species, in which its population is $\approx 88\%$, while the populations of the ground state and the first excited

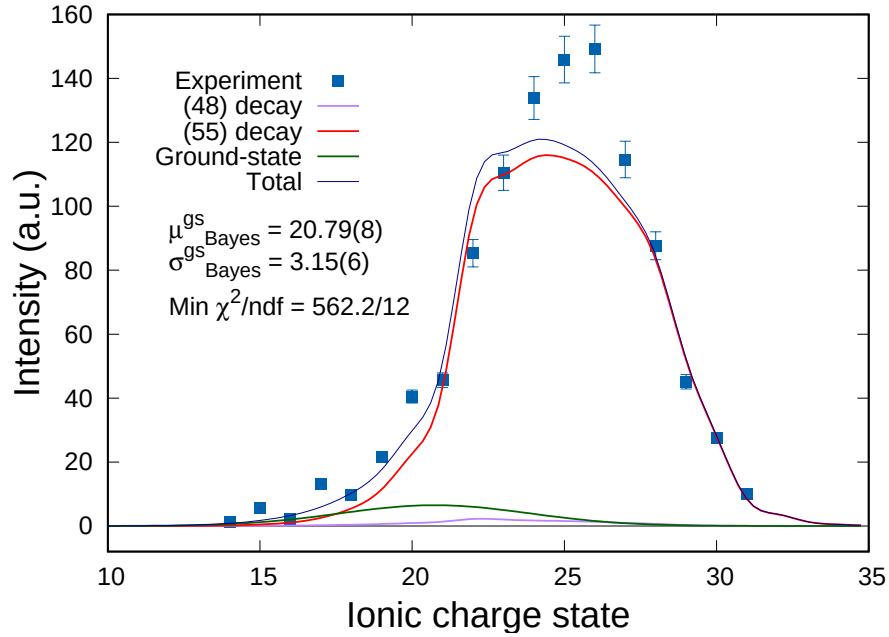


Figure 6.24: Experimental ionic-charge distribution of ^{141}Ba is fitted with a Bayes-derived distribution assuming the uncertainty of each experimental data point is 5%.

state are highly suppressed. This is because only the charge distribution perturbed by the decay of the second excited state is able to explain the width of the full distribution given that the mean of the equilibrium Gaussian distribution is $\bar{q}_{\text{Bayes}}^{\text{gs}} = 20.79 \pm 0.08$. Figure 6.25 demonstrates that the derived solution is now much more precise than that the one shown in 6.23. It is important to note that the derived mean is now in better agreement with the semi-empirical values (see Table 6.4).

The multipolarity of the transition between the first and second excited states in ^{141}Ba was assigned as $M1$ by Yamamoto et al. [220] based on their measurement of L-shell X-rays created after the internal conversion of the transition, where the intensity of $L_{2,3}$ X-rays is not much larger than the intensity of L_1 X-rays. Therefore, it was concluded that the transition has a negligible $E2$ component since the internal conversion coefficients of L_2 and L_3 subshells should then be much larger than the one for L_1 . There was not enough sensitivity in the measurement to determine the mixing ratio of the transition at that time. A different story can be told through the charge-state distribution since it depends strongly on the distribution of initial vacancies. The distribution of initial vacancies varies significantly from an $M1$ transition to an $E2$. Table 6.6 shows the distribution of initial vacancies created by the transition between the first and second excited states in ^{141}Ba for a Kr-like electron configuration. If the transition is pure $M1$, only 26% of the time an L-shell vacancy is created. In contrast, an L-shell vacancy is created for 77% of the time for a pure $E2$ transition. The table also shows that the distribution of initial vacancies for a mixed $M1 + E2$ transition quickly converges to the one for a pure $E2$ transition.

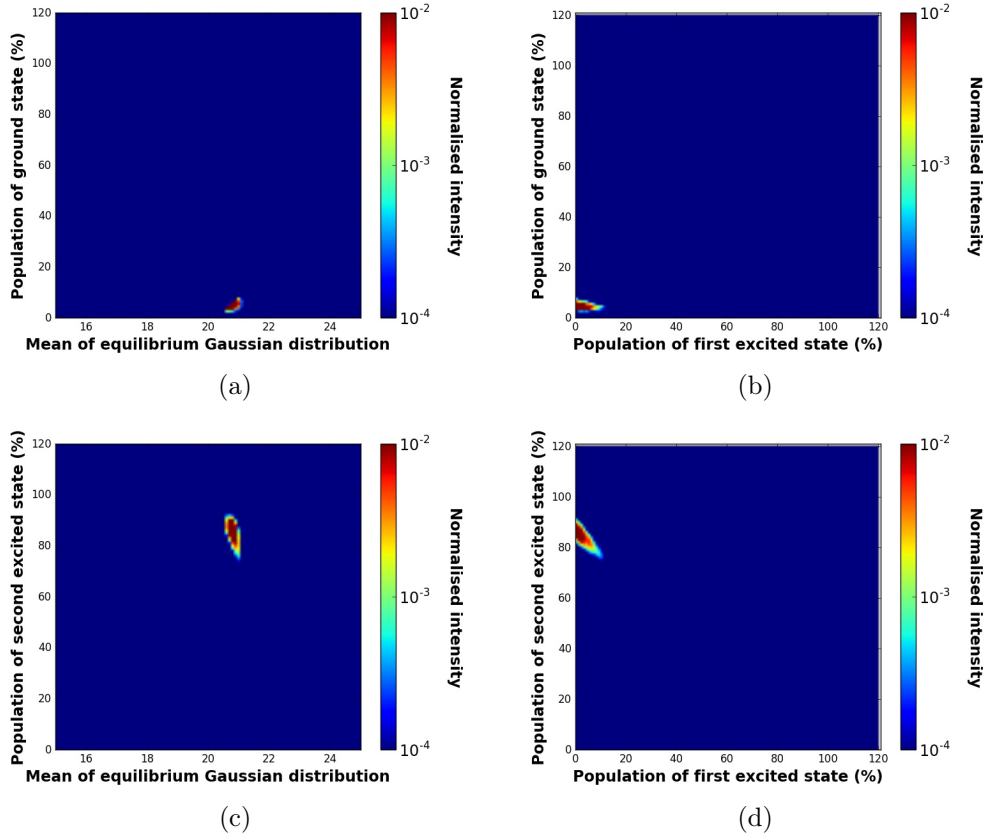


Figure 6.25: Similar to Figure 6.23 but the uncertainty of each experimental data point is assumed to be 5%.

Moreover, the distribution changes drastically from $\delta = 0.00$ to $\delta = 0.03$. This means that the perturbed charge-state distribution could be sensitive to the mixing ratio of this low-energy transition.

Figure 6.26 shows the Bayes-derived perturbed distribution if the 6.469 keV transition is assumed to be a mixed $M1 + E2$ transition with a mixing ratio $\delta = 0.03$ and the uncertainty of each experimental data point is 5%. The figure demonstrates that the increase in the number of vacancies created in the L shell due to the $E2$ multipolarity, increases the intensity of Ba^{25+} and Ba^{26+} and also decreases the mean of the equilibrium Gaussian distribution from $\bar{q}_{\text{Bayes}}^{gs} = 20.79 \pm 0.08$ to $\bar{q}_{\text{Bayes}}^{gs} = 20.18 \pm 0.07$. The changes in the distribution of initial vacancies due to a small mixing of $E2$ lead to a better agreement with the experiment. Nevertheless, the high intensities of Ba^{26+} and Ba^{27+} still could not be explained. It is important to note that these experimental values could possibly be subject to systematic errors incurred during the analysis performed by colleagues at the Laboratory of Subatomic Physics & Cosmology (LPSC). The analysis was initially performed to obtain the integral of the distribution, and therefore the accuracy of each experimental data point was not the biggest concern. Our colleagues at LPSC have agreed that they will pursue a more careful approach in determining the intensity of each charge

Table 6.6: Distribution of initial vacancies created by the 6.469 keV transition between the first and second excited states in ^{141}Ba for Kr-like electron configuration. The L_1 subshell is excluded since its binding energy exceeds the transition energy for Ba^{18+} and higher charged ions.

Atomic shell	Distribution of initial vacancies				
	$M1$	$E2$	$M1+E2$		
			$\delta = 0.01$	$\delta = 0.03$	$\delta = 0.1$
L_2	0.212	0.285	0.230	0.267	0.283
L_3	0.052	0.484	0.160	0.376	0.472
M_1	0.522	0.001	0.391	0.131	0.016
M_2	0.051	0.069	0.055	0.064	0.068
M_3	0.012	0.112	0.037	0.087	0.109
M_4	0.001	0.001	0.001	0.001	0.001
M_5	0.001	0.002	0.001	0.002	0.002
N_1	0.134	0.000	0.100	0.034	0.004
N_2	0.013	0.017	0.014	0.016	0.017
N_3	0.003	0.028	0.009	0.022	0.028

state for future studies.

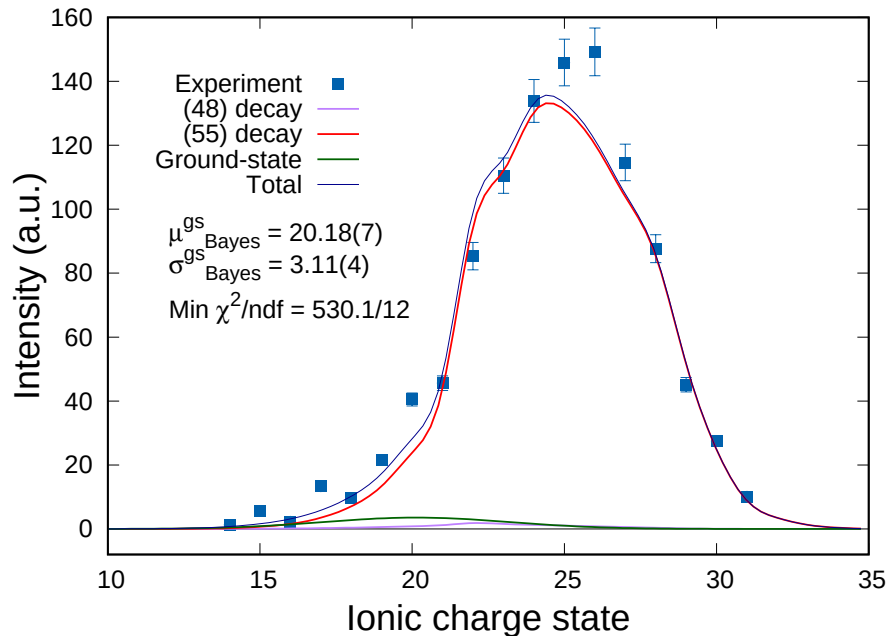


Figure 6.26: Experimental ionic-charge distribution of ^{141}Ba is fitted with a Bayes-derived distribution assuming the transition between the first and second excited states is a mixed $M1 + E2$ transition with a mixing ratio $\delta = 0.03$ and that the uncertainty of each experimental data point is 5%.

Figure 6.27 shows the Bayes-derived correlation density plots for several free parameters that are similar to those shown in Figure 6.25 given that the 6.469 keV transition is assumed to be mixed $M1 + E2$ with $\delta = 0.03$. A slightly smaller solution space is derived

in comparison to the case where the nuclear transition is assumed to be pure $M1$. The derived population of the second excited state is approximately 92%, which dwarfs the populations of the ground state and the first excited state.

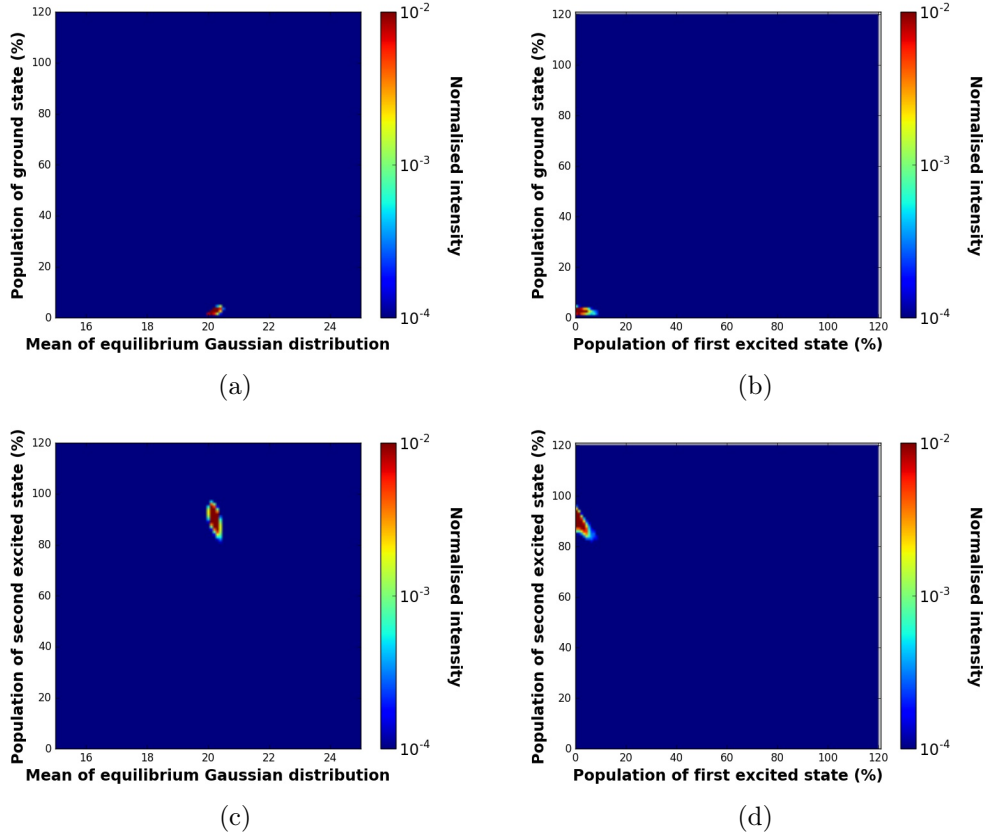


Figure 6.27: Similar to Figure 6.23 but the 6.469 keV transition between the first and second excited states of ^{141}Ba is assumed to be a mixed $M1 + E2$ transition with a mixing ratio, $\delta = 0.03$ and the uncertainty of each experimental data point is assumed to be 5%.

The analysis of the charge-state distribution of ^{141}Ba in this section indicates that the transition between the first and second excited states is expected to be a mixed transition rather than a pure $M1$. This result is supported by shell model calculations which are described in the next section.

Shell model calculations

Shell model calculations for ^{141}Ba were performed (by Prof. Andrew Stuchbery) with the NuShellX@MSU code [221]. As described in Refs. [222, 223], the interactions for the proton-proton space were based on the CD Bonn potential and the proton-neutron and neutron-neutron interactions, designated $jj56pnb$, were obtained from the N^3LO potential. The effective charges e_p and e_n were set by reference to observed $E2$ transitions in ^{134}Te and ^{134}Sn , respectively, resulting in $e_p = 1.56e$ and $e_n = 0.66e$. The effective $M1$ operator applied a correction $\delta g_l(p) = 0.13$ to the proton orbital g factor and quenched the spin

g factors for both protons and neutrons to 70% of their bare values. (The tensor term was ignored.) The effective $M1$ operator is then similar to that of Jakob et al. [224] and in reasonable agreement with that of Brown et al. [222].

Shell model calculation results are compared with the data in ENSDF [113] in Table 6.7. It is evident that the agreement between theory and experiment is not very good. This may in part be due to the experimental data, but it is also the case that ^{141}Ba is on the borderline where collective effects are expected to set in, so the shell model may not be adequate. Nevertheless we observe that the theoretical $M1$ transitions are about an order of magnitude larger than experiment, whereas the $B(E2)$ values are at least of the correct order of magnitude.

Table 6.7: Comparison of the theoretical $B(M1)$ and $B(E2)$ values from shell model calculations with the experimental data in ENSDF [113] for transitions between the ground state ($3/2^-$), the first excited state ($5/2^-$) and the second excited state ($7/2^-$) in ^{141}Ba . I_i and I_f represent the initial and final states of a transition, respectively.

I_i	I_f	$B(M1) (\mu_N^2)$		$B(E2) (e^2 fm^4)$	
		theory	exp	theory	exp
$5/2^-$	$3/2^-$	0.159	0.012(3)	2450	9200(5700)
$7/2^-$	$3/2^-$	-	-	970	390(50)
$7/2^-$	$5/2^-$	0.262	0.056(9)	1530	~ 0

The mixing ratio is given by

$$\delta^2 = 6.97 \times 10^{-5} E_\gamma^2 \frac{B(E2)}{B(M1)}, \quad (6.11)$$

where the transition energy E_γ is in MeV, the $B(E2)$ is in $e^2 \text{fm}^4$, and the $B(M1)$ is in μ_N^2 . If we use the experimental $B(M1)$ with the theoretical $B(E2)$ we get $|\delta| \sim 0.07$, which is of the order inferred from the observed charge-state distribution together with the analysis of the atomic relaxation.

6.4.4 Comparison with semi-empirical calculations

Table 6.8 compares the Bayes-derived mean of the equilibrium Gaussian distribution with semi-empirical values calculated based on Eqs. (6.3)-(6.6). For ^{143}Ba and ^{145}Ba , the mean derived from fitting the experimental data with a Gaussian function is too high relative to the semi-empirical values. Meanwhile, if the vacancy cascades following the decays of nuclear isomers are taken into consideration, the Bayes-derived means for the two isotopes are consistently higher than their semi-empirical counterparts by about 1.2 - 1.5 charge states. Since the semi-empirical calculations are expected to overestimate by roughly 0.5 for Kr-like and Rb-like electron configurations [214], the differences between the Bayes-derived values and their semi-empirical counterparts are brought down to about 0.7 -

1.0. Moreover, several secondary effects, such as shake-off and possible non-ground-state electron configurations, are neglected in the vacancy cascade simulation. By inducing these effects, the differences could be further reduced¹⁴. Therefore, overall a fairly good agreement has been achieved for ^{143}Ba and ^{145}Ba .

Table 6.8: Comparison of Bayes-derived mean of the equilibrium Gaussian distribution with semi-empirical values calculated using equations from Nikolaev and Dmitriev [212], Betz [213] and Schiwietz and Grande [214] (also see Eqs. (6.3)-(6.5)).

Isotope	$\bar{q}_{\text{Bayes}}$	$\bar{q}_{\text{N\&D}}$	$\bar{q}_{\text{Betz}}$	$\bar{q}_{\text{S\&G}}$
^{141}Ba	23.12(73) ^d			
$^{141}\text{Ba}^a$	20.79(8)	19.8(2)	20.1(2)	19.9(1)
$^{141}\text{Ba}^{a,b}$	20.18(7)			
^{143}Ba	20.76(23)			
$^{143}\text{Ba}^a$	20.65(12)	19.2(2)	19.5(2)	19.3(1)
$^{143}\text{Ba}^c$	21.36(5)			
^{145}Ba	20.38(20)			
$^{145}\text{Ba}^a$	20.30(16)	18.8(2)	19.1(2)	18.9(1)
$^{145}\text{Ba}^c$	20.71(5)			

^aThe uncertainty of individual experimental data points is assumed to be 5%.

^bThe 6.469 keV transition between the first and second excited states is assumed to be a mixed $M1+E2$ transition with a mixing ratio, δ , of 0.03.

^cThe experimental charge-state distribution is fitted with a Gaussian distribution.

^d23.12(73) means 23.12 ± 0.73 .

It seems from Table 6.8 that the Bayes-derived means for ^{141}Ba agree well with the semi-empirical calculation if the uncertainty of each experimental data point is set at 5%. This is at odds with results for ^{143}Ba and ^{145}Ba where the Bayes-derived means are higher than their semi-empirical counterparts by at least one charge state. However, Figures 6.25 and 6.27 show that the centroid of the Bayes-derived distribution is in between Ba^{24+} and Ba^{25+} while the centroid of the experimental distribution is in between Ba^{25+} and Ba^{26+} . Therefore, the derived means of the perturbed distributions are probably underestimated in the current analysis. It is important to note that the experimental uncertainties need to be smaller for the Bayesian analysis to generate reliable information, especially for cases where there are two or more nanosecond isomers. For example, the mean of the equilibrium Gaussian distribution is calculated to be 23.12 ± 0.73 for ^{141}Ba , which is much higher than its semi-empirical counterparts, when the experimental uncertainties are large. Moreover,

¹⁴Secondary effects cause slightly more electrons to be ejected in a vacancy cascade. This will increase the separation of the centroids of the equilibrium Gaussian and perturbed distributions. Therefore, the inclusion of secondary effects will reduce the mean of the equilibrium Gaussian distribution derived from a Bayesian analysis.

the derived probability density function of the mean is highly distorted and exhibits two local maxima (see Figure 6.22). This demonstrates the sensitivity of the Bayesian analysis towards experimental uncertainties.

Table 6.9: Comparison of Bayes-derived width of the equilibrium Gaussian distribution with semi-empirical values calculated using equations from Nikolaev and Dmitriev [212], Betz [213] and Schiwietz and Grande [214] (also see Eqs. (6.3)-(6.6)).

Isotope	$\bar{q}_{\text{Bayes}}$	$\bar{q}_{\text{N\&D}}$	$\bar{q}_{\text{Betz}}$	$\bar{q}_{\text{S\&G}}$
^{141}Ba	2.65(16) ^d			
$^{141}\text{Ba}^a$	3.15(6)	2.02(1)	2.02 ^e	1.883(1)
$^{141}\text{Ba}^{a,b}$	3.11(4)			
^{143}Ba	2.96(6)			
$^{143}\text{Ba}^a$	2.91(3)	2.00(1)	2.02	1.879(1)
$^{143}\text{Ba}^c$	3.11(2)			
^{145}Ba	3.40(8)			
$^{145}\text{Ba}^a$	3.41(6)	1.98(1)	2.02	1.877(1)
$^{145}\text{Ba}^c$	3.52(4)			

^aThe uncertainty of individual experimental data points is assumed to be 5%.

^bThe 6.469 keV transition between the first and second excited states is assumed to be a mixed $M1 + E2$ transition with a mixing ratio, δ , of 0.03.

^cThe experimental charge-state distribution is fitted with a Gaussian distribution.

^d2.65(16) means 2.65 ± 0.16 .

^eWidth from Betz's equation is only dependent on the projectile's nuclear charge Z_p (see Eq. (6.4)), so no uncertainty is propagated from the uncertainty in the fragments' kinetic energies, E_0 , before they enter the nickel foil.

Table 6.9 compares the Bayes-derived width of the equilibrium Gaussian distribution with semi-empirical values. The widths calculated using the Bayesian analysis are consistently higher than their semi-empirical counterparts by 1 or more charge state. This may be largely due to the different ways of producing the projectiles. Semi-empirical equations are derived from experimental data based on projectiles generated by accelerators. The kinetic energy distribution of the projectiles created in this way is usually much sharper than its counterpart for fission fragments. Therefore, the semi-empirical width is expected to be smaller than the experimental width of a charge-state distribution measured for fission fragments (see Figure 6.2a). Overall, the derived widths are similar in magnitude, around 3, apart from ^{145}Ba which is close to 3.5. This could be attributed to the differences in kinetic energy distributions which depend on the nuclear charge and mass of the fission fragments. The kinetic energy distribution of fission fragments before they enter the target

nickel foil is difficult to measure and it is usually modified in the target.

6.5 Extraction of isomeric ratio

One of the main goals of studying the perturbed charge-state distribution of fission fragments is to extract isomeric ratios. The isomeric ratio (IR) is defined as follows:

$$\text{IR} = \frac{\sum_i P_{es_i}}{P_{gs} + \sum_i P_{es_i}}, \quad (6.12)$$

where P_{es_i} is the population of an isomeric state i and P_{gs} is the population of the ground state. The quantity IR is useful for the calculations of the spin distribution and excitation energy of a given isotope created in a fission using Monte Carlo codes that simulate the fission fragment de-excitation process such as FIFRELIN [225, 226, 227].

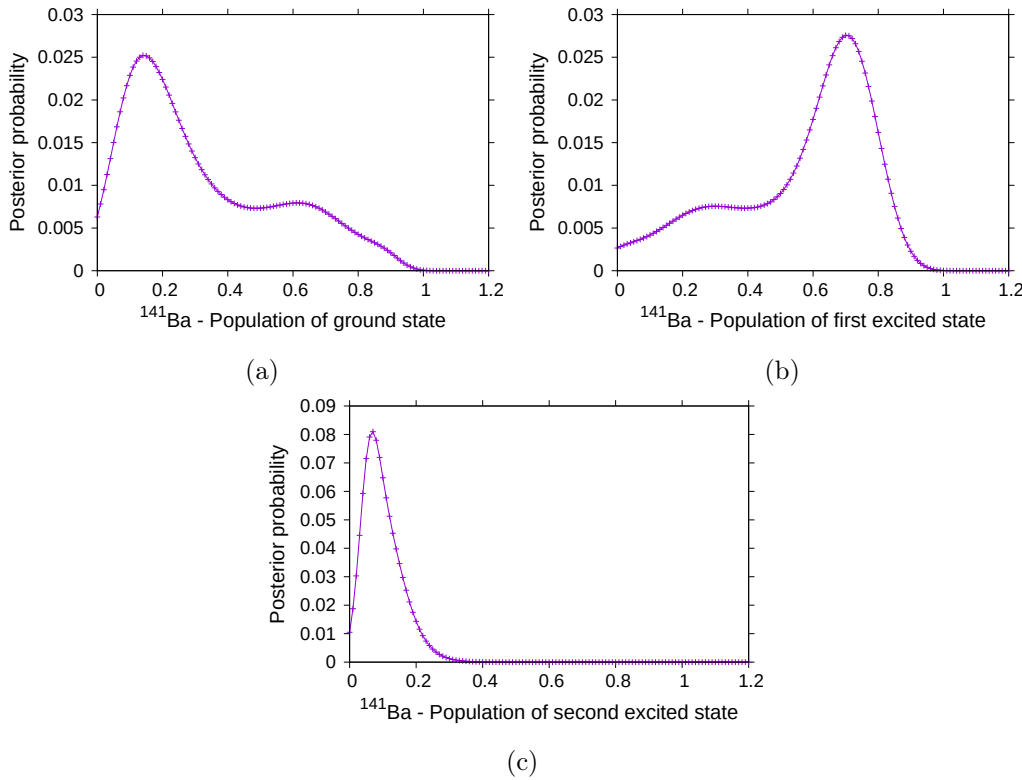


Figure 6.28: Bayes-derived probability density functions for the populations of (a) the ground state, (b) the first excited state and (c) the second excited state in ^{141}Ba .

Once the equilibrium Gaussian and isomeric-perturbed distributions can be separated through the phenomenon of the vacancy cascade, one could determine the population of each component that makes up the entire charge-state distribution. The probability density function (pdf) of the population of a nuclear state could be derived from a Bayesian analysis. Figure 6.28 shows the pdfs for the populations of (a) the ground state, (b) the first excited state and (c) the second excited states in ^{141}Ba derived from a Bayesian

analysis using the experimental data shown in Figure 6.16. Due to the relatively large uncertainties, every distribution shown in the figure has a large width. This is especially true for the ground state and the first excited state where their population distributions span 0 - 1. Based on these probability distributions, one can calculate the pdf of the IR using Eq. (6.12). Here, the pdf of the IR is derived using a Monte Carlo technique. Firstly, a value of P_{gs} is obtained from sampling the distribution shown in Figure 6.28a. Secondly, a value of P_{es1} is selected based on the chosen P_{gs} using the correlation density plot for the populations of the ground state and the first excited state shown in Figure 6.23b. Finally, a value of P_{es2} is acquired from the population distribution of the second excited state *given* the taken values of P_{gs} and P_{es1} , which is extracted from the correlations among the population distributions of all three states. An IR value is then calculated according to Eq. (6.12). The same selection procedure is repeated 1 million times and the resulting distribution is shown in Figure 6.29. The figure indicates that the distribution is highly distorted and non-Gaussian. The mean and standard deviation of the distribution is 0.67 and 0.24, respectively. However, if the experimental uncertainties could be reduced to 5%, then the resulting pdf of the IR would be approximately Gaussian (see Figure 6.30).

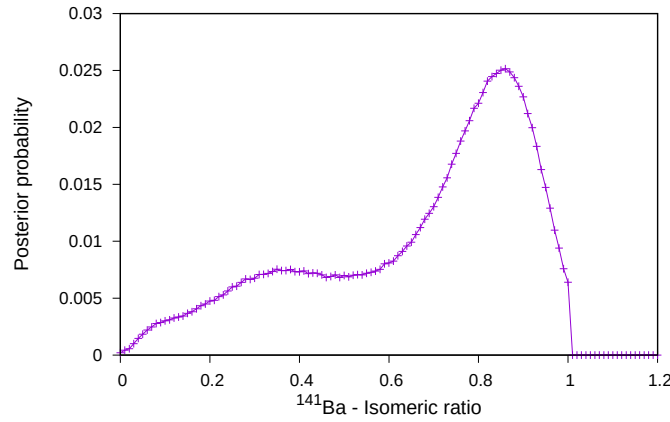


Figure 6.29: The probability density function for the isomeric ratio of ^{141}Ba created in the $^{233}\text{U}(\text{n}_{\text{th}},\text{f})$ reaction.

One could apply the same procedure to ^{143}Ba and ^{145}Ba to obtain the pdf of their IR. Figures 6.31a and 6.31c show these pdfs while Figures 6.31b and 6.31d demonstrate that the pdf of IR could become Gaussian-like if the experimental uncertainties could be reduced. Table 6.10 compares the extracted isomeric ratios from this work with values calculated using a multi-Gaussian fitting from Ref. [211] for the three barium isotopes studied in this chapter. The multi-Gaussian fitting produces values with smaller uncertainties, except for ^{141}Ba which has a highly distorted charge-state distribution. Overall, there is a good agreement between the two methods. This can be attributed to the fact that the perturbed charge-state distribution is not too far from a Gaussian distribution.

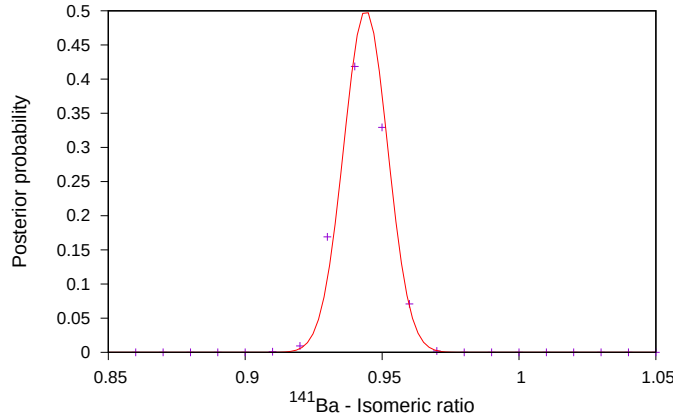


Figure 6.30: Similar to Figure 6.29 but the uncertainty of each experimental data point is assumed to be 5%.

Table 6.10: Comparison of calculated isomeric ratios from this work with results extracted using a multi-Gaussian fitting method for three barium isotopes produced in the $^{233}\text{U}(\text{n}_{\text{th}},\text{f})$ reaction.

Isotope	This work			Multi-Gaussian fitting ^a
	A	B	C	
^{141}Ba	0.67(24)	0.948(8)	0.973(5)	0.90(34)
^{143}Ba	0.42(16)	0.50(9)		0.44(5)
^{145}Ba	0.29(16)	0.37(13)		0.33(3)

A. Based on the original experimental uncertainties provided by colleagues at LPSC.

B. Based on 5% uncertainty.

C. Similar to **B** but the 6.469 keV transition between the first and second excited states in ^{143}Ba is assumed to be a mixed $M1 + E2$ transition with a mixing ratio δ , of 0.03.

^aFrom Ref. [211].

6.6 Discussion

It is well accepted since the 1970s that the perturbed charge-state distributions of fission fragments measured using the LOHENGRIN mass separator are due to the decays of nuclear isomers and the subsequent vacancy cascades which eject more electrons through Auger transitions [12]. However, little effort has been made to fully understand these distributions from underlying principles. This chapter has detailed a novel technique to analyse perturbed ionic-charge distributions using the vacancy-cascade simulations by **BrIccEmis** together with Bayesian inferences. Overall, the derived charge-state distributions are able to explain most of the important features observed in the experiments even though some secondary effects have been neglected in the simulations.

There are two main secondary effects that could have non-negligible impact on the simulated charge-state distributions. The first one is the shake-off process that has been

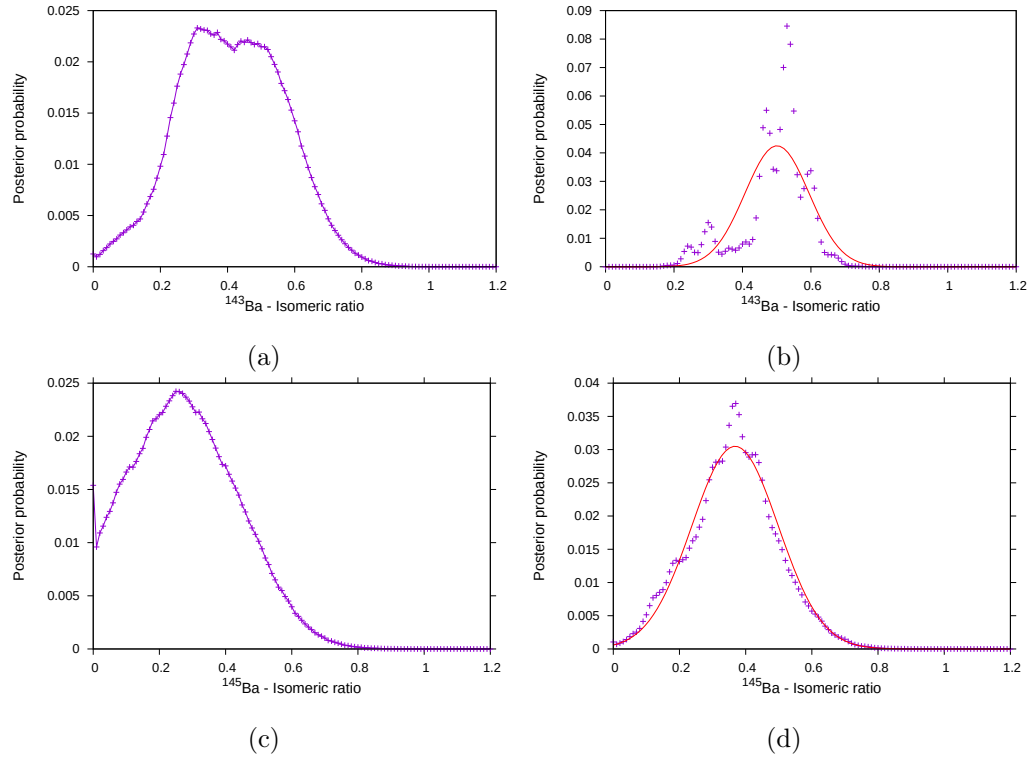


Figure 6.31: Bayes-derived probability density functions for isomeric ratio of (a) ^{143}Ba and (c) ^{145}Ba . (b) and (d) show the derived distributions if the experimental uncertainties are reduced to 5% for ^{143}Ba and ^{145}Ba , respectively.

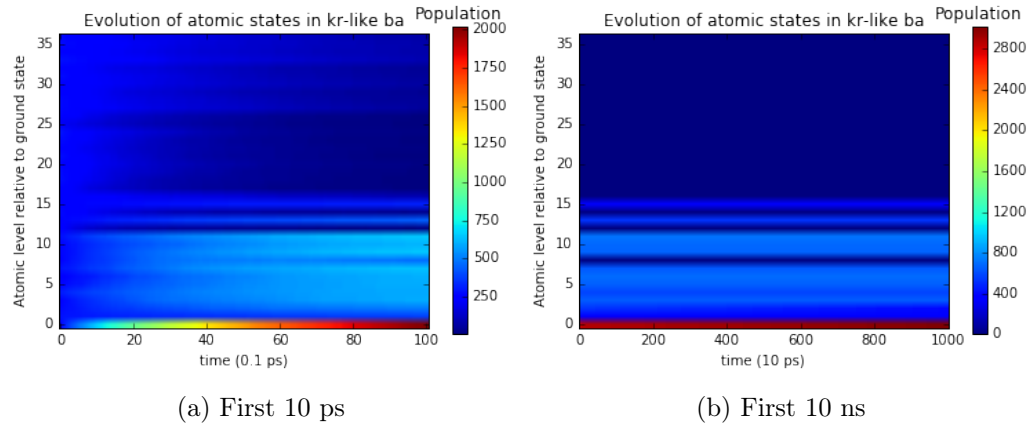


Figure 6.32: Simulated population density plots for Kr-like barium assuming all lowest 37 atomic states have equal probability to be populated at the beginning, i.e. a uniform initial distribution, for the (a) first 10 ps and (b) 10 ns. All plots presented are based on 10,000 simulations therefore the total population at any time point is 10,000.

discussed in Section 6.2. This phenomenon results in a small probability (about 5% per K-shell ionisation in Kr-like barium) to eject an outer-shell electron together with a primary ionisation such as internal conversion of a γ -transition or non-radiative atomic transitions. The shake-off probability decreases as the charge-state increases; thus it is usually more important for K-shell conversion rather than subsequent Auger transitions. Therefore,

extra electrons of about 3-5%, will be emitted following an internal conversion after shake-off is taken into account. The second effect that needs to be carefully investigated in future studies is the impact of non-ground-state electron configurations on the vacancy-cascade simulation. The ground-state electron configuration is used for generating the results presented in this chapter. However, the truth is that the atomic ground state makes up only a small portion of the total population when nanosecond isomers decay. Table A.7 in Appendix A.7 shows the lifetimes of the lowest 37 atomic states in Kr-like barium, i.e. Ba²⁰⁺. Although some higher excited states have picosecond lifetimes, many of the lowest 16 atomic states are, in fact, very long-lived. These states can be populated during the projectile's passage through the nickel target. Based on the atomic transitions among these states that are calculated using the atomic structure package **Grasp2K**¹⁵ [150], one can simulate the decays of these atomic excited states using a Monte Carlo method. Figure 6.32 shows the population density plots at different times. Many short-lived excited states decay in the first 5 ps but the population distribution becomes static after 10 ps up to 10 ns. This indicates that most ions are in the excited atomic states which have more energy to be released in a vacancy cascade, thus emitting more electrons.

Table 6.11: Simulated distribution of atomic states in Kr-like barium after 1 ns. Only the lowest 16 atomic states are presented here as other higher excited states are all short-lived, in which their lifetimes are in the order of 1 ps. Data presented are based on 10,000 simulations.

Atomic state	Normalised intensity
$4s^2 4p^6 \ ^1S_0$	0.2873
$4s^2 4p^5 \ ^2P \ 4d \ ^3P_0^o$	0.0369
$4s^2 4p^5 \ ^2P \ 4d \ ^3P_1^o$	0.0498
$4s^2 4p^5 \ ^2P \ 4d \ ^3F_3^o$	0.0637
$4s^2 4p^5 \ ^2P \ 4d \ ^3D_2^o$	0.0677
$4s^2 4p^5 \ ^2P \ 4d \ ^3F_4^o$	0.0632
$4s^2 4p^5 \ ^2P \ 4d \ ^1D_2^o$	0.0736
$4s^2 4p^5 \ ^2P \ 4d \ ^3D_3^o$	0.0669
$4s^2 4p^5 \ ^2P \ 4d \ ^3D_1^o$	0.0000
$4s^2 4p^5 \ ^2P \ 4d \ ^3F_2^o$	0.0719
$4s^2 4p^5 \ ^2P \ 4d \ ^3D_2^o$	0.0708
$4s^2 4p^5 \ ^2P \ 4d \ ^1F_3^o$	0.0625
$4s^2 4p^5 \ ^2P \ 4d \ ^1P_1^o$	0.0000
$4s^2 4p^5 \ ^2P \ 5s \ ^3P_2^o$	0.0525
$4s^2 4p^5 \ ^2P \ 5s \ ^1P_1^o$	0.0000
$4s^2 4p^5 \ ^2P \ 5s \ ^3P_0^o$	0.0332

Table 6.11 shows the simulated distribution of atomic states in Kr-like barium after 1 ns. A Kr-like barium ion is only in its ground-state electron configuration for about 29% of the time. This could have significant impact on the simulation of resid-

¹⁵As a first approximation, no correlation effect is taken into account, i.e. a Dirac-Fock calculation.

ual charge-state distributions due to isomeric decays. Figure 6.33 compares the simulated residual charge-state distribution using only the ground-state electron configuration with the one simulated using the distribution shown in Table 6.11 for ^{141}Ba and ^{143}Ba . It is important to note that **BrIccEmis** considers a combination of subshell occupation numbers in the jj coupling scheme rather than the individual state listed in Table 6.11 as a unique configuration. So all states listed in Table 6.11 are grouped based on the combination of subshell occupation numbers of their leading terms before the **BrIccEmis** simulation. For example, both $4s^2 4p^5 {}^2P 4d {}^3P_0^o$ and $4s^2 4p^5 {}^2P 4d {}^3D_2^o$ will be categorised in the $4s^2 4p_{1/2}^2 4p_{3/2}^3 4d_{3/2}^1$ group. Therefore, the distribution of initial-electron configurations used in the **BrIccEmis** simulation is 28.73% $4s^2 4p_{1/2}^2 4p_{3/2}^4$ (ground state), 29% $4s^2 4p_{1/2}^2 4p_{3/2}^3 4d_{3/2}^1$, 33.70% $4s^2 4p_{1/2}^2 4p_{3/2}^3 4d_{5/2}^1$, and 8.57% $4s^2 4p_{1/2}^2 4p_{3/2}^3 5s_{1/2}^1$.

The inclusion of non-ground-state electron configurations results in distributions with a more flat peak than those that only consider the ground-state electron configuration. This feature agrees well with the experimentally obtained distributions for both ^{141}Ba and ^{143}Ba (see Figures 6.16 and 6.6). This strongly indicates that non-ground-state configurations could play a significant role in determining the final charge-state distributions and they need to be considered in future studies.

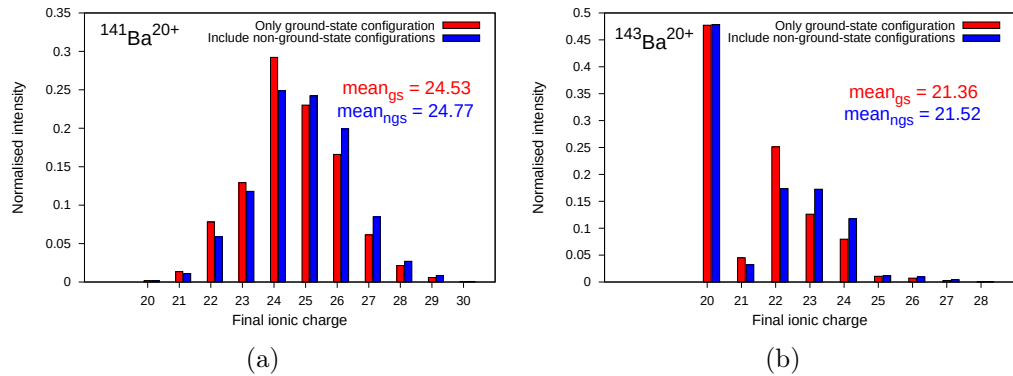


Figure 6.33: Comparison of simulated residual charge-state distributions using only the ground-state electron configuration (red) and the distribution shown in Table 6.11 (blue) for the decays of (a) the second excited state of ^{141}Ba and (b) the first excited state of ^{143}Ba . The 6.469 keV transition between the first and second excited states of ^{141}Ba is assumed to be a mixed $M1 + E2$ transition with a mixing ratio of 0.03.

The residual charge-state distribution has been shown to be sensitive to the calculation of Coster-Kronig transition energies in Chapter 5. Due to the increase in atomic binding energies, most Coster-Kronig transitions become energetically forbidden in highly-ionised systems. Mainly Auger electrons are emitted following inner-shell ionisation by internal conversion at high-charge state. Therefore, the impact of the underestimation of Coster-Kronig transition energies is minimised. Moreover, the accuracy of simulated charge-state distribution is also less affected by the inaccurate Coster-Kronig yields adopted from the EADL [27, 135].

Recent charge-state measurements of highly-ionised iron, germanium and selenium at the Australian National University have shown that atomic closed-shell effects can be pronounced for projectiles produced by accelerators [228]. In these measurements, the intensity of the Ne-like species is higher than the value from semi-empirical calculations by about 20-30%. However, this effect seems to be less significant for charge-state distributions of fission fragments as there is no sudden increase in the intensity of Kr-like barium (Ba^{20+}) for all three isotopes studied in this chapter. This could be attributed to broader energy distribution of fission fragments than those produced by accelerators. It is important to note that the broader energy distribution is also the reason behind the larger width of the equilibrium Gaussian distribution.

The overarching aim of studying the perturbed charge-state distribution of fission fragments is to fully understand all underlying principles that cause the distortion. Once all important principles and factors have been pinpointed, then one could utilise them to probe the nuclear properties of unknown nanosecond isomers, which usually have very low excitation energies, that can only be produced from a fission reaction. It has been shown from the analysis of the perturbed charge-state distribution of ^{141}Ba that the novel technique used in this chapter can be sensitive to the multipolarity of the nuclear transition to a certain extent. The residual charge-state distribution following internal conversion is only dependent on the distribution of initial vacancies rather than the absolute internal conversion coefficients. The distribution of vacancies created by an $M1$ transition is different from the one associated with an $E2$ transition, and the difference will be propagated to the residual ionic-charge distribution. Table 6.6 shows the drastic changes in the initial-vacancy distribution from a pure $M1$ transition to a mixed $M1 + E2$, when the transition energy is low and the mixing ratio is small. By the time the mixing ratio becomes high enough that the initial-vacancy distribution created by a mixed $M1 + E2$ transition is almost the same as the one generated by a pure $E2$, the simulation of the charge-state distribution has lost its sensitivity to the mixing ratio.

6.7 Summary

In summary, a satisfactory agreement has been achieved between the calculated charge-state distributions and the measured ones for three barium isotopes, i.e. ^{141}Ba , ^{143}Ba and ^{145}Ba , produced in the $^{233}\text{U}(\text{n}_{\text{th}},\text{f})$ reaction. This shows theoretically, for the first time, that the phenomenon of the vacancy cascade is responsible for the perturbed charge-state distributions of fission fragments. Several secondary effects, such as shake-off and the existence of atomic excited states, could be crucial in improving the agreement with the experiments. The main weaknesses of the technique developed for this chapter are the dependency on experimental uncertainties, which must be small to obtain reliable results, and the reliance on nuclear structure data from ENSDF [113], which can be incomplete.

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The author thanks Dr. Grégoire Kessedjian and Dr. Christophe Sage of LPSC for providing the experimental charge-state distributions of barium isotopes produced in the $^{233}\text{U}(\text{n}_{\text{th}},\text{f})$ reaction. The author is indebted to Dr. Ulli Köster of ILL for facilitating this study. The author is grateful to Daniel Sean Cusack, who did a third-year undergraduate project under the author's supervision, for developing the Monte Carlo code to simulate the decays of atomic excited states, and Prof. Andrew Stuchbery who suggested the project. Furthermore, the author is thankful to Prof. Stuchbery for his shell model calculations for ^{141}Ba . The author thanks Shao Qi Lim for helping the author to develop the initial concept of the novel technique described in this chapter during her third-year undergraduate project with the author, which was facilitated by Prof. Stuchbery and Dr. Tibor Kibédi. The author is also grateful to Dr. Matthew Reed, who used to be one of the author's colleagues at ANU, for his valuable comments on the increase in lifetime of isomeric states in ions. Last but not least, the author is immensely grateful for the financial support provided by my supervisors Dr. Kibédi, Prof. Stuchbery and Dr. Vos, for my visit to ILL.

Absorbed dose evaluation of Auger-emitting radioisotopes

7.1 Overview

In this chapter, the impact of new decay spectra, which are simulated using the `BrIccEmis` code, on the distribution of dose deposition for Auger-emitting medical radioisotopes is investigated.

7.2 Background

One of the overarching aims of the vacancy-cascade modelling is to provide updated decay spectra of Auger-emitting medical radioisotopes to assist advances in Auger therapy. In fact, the Auger project at the Australian National University was initiated due to the demand for improved decay data of Auger-emitting radioisotopes [11]. Recently, the impact of different Monte Carlo codes on the medically relevant quantities, such as dose point kernels (DPKs - radial dose distribution around a unit source) and S -values (absorbed dose per unit cumulated activity), has been extensively studied [229, 230]. Moreover, the effects of cell geometry on these quantities have also been reported [160, 186]. However, the impact of the decay spectra of Auger-emitting radioisotopes has never been studied in detail. Previous studies mostly adopted the decay spectra reported by Howell [38], in which the non-radiative transition energies were calculated based on the $Z/Z + 1$ rule, or those generated from the Medical Internal Radiation Dose (MIRD) RADTABS program [81] that adopted the terminology of the `RELAX` program written by Cullen [84] and neglects the effect of spectator vacancies. Cullen's program calculates the transition energies based on neutral binding energies using Eqs. (4.1) and (4.7).

In this joint study with Dr. Nadia Falzone of the University of Oxford, the impact of the input decay spectra on the dosimetric evaluation of Auger-emitting radioisotopes is investigated. The DPKs and S -values derived from the `BrIccEmis`-based spectra are compared with those calculated using the RADTABS-based spectra [81]. In addition, the differences between the isolated-atom and condensed-phase assumptions (see Section 3.4.7

for the definition of these assumptions), which concern the fate of valence vacancies during the vacancy cascade, are also studied in detail. The ionisation potential associated with the residual ion, ΔV , in Eq. (3.29), as well as the neutralisation energy, E_N , in Eq. (3.31), have been left out of all calculations in this study. Realistic modelling of the charge neutralisation at the decay site is required to properly account for ΔV and E_N . Such a neutralisation event is beyond the scope of this thesis but its impact on the overall energy deposition by Auger-emitting radioisotopes will be discussed.

Supplementary material

Supplementary figures and tables of this chapter are provided in Appendix B.

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Statement of authorship

Absorbed dose evaluation of Auger electron-emitting radionuclides: impact of input decay spectra on dose point kernels and S -values

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Lee, B.Q.(Candidate)

Generated decay spectra; interpreted simulated results from PENELOPE; joint-wrote manuscript and produced figures.

Falzone, N.

Performed PENELOPE simulations; calculated S -values and DPKs; joint-wrote manuscript and produced tables; acted as corresponding author.

Fernández-Varea, J.M.

Provided software support; manuscript editing.

Kartsonaki, C.

Manuscript editing.

Stuchbery, A.E.

Manuscript editing.

Kibédi, T.

Manuscript editing.

Vallis, K.A.

Manuscript editing.

Absorbed dose evaluation of Auger electron-emitting radionuclides: impact of input decay spectra on dose point kernels and S-values

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Abstract

The aim of this study was to investigate the impact of decay data provided by the newly developed stochastic atomic relaxation model *BrIccEmis* on dose point kernels (DPKs - radial dose distribution around a unit point source) and S-values (absorbed dose per unit cumulated activity) of 14 Auger electron (AE) emitting radionuclides, namely ⁶⁷Ga, ^{80m}Br, ⁸⁹Zr, ⁹⁰Nb, ^{99m}Tc, ¹¹¹In, ^{117m}Sn, ¹¹⁹Sb, ¹²³I, ¹²⁴I, ¹²⁵I, ¹³⁵La, ^{195m}Pt and ²⁰¹Tl.

Radiation spectra were based on the nuclear decay data from the medical internal radiation dose (MIRD) RADTABS program and the *BrIccEmis* code, assuming both an isolated-atom and condensed-phase approach. DPKs were simulated with the PENELOPE Monte Carlo (MC) code using event-by-

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event electron and photon transport. *S*-values for concentric spherical cells of various sizes were derived from these DPKs using appropriate geometric reduction factors.

The number of Auger and Coster–Kronig (CK) electrons and x-ray photons released per nuclear decay (yield) from MIRD-RADTABS were consistently higher than those calculated using *BrIccEmis*. DPKs for the electron spectra from *BrIccEmis* were considerably different from MIRD-RADTABS in the first few hundred nanometres from a point source where most of the Auger electrons are stopped. *S*-values were, however, not significantly impacted as the differences in DPKs in the sub-micrometre dimension were quickly diminished in larger dimensions.

Overestimation in the total AE energy output by MIRD-RADTABS leads to higher predicted energy deposition by AE emitting radionuclides, especially in the immediate vicinity of the decaying radionuclides. This should be taken into account when MIRD-RADTABS data are used to simulate biological damage at nanoscale dimensions.

Keywords: Auger electron-emitters, single-cell dosimetry, *S*-values, dose point kernels

 Supplementary material for this article is available [online](#)

(Some figures may appear in colour only in the online journal)

1. Introduction

When evaluating the efficacy of novel radionuclide constructs used in diagnostic and therapeutic nuclear medicine, it is often necessary to estimate the absorbed dose from intracellularly localised radionuclides at a single-cell level. To enable these calculations, the Medical Internal Radiation Dose (MIRD) Committee (Goddu *et al* 1997) has compiled comprehensive tables of cellular *S*-values (absorbed dose per unit cumulated activity). An important aspect of cellular *S*-value calculations is the adopted decay radiation spectrum. The radiation spectra used in the MIRD compilation of *S*-values (Goddu *et al* 1997) were taken from the work of Eckerman *et al* (1993), which is based on the nuclear structure data from the Evaluated Nuclear Structure Data Files (ENSDF). More recently, MIRD (Eckerman and Endo 2008a) published up-to-date tabulations of radiation spectra for 333 radionuclides. In the calculation of atomic relaxation spectra emitted after electron capture (EC) or internal conversion (IC), several assumptions were made in the MIRD tabulations partly due to computational limits at the time and the paucity of experimental data. These include the use of binding energies of neutral atoms and neglect of spectator vacancies that could potentially lead to inaccurate estimates of the emissions of low-energy x-rays and AE. An improved computational route is therefore required to address the limitations of the MIRD committee data as suggested by the International Nuclear Data Committee of the International Atomic Energy Agency (Nichols *et al* 2011).

The nuclear structure data from ENSDF is used to evaluate the distribution of initial vacancies, which determines the initial atomic state before the cascade simulation commences. Furthermore, two approaches toward the fate of valence vacancies produced during the cascade have been considered in the literature, namely the isolated-atom and condensed-phase conditions (Howell 1992, Pomplun 2000, Stepanek 2000). The distinction between these two

approaches is the assumption regarding the neutralisation of any valence vacancy during a cascade. In the condensed-phase approach, charge transfer between the environment and the valence shell is assumed to take place during the cascade, leaving the atom completely neutralised at the end of the cascade process, while with the isolated-atom approach, the atom becomes ionised. Detailed comparison of isolated-atom and condensed-phase approximations and the impact of partial neutralisation have been discussed by Charlton *et al* (1983). There is no consensus in the literature as to which approach should be adopted, due to the lack of experimental evidence for the time-scale of electron transfer from DNA, proteins or a liquid water environment to an Auger-emitter. For example, radiation spectra in the MIRD tabulations (Howell 1992, Eckerman and Endo (2008a) were calculated using the condensed-phase approach, while Pomplun (2000) adopted the isolated-atom approximation. This is one of the main factors contributing to the variability in published emission spectra of selected Auger-emitters (Howell 1992, Pomplun 2000, Stepanek 2000, Eckerman and Endo 2008a). Recently Lee *et al* (2012, 2016) published a stochastic atomic relaxation cascade (referred to as vacancy cascade) model called *BrIccEmis* to simulate the AE and x-ray spectra of elements up to Fermium, $Z = 100$. In the present work, emission spectra based on both approaches were calculated using *BrIccEmis* for detailed comparison.

The main aim of this study is therefore to investigate the impact of new decay data provided by *BrIccEmis* (Lee *et al* 2012, 2016) on dose point kernels (DPKs) and S -values of 14 Auger electron-emitting radionuclides⁹, namely ^{67}Ga (EC), $^{80\text{m}}\text{Br}$ (IT), ^{89}Zr (EC β^+), ^{90}Nb (EC β^+), $^{99\text{m}}\text{Tc}$ (IT β^-), ^{111}In (EC), $^{117\text{m}}\text{Sn}$ (IT), ^{119}Sb (EC), ^{123}I (EC), ^{124}I (EC β^+), ^{125}I (EC), ^{135}La (EC), $^{195\text{m}}\text{Pt}$ (IT) and ^{201}Tl (EC) (Eckerman and Endo 2008a).

2. Materials and methods

2.1. Stochastic cascade model

A detailed description of *BrIccEmis* has been published elsewhere (Lee *et al* 2012, 2016). The initial vacancy distributions due to EC and internal conversion (IC) of γ -emission of 14 radionuclides were evaluated using the nuclear structure data in the ENSDF format, available from www.nndc.bnl.gov/ensdf/. The energies of all possible radiative and non-radiative atomic transitions to fill a vacancy were checked during the cascade and any energetically forbidden transitions were discarded before a propagation step. A transition energy was set equal to the difference between the total energies of the atom before and after a transition, calculated using the RAINE code (Band *et al* 2002), which is based on the relativistic Dirac–Fock self-consistent method. The atomic transition probabilities were taken from the Evaluated Atomic Data Library (EADL) (Perkins *et al* 1991). Since all probabilities from EADL were calculated for singly-ionised systems, the empirical correction of Krause and Carlson (Krause and Carlson 1967) was applied to modify probabilities according to the actual number of electrons available in the participating atomic subshells. Radiation spectra are provided as supplementary data (supplementary figures 1–7 (stacks.iop.org/PMB/62/2239/mmedia)).

2.2. Dose point kernels

Electron and x-ray transport was performed by event-by-event Monte Carlo (MC) simulations with the general-purpose MC code PENELOPE (Salvat *et al* 2011). MC transport of the complete radiation spectra based on the unabridged decay data from MIRD tabulation

⁹Decay mode: β^- beta minus, β^+ positron emission/beta plus, EC electron capture, and IT internal transition.

(MIRD-RADTABS software Ver. 2.2) (Eckerman and Endo 2008a), as well as the unabridged spectra generated by *BrIccEmis* based on the isolated-atom and condensed-phase approximations were used for all radionuclides considered. In the case of ^{90}Nb , $^{117\text{m}}\text{Sn}$, and ^{124}I , the abridged AE + CK + IC spectra were utilised as their unabridged spectra were not provided in MIRD-RADTABS. Since ^{135}La was not included in MIRD-RADTABS, the abridged AE + CK + IC spectrum of ^{135}La was adopted from the International Commission on Radiological Protection (ICRP) Publication 107 (Eckerman and Endo 2008b).

An isotropic point source was placed in an infinite liquid water medium (mass density $\rho = 1 \text{ g cm}^{-3}$) and the mean absorbed dose from the emitted ionising radiation (AE, CK, IC, β^+ particles and x-rays) was recorded in 1 nm-thick spherical shells around the decay site. Mean absorbed doses were tallied up to a radial distance of 50 μm from the point source. Electron and photon cut-off energies were set at 50 eV. Typically 2×10^9 primary particles were simulated in each run.

2.3. S-values from simulated DPKs

Cell (R_C) and nucleus (R_N) radii combinations for concentric spherical cells as tabulated by MIRD (Goddu *et al* 1997) were expanded to include larger cell sizes (up to $R_C = 12 \mu\text{m}$ and $R_N = 11 \mu\text{m}$). S -values ($\text{Gy/Bq} \cdot \text{s}$) were calculated as previously described (Falzone *et al* 2015). Briefly, taking the cell nucleus as the target, S -values were calculated from the simulated DPKs to determine contributions from the nucleus ($N \leftarrow N$), the cytoplasm ($N \leftarrow \text{Cy}$) or the cell surface ($N \leftarrow \text{CS}$) as;

$$S(\text{T} \leftarrow \text{S}) = \frac{1}{m_T} \int_0^\infty dr 4\pi r^2 \rho \psi_{\text{T} \leftarrow \text{S}}(r) D(r), \quad (1)$$

where the DPK $D(r)$ is the mean absorbed dose scored in a 1 nm-thick spherical shell at a distance r from the point source, ρ is the mass density, m_T is the mass of the target volume and $\psi_{\text{T} \leftarrow \text{S}}(r)$ are the geometric reduction factors. $\psi_{\text{T} \leftarrow \text{S}}(r)$ denotes the probability of a randomly directed vector of length r that starts from a random point within the source region (S) and ends within the target region (T) (Goddu *et al* 1997); these factors become analytical functions in the case of concentric nucleus and cell configurations.

Tabulated MIRD S -values (Goddu *et al* 1997) were calculated with DPKs assuming straight line trajectories and the continuous slowing down approximation. To enable a direct comparison of S -values derived from MC DPKs, thus just taking the differences in spectra (Eckerman and Endo 2008a, 2008b) versus *BrIccEmis* and not calculation method into account, we compared S -values derived from event-by-event MC simulations as previously published (Falzone *et al* 2015) with the *BrIccEmis*-derived S -values.

3. Results

3.1. AE and x-ray yields

A summary of AE, CK, and x-ray yields, where yield is defined as the number of a particular radiation type released per nuclear decay, comparing MIRD-RADTABS (Eckerman and Endo 2008a) and *BrIccEmis* data is provided in table 1. AE and CK yields calculated using *BrIccEmis* are generally lower than the values provided by MIRD-RADTABS even though the condensed-phase approximation is in better agreement with MIRD-RADTABS for several radionuclides. Most of the discrepancies come from the differences in yields of outer-shell

Table 1. Comparison of AE + CK and x-ray yields from *BrIccEmis* and MIRD-RADTABS (Eckerman and Endo 2008a).

Nuclide	Yield of AE + CK			Yield of x-rays		
	<i>BrIccEmis</i> isolated	<i>BrIccEmis</i> condensed	MIRD	<i>BrIccEmis</i> isolated	<i>BrIccEmis</i> condensed	MIRD
⁶⁷ Ga	4.56	4.85	4.96	0.57	0.57	6.87
^{80m} Br	7.33	8.96	9.60	0.82	0.81	12.0
⁸⁹ Zr	4.58	6.88	9.45	0.51	0.51	10.7
⁹⁰ Nb	4.33	6.88	8.77	0.52	0.52	10.1
^{99m} Tc	3.52	4.39	4.41	0.08	0.08	5.58
¹¹¹ In	5.84	7.17	7.43	0.91	0.91	9.50
^{117m} Sn	5.38	5.72	14.2	0.80	0.76	16.1
¹¹⁹ Sb	10.0	14.4	23.7	0.97	0.88	26.4
¹²³ I	7.38	12.3	13.7	1.09	0.97	15.8
¹²⁴ I	5.04	8.38	9.17	0.72	0.65	10.6
¹²⁵ I	11.9	20.0	23.0	1.76	1.57	26.5
¹³⁵ La	7.44	10.9	12.3 ^a	0.93	0.81	14.3 ^a
^{195m} Pt	20.4	30.6	36.6	1.21	1.20	41.1
²⁰¹ Tl	10.9	18.8	20.9	1.53	1.45	24.3

^a ICRP Publication 107 (Eckerman and Endo 2008b).

non-radiative transitions (M-shell and above). There are significant differences between the condensed-phase approximation and MIRD-RADTABS data for ^{117m}Sn and ¹¹⁹Sb. In the case of ^{117m}Sn, the main source of the discrepancy is an unassigned non-radiative transition in Sn at 11.33 eV with a yield of 7.6. For ¹¹⁹Sb, in which the vacancy cascade occurs in the atomic system of Sn following EC decay, MIRD-RADTABS data included 9.3 non-radiative transitions that involve the O₃-subshell, which is only occupied in the excited atomic states of Sn. *BrIccEmis* ignored these transitions because the probability of finding a Sn atom in its excited state at human body temperature is extremely low (<0.001) according to the Boltzmann distribution (Kramida *et al* 2015).

Both condensed-phase and isolated-atom approximations result in similar x-ray yields. However, both approximations lead to significantly lower x-ray yields than MIRD-RADTABS data because, in the latter, energy released during the neutralisation of a valence vacancy is assigned to radiative recombination, thus producing a very high number of ‘free-bound low-energy x-rays’. This approach could be misleading as femtosecond spectroscopy has revealed that vibrational relaxation is competing with electronic relaxation in electron transfer at a molecular level within the timescale of the vacancy cascade (Anderson and Lian 2004, Frischkorn and Wolf 2006). The correct treatment of the neutralisation of valence vacancies, which is dependent on the environment, is currently beyond the scope of *BrIccEmis*. Hence, the code only reports bound–bound radiative transitions.

3.2. Dose point kernels

3.2.1. Electrons. DPKs of ¹¹¹In, ¹²⁴I, ¹²⁵I and ²⁰¹Tl derived from MIRD-RADTABS data (Eckerman and Endo 2008a) are shown in the upper panels of figure 1 in which the contributions from AE, CK, IC, and β^+ are included. The shoulders in the DPKs are a result of the energy deposition contributions of the aforementioned particulate radiations. The DPKs derived from *BrIccEmis* data, which are given as ratios to the DPKs calculated using MIRD-RADTABS

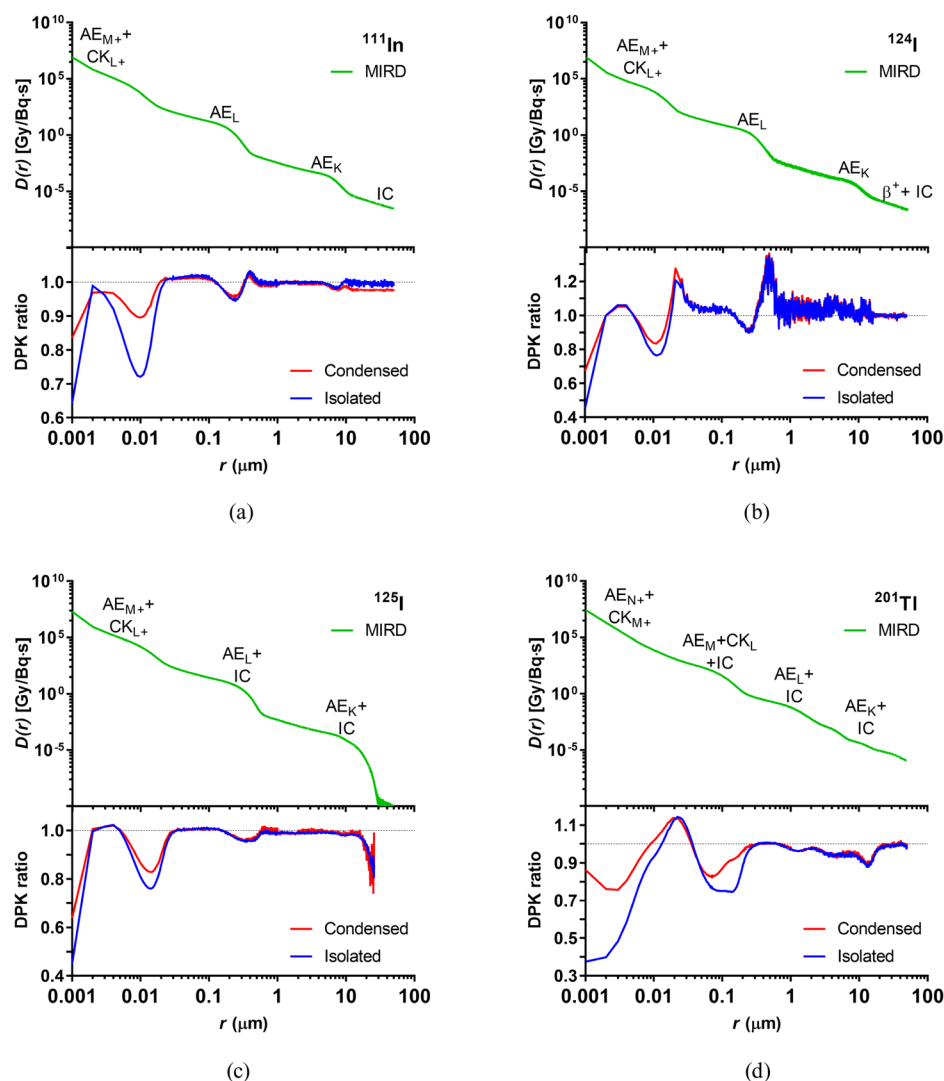


Figure 1. DPKs (Gy/Bq · s) for the AE + CK + IC spectra from MIRD-RADTABS (Eckerman and Endo 2008a) of (a) ^{111}In , (b) ^{124}I , (c) ^{125}I , and (d) ^{201}Tl plotted on the top panel as a function of distance from a point source. DPK ratios, which are defined as $\text{DPK}_{\text{BrIccEmis}} / \text{DPK}_{\text{MIRD-RADTABS}}$, comparing dose distribution from MIRD-RADTABS data with spectra generated from *BrIccEmis* using condensed-phase and isolated-atom approximations plotted on the bottom panel.

data, are shown in the lower panels of figure 1. DPKs of the remaining radionuclides are supplied as supplementary figures (supplementary figures 8–10).

In general, little discrepancy exists between the condensed-phase and isolated-atom approximations at large radii. The two deviate at small radii as this is the domain where low-energy AE and CK electrons, which are emitted from M and higher shells towards the final stages of the vacancy cascade, deposit their energies. The radial interval where the two approaches deviate from each other is radionuclide dependent; the deviation begins at about

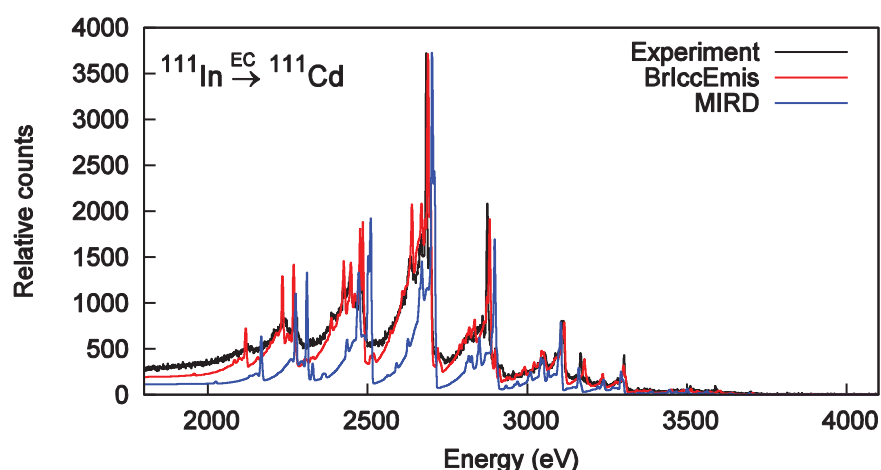


Figure 2. Comparison of Auger spectra from *BrIccEmis* and MIRD-RADTABS (Eckerman and Endo 2008a) with experimental data (Yakushev *et al* 2005) within the energy range from 1800 to 3600 eV. There is a shift towards higher energies in the MIRD-RADTABS data. Both *BrIccEmis* and MIRD-RADTABS data were folded with the same approximate theoretical lineshape. Theoretical spectra were scaled to match the maximum intensity of the main L-Auger group at about 2700 eV.

200 nm and 300 nm in ^{111}In and ^{201}Tl , respectively (figures 1(a) and (d)). DPKs derived from the *BrIccEmis* data differ from DPKs calculated using MIRD-RADTABS data up to the maximum range of L-Auger electrons. At large radii, DPKs derived from all three approaches are alike for ^{111}In and ^{124}I . For ^{125}I and ^{201}Tl (figures 1(c) and (d)), *BrIccEmis*-based DPKs are dissimilar from MIRD-based DPKs due to the differences in IC yields. Besides, many low-energy AE and CK electrons are stopped at the decay site due to the cut-off energy of 50 eV in electron transport, resulting in the discontinuities in DPKs.

There are noticeable oscillations in the DPK plots (lower panels). These are due to the differences between MIRD-RADTABS and *BrIccEmis* AE and CK electron energies emitted from the L-shell and above. Figure 1(a) shows that *BrIccEmis*-based DPKs are slightly higher than MIRD-based DPKs near the maximum range of the L-Auger electrons. The latter becomes dominant at smaller radii around the median range of the L-Auger group before the former overtakes it again near the maximum range of the M-Auger group. This is a result of an energy shift in the L-Auger spectrum from MIRD-RADTABS. Figure 2 compares spectra from *BrIccEmis* (condensed-phase and isolated-atom approximations are the same for L-Auger) and MIRD-RADTABS with experimental data (Yakushev *et al* 2005). The comparison shows an energy shift towards higher energies in MIRD-RADTABS data while *BrIccEmis* data are in good agreement with the experiment within the given energy range. Similar behaviour is found at smaller radii where AE emitted from higher atomic shells are dominant. This is prevalent in all other radionuclides except ^{90}Nb , $^{117\text{m}}\text{Sn}$, ^{124}I , and ^{135}La in which the abridged spectra from MIRD-RADTABS were used (figure 1(b) and supplementary figures 8(b), 9(b) and 10(a)).

In cases where MIRD-RADTABS's abridged spectra (^{90}Nb , $^{117\text{m}}\text{Sn}$ and ^{124}I), which are sometimes called 'average spectra' were used, *BrIccEmis*-based DPKs are significantly higher than MIRD-based DPKs near the maximum range of an Auger group. Around the median range of a group, the latter surpasses the former. Since transition intensity is concentrated at the mean energy of a group in the abridged spectra, *BrIccEmis*-based DPKs eventually overtake MIRD-based DPKs at smaller radii.

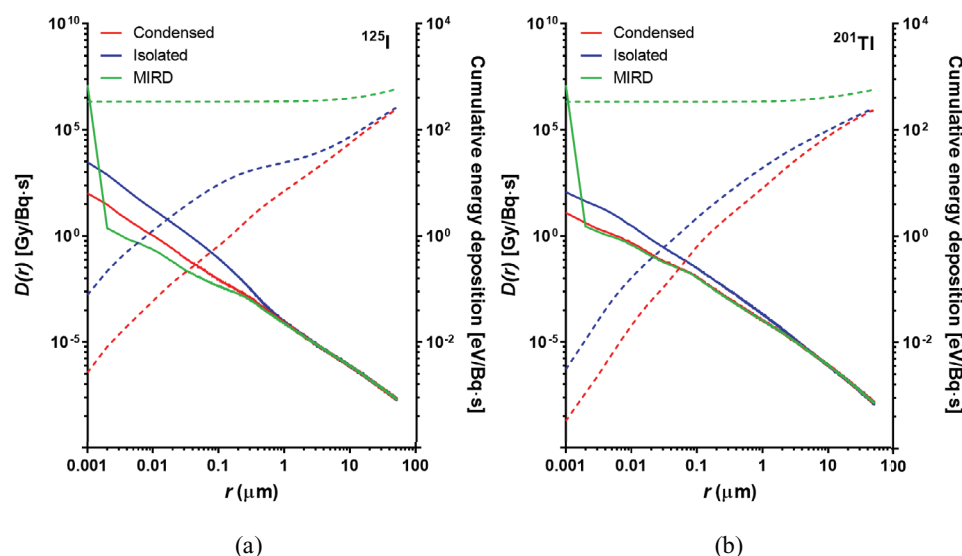


Figure 3. DPKs (Gy/Bq · s) for the x-ray spectra of (a) ^{125}I and (b) ^{201}Tl from *BrIccEmis* and MIRD-RADTABS (Eckerman and Endo 2008a) plotted on the left y-axis as a function of distance from a point source (solid lines). Cumulated energy deposition (eV/Bq · s) as a function of distance from the point source is plotted on the right y-axis (dashed lines).

3.2.2. X-rays. DPKs of ^{125}I and ^{201}Tl for all three x-ray spectra (*BrIccEmis*' condensed-phase and isolated-atom assumptions and MIRD-RADTABS) are depicted in figure 3. The DPKs derived from the isolated-atom approximation are significantly higher than the other two DPKs over the range 2 nm–1 μm for both radionuclides. This is a consequence of the higher yields of low-energy x-rays (25–500 eV) from the isolated-atom approximation (supplementary figures 1–7). The contributions from the MIRD-RADTABS x-ray spectra, for distances smaller than 2 nm from the point source, exceed those of the x-ray spectra from the *BrIccEmis* code. The so-called free-bound x-rays reported in MIRD-RADTABS data are very low in energy (<50 eV). Thus, they were absorbed at the site of decay in the PENELOPE simulations resulting in the spikes in x-ray DPKs. This is the same in the x-ray DPKs derived for all other radionuclides (supplementary figures 11–13).

3.3. Cellular *S*-values

Comparisons of *S*-values derived from MIRD-RADTABS data (Eckerman and Endo 2008a) as previously published (Falzone *et al* 2015) with those presently calculated from the *BrIccEmis* spectra are shown in supplementary tables 1–6 (except ^{124}I and ^{135}La , which were not reported in the previous publication). MC calculated *S*-values from *BrIccEmis* data are generally in good agreement with the values derived from MIRD-RADTABS data, even though the difference between the data is significant in the immediate vicinity of the decay site (see figure 1), the calculations of *S*-values at a cellular scale diminish the contributions of nanoscale dose where Auger electrons dominate. For low atomic number radionuclides (^{67}Ga and $^{80\text{m}}\text{Br}$), differences are less than 1% for $S(\text{N} \leftarrow \text{CS})$ and $S(\text{N} \leftarrow \text{Cy})$ values and less than 3% for $S(\text{N} \leftarrow \text{N})$ values (supplementary table 1). However, with increasing atomic number, the variation in $S(\text{N} \leftarrow \text{N})$ contributions from the various spectra data increase markedly, with the

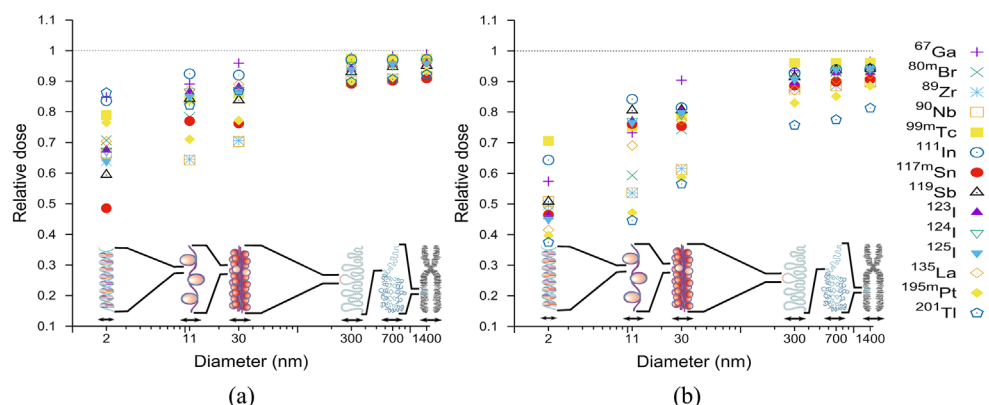


Figure 4. Relative dose, $S_{BrIccEmis} / S_{MIRD-RADTABS}$, comparing energy deposition based on MIRD-RADTABS data (Eckerman and Endo 2008a) and *BrIccEmis* data calculated using condensed-phase (a) and isolated-atom (b) approximations. Relative dose is given as the ratio of energy deposited in spheres with diameters representing different DNA condensation states, i.e. DNA double helix (2 nm), DNA wrapped around histones (chromatin, 11 nm), chromatin fibre of packed nucleosomes (30 nm), chromosome section in extended form (300 nm), condensed section of chromosome (700 nm), and entire mitotic chromosome (1400 nm).

isolated-atom assumption contributing to differences up to 18% (for ^{201}Tl) for small R_C and R_N (supplementary table 6) while the condensed-phase assumption differs by less than 10% from MIRD-RADTABS data for all radionuclides (supplementary tables 1–6). In summary, a decrease in S -values is more prominent for small cell sizes, as the differences between MIRD-RADTABS and *BrIccEmis* data are most noticeable for electrons emitted from the L-shell and above which usually have a maximum range of less than 1 μm . This point is further illustrated in figure 4, dose point kernels of MIRD-RADTABS and *BrIccEmis* data can be significantly different at close range from the source whereas S -values remain similar at the cell level. Over shorter dimensions at a DNA scale, MIRD-RADTABS data would overestimate the calculated dose as compared to *BrIccEmis* data.

As S -values have not been supplied for ^{135}La in the MIRD monograph of cellular S -values (Goddu *et al* 1997), we have included these calculations for both the isolated-atom and condensed-phase assumptions (table 2). IC electrons contributed at most 0.02% when R_N was large relative to R_C in ^{135}La . Similarly, S -values of ^{124}I not included in our previous work (Falzone *et al* 2015), are now derived from *BrIccEmis* data and are shown in supplementary table 7. β^+ particles contribute up to 12% of the S -values while IC, similar to the case of ^{135}La , contribute at most 0.18% when R_N is large relative to R_C .

Where available, a comparison of S -values ($R_C = 5 \mu\text{m}$, $R_N = 4 \mu\text{m}$) with those derived from other computational codes in the literature is also presented (table 3). For overlapping source and target configurations, *BrIccEmis* data compare well with MC4 (Bousis *et al* 2010), ETRACK (Ftániková and Böhm 2000) and GEANT4 (Šefl *et al* 2015) data, with differences <10%, apart from ^{201}Tl . However, differences greater than 40% between *BrIccEmis* and ETRACK data are seen for $S(N \leftarrow CS)$ configurations. All aforementioned publications adopted the average spectra from Howell (1992) which are similar to the abridged spectra of MIRD-RADTABS. From the comparisons of S -values derived from *BrIccEmis* and MIRD-RADTABS for ^{90}Nb and ^{117m}Sn , differences due to input decay spectra are expected not to be

Table 2. Cellular S -values (Gy/Bq · s) for ^{135}La , calculated both with the condensed-phase and isolated-atom assumptions of the *BrIccEmis* code.

R_N (μm)	R_C (μm)	Condensed-phase spectra			Isolated-atom spectra		
		$S(N \leftarrow N)$	$S(N \leftarrow \text{Cy})$	$S(N \leftarrow \text{CS})$	$S(N \leftarrow N)$	$S(N \leftarrow \text{Cy})$	$S(N \leftarrow \text{CS})$
1	3	1.40×10^{-1}	1.11×10^{-3}	1.65×10^{-4}	1.33×10^{-1}	1.10×10^{-3}	1.62×10^{-4}
2	3	1.91×10^{-2}	7.97×10^{-4}	1.76×10^{-4}	1.83×10^{-2}	7.88×10^{-4}	1.72×10^{-4}
2	4	1.91×10^{-2}	3.59×10^{-4}	1.09×10^{-4}	1.83×10^{-2}	3.54×10^{-4}	1.07×10^{-4}
3	4	5.92×10^{-3}	3.36×10^{-4}	1.16×10^{-4}	5.66×10^{-3}	3.32×10^{-4}	1.13×10^{-4}
2	5	1.91×10^{-2}	2.20×10^{-4}	7.99×10^{-5}	1.83×10^{-2}	2.16×10^{-4}	7.82×10^{-5}
3	5	5.92×10^{-3}	1.86×10^{-4}	8.17×10^{-5}	5.66×10^{-3}	1.83×10^{-4}	8.00×10^{-5}
4	5	2.58×10^{-3}	1.89×10^{-4}	8.53×10^{-5}	2.47×10^{-3}	1.86×10^{-4}	8.35×10^{-5}
3	6	5.92×10^{-3}	1.31×10^{-4}	6.36×10^{-5}	5.66×10^{-3}	1.29×10^{-4}	6.22×10^{-5}
4	6	2.58×10^{-3}	1.20×10^{-4}	6.42×10^{-5}	2.47×10^{-3}	1.17×10^{-4}	6.28×10^{-5}
5	6	1.37×10^{-3}	1.23×10^{-4}	6.58×10^{-5}	1.31×10^{-3}	1.21×10^{-4}	6.44×10^{-5}
3	7	5.92×10^{-3}	1.01×10^{-4}	5.14×10^{-5}	5.66×10^{-3}	9.94×10^{-5}	5.03×10^{-5}
4	7	2.58×10^{-3}	9.10×10^{-5}	5.10×10^{-5}	2.47×10^{-3}	8.93×10^{-5}	4.99×10^{-5}
5	7	1.37×10^{-3}	8.49×10^{-5}	5.08×10^{-5}	1.31×10^{-3}	8.33×10^{-5}	4.97×10^{-5}
6	7	8.16×10^{-4}	8.69×10^{-5}	5.13×10^{-5}	7.82×10^{-4}	8.54×10^{-5}	5.02×10^{-5}
4	8	2.58×10^{-3}	7.39×10^{-5}	4.10×10^{-5}	2.47×10^{-3}	7.25×10^{-5}	4.01×10^{-5}
5	8	1.37×10^{-3}	6.75×10^{-5}	4.02×10^{-5}	1.31×10^{-3}	6.62×10^{-5}	3.94×10^{-5}
6	8	8.16×10^{-4}	6.29×10^{-5}	3.97×10^{-5}	7.82×10^{-4}	6.17×10^{-5}	3.89×10^{-5}
7	8	5.30×10^{-4}	6.37×10^{-5}	3.99×10^{-5}	5.08×10^{-4}	6.26×10^{-5}	3.90×10^{-5}
5	9	1.37×10^{-3}	5.61×10^{-5}	3.18×10^{-5}	1.31×10^{-3}	5.50×10^{-5}	3.12×10^{-5}
6	9	8.16×10^{-4}	5.12×10^{-5}	3.12×10^{-5}	7.82×10^{-4}	5.02×10^{-5}	3.06×10^{-5}
7	9	5.30×10^{-4}	4.75×10^{-5}	3.09×10^{-5}	5.08×10^{-4}	4.67×10^{-5}	3.03×10^{-5}
8	9	3.65×10^{-4}	4.78×10^{-5}	3.11×10^{-5}	3.50×10^{-4}	4.70×10^{-5}	3.05×10^{-5}
5	10	1.37×10^{-3}	4.74×10^{-5}	2.48×10^{-5}	1.31×10^{-3}	4.65×10^{-5}	2.43×10^{-5}
6	10	8.16×10^{-4}	4.30×10^{-5}	2.44×10^{-5}	7.82×10^{-4}	4322×10^{-5}	2.39×10^{-5}
7	10	5.30×10^{-4}	3.92×10^{-5}	2.42×10^{-5}	5.08×10^{-4}	3.84×10^{-5}	2.37×10^{-5}
8	10	3.65×10^{-4}	3.64×10^{-5}	2.41×10^{-5}	3.50×10^{-4}	3.57×10^{-5}	2.36×10^{-5}
9	10	2.63×10^{-4}	3.66×10^{-5}	2.44×10^{-5}	2.53×10^{-4}	3.60×10^{-5}	2.39×10^{-5}
5	11	1.37×10^{-3}	4.03×10^{-5}	1.88×10^{-5}	1.31×10^{-3}	3.95×10^{-5}	1.84×10^{-5}
6	11	8.16×10^{-4}	3.66×10^{-5}	1.88×10^{-5}	7.82×10^{-4}	3.59×10^{-5}	1.84×10^{-5}
7	11	5.30×10^{-4}	3.32×10^{-5}	1.88×10^{-5}	5.08×10^{-4}	3.25×10^{-5}	1.84×10^{-5}
8	11	3.65×10^{-4}	3.03×10^{-5}	1.88×10^{-5}	3.50×10^{-4}	2.97×10^{-5}	1.84×10^{-5}
9	11	2.63×10^{-4}	2.83×10^{-5}	1.90×10^{-5}	2.53×10^{-4}	2.78×10^{-5}	1.86×10^{-5}
10	11	1.96×10^{-4}	2.85×10^{-5}	1.94×10^{-5}	1.88×10^{-4}	2.80×10^{-5}	1.90×10^{-5}
6	12	8.16×10^{-4}	3.13×10^{-5}	1.41×10^{-5}	7.82×10^{-4}	3.06×10^{-5}	1.38×10^{-5}
7	12	5.30×10^{-4}	2.84×10^{-5}	1.43×10^{-5}	5.08×10^{-4}	2.78×10^{-5}	1.40×10^{-5}
8	12	3.65×10^{-4}	2.58×10^{-5}	1.45×10^{-5}	3.50×10^{-4}	2.53×10^{-5}	1.43×10^{-5}
9	12	2.63×10^{-4}	2.37×10^{-5}	1.48×10^{-5}	2.53×10^{-4}	2.33×10^{-5}	1.45×10^{-5}
10	12	1.96×10^{-4}	2.23×10^{-5}	1.51×10^{-5}	1.88×10^{-4}	2.19×10^{-5}	1.48×10^{-5}
11	12	1.50×10^{-4}	2.26×10^{-5}	1.56×10^{-5}	1.44×10^{-4}	2.22×10^{-5}	1.53×10^{-5}

Table 3. Comparison between MC calculated S -values (Gy/Bq · s) of spherical concentric cells with $R_C = 5 \mu\text{m}$ and $R_N = 4 \mu\text{m}$.

	MC code	$S(N \leftarrow N)$	<i>BrIccEmis</i> /MC code	$S(N \leftarrow Cy)$	<i>BrIccEmis</i> /MC code	$S(N \leftarrow CS)$	<i>BrIccEmis</i> /MC code
⁶⁷ Ga	<i>BrIccEmis</i> -condensed	3.76×10^{-3}		5.79×10^{-4}		1.69×10^{-4}	
	<i>BrIccEmis</i> -isolated	3.71×10^{-3}		5.78×10^{-4}		1.69×10^{-4}	
	MC4 ^a	3.62×10^{-3}	+4%/+ 2%	5.17×10^{-4}	+12%/+ 12%	1.41×10^{-4}	+20%/+ 20%
	ETRACK ^b	3.38×10^{-3}	+11%/+ 10%	5.81×10^{-4}	0%/− 1%	2.22×10^{-4}	−24%/− 24%
^{99m} Tc	<i>BrIccEmis</i> -condensed	1.54×10^{-3}		9.08×10^{-5}		5.43×10^{-5}	
	<i>BrIccEmis</i> -isolated	1.52×10^{-3}		9.15×10^{-5}		5.47×10^{-5}	
	MC4 ^a	1.59×10^{-3}	−3%/− 4%	8.46×10^{-5}	+7%/+ 8%	4.80×10^{-5}	+13%/+ 14%
	GEANT4 ^c	1.57×10^{-3}	−2%/− 3%	8.29×10^{-5}	+10%/+ 10%	4.99×10^{-5}	+9%/+ 10%
¹¹¹ In	ETRACK ^b	1.53×10^{-3}	+1%/− 1%	7.41×10^{-5}	+23%/+ 23%	3.90×10^{-5}	+39%/+ 40%
	<i>BrIccEmis</i> -condensed	2.77×10^{-3}		3.82×10^{-4}		2.68×10^{-4}	
	<i>BrIccEmis</i> -isolated	2.70×10^{-3}		3.83×10^{-4}		2.69×10^{-4}	
	MC4 ^a	2.85×10^{-3}	−3%/− 5%	3.01×10^{-4}	+27%/+ 27%	2.64×10^{-4}	+2%/+ 2%
¹²³ I	GEANT4 ^c	2.78×10^{-3}	0%/− 3%	3.60×10^{-4}	+6%/+ 6%	2.43×10^{-4}	+10%/+ 10%
	ETRACK ^b	2.72×10^{-3}	+2%/− 1%	2.95×10^{-4}	+29%/+ 30%	1.88×10^{-4}	+43%/+ 43%
	<i>BrIccEmis</i> -condensed	2.86×10^{-3}		3.06×10^{-4}		1.94×10^{-4}	
	<i>BrIccEmis</i> -isolated	2.78×10^{-3}		3.03×10^{-4}		1.92×10^{-4}	
¹²⁵ I	MC4 ^a	3.05×10^{-3}	−6%/− 9%	3.35×10^{-4}	−9%/− 10%	2.14×10^{-4}	−9%/− 10%
	ETRACK ^b	2.93×10^{-3}	−2%/− 2%	2.66×10^{-4}	+15%/+ 15%	1.42×10^{-4}	+37%/+ 35%
	<i>BrIccEmis</i> -condensed	6.51×10^{-3}		6.44×10^{-4}		3.72×10^{-4}	
	<i>BrIccEmis</i> -isolated	6.40×10^{-3}		6.40×10^{-4}		3.70×10^{-4}	
²⁰¹ Tl	MC4 ^a	6.87×10^{-3}	−5%/− 7%	6.94×10^{-4}	−7%/− 8%	4.18×10^{-4}	−11%/− 11%
	ETRACK ^b	6.78×10^{-3}	−4%/− 6%	6.54×10^{-4}	−2%/− 2%	3.61×10^{-4}	+3%/+ 2%
	GEANT4 ^c	6.25×10^{-3}	+4%/+ 2%	6.37×10^{-4}	+1%/0%	3.16×10^{-4}	+18%/+ 17%
	<i>BrIccEmis</i> -condensed	7.74×10^{-3}		1.25×10^{-3}		6.07×10^{-4}	
²⁰¹ Tl	<i>BrIccEmis</i> -isolated	7.14×10^{-3}		1.24×10^{-3}		6.05×10^{-4}	
	MC4 ^a	8.26×10^{-3}	−6%/− 14%	1.20×10^{-3}	+4%/+ 4%	5.51×10^{-4}	+10%/+ 10%
	ETRACK ^b	7.83×10^{-3}	−1%/− 9%	1.27×10^{-3}	−1%/− 2%	6.48×10^{-4}	−6%/− 7%

^a Data from Bousis *et al* (2010).^b Data from Ftáňíková and Böhm (2000).^c Data from Šefl *et al* (2015).

more than 10%. Thus the large discrepancies between *BrIccEmis* and ETRACK are mainly due to the differences in the MC transport codes rather than the radiation spectra.

The contributions of x- and γ -rays to the S -values were ignored in the MIRD Cellular S -values monograph (Goddu *et al* 1997) as they reportedly contributed less than 2% to the total S -value, $S_{\text{TOT}} = S(N \leftarrow N) + S(N \leftarrow \text{Cy}) + S(N \leftarrow \text{CS})$. However, for very small R_C and R_N and for increasing atomic numbers, the x-ray spectra taken from MIRD-RADTABS data (Eckerman and Endo 2008a) may contribute as much as 5% to the total S -values calculated from equation (1) (supplementary table 8). In contrast, the total contribution of x-rays to S -value calculations from *BrIccEmis* is less than 1% for all radionuclides considered (supplementary table 9—data presented for isolated-atom assumption only).

4. Discussion

Energies of x-rays, AE and CK electrons in the spectra from MIRD-RADTABS (Eckerman and Endo 2008a) were extracted directly from EADL (Perkins *et al* 1991) which is based on singly-ionised atomic systems. This simplification, which was implemented partly due to computational limitations at the time, neglects the variations in transition energies caused by the presence of spectator vacancies during the vacancy cascade. Previous findings of cascade modelling agreed that each atomic transition energy should be calculated from the electronic configurations involved in that particular event (Howell 1992, Pomplun 2000). Even in the case of the L-Auger group, where the effect of spectator vacancies is expected to be minimal, the spectrum derived from EADL without modification exhibits a shift towards higher energies (see figure 2).

Most non-radiative transitions, especially those involving higher atomic subshells, have lower energies than the values reported in EADL and some transitions simply become energetically forbidden after the effects of spectator vacancies are properly accounted for. Consequently, *BrIccEmis* spectra yielded lower energy deposition than MIRD-RADTABS spectra within the first 100 nm, where most of the AE and CK electrons are stopped (figure 1 and supplementary figures 8–10). Experimental spectra of low-energy AE and CK electrons (<1200 eV) from radioactive decays are extremely scarce in the literature. One such energy spectrum (Kovalík *et al* 2004) is highly distorted thus useful comparisons could not be made. Future measurements of low-energy electrons with cleaner background are desired for more detailed comparison between theory and experiment.

The isolated-atom approximation produced more low-energy x-rays (bound-bound transitions) than the condensed-phase approximation (supplementary figures 1–7) owing to the inhibition of low-energy AE and CK electrons near the end of the vacancy cascade. The so-called free-bound x-rays from MIRD-RADTABS could increase the total contribution of x-rays to the S -values up to 5% at small cell sizes (see subsection Cellular S -values). Nevertheless, simulations of photon transport below the current cut-off energy (50 eV) would resolve the sharp fall seen in x-ray DPKs (figure 3 and supplementary figures 11–13) derived from MIRD-RADTABS and reduce the total contribution of x-rays to the S -values.

Notwithstanding the fact that vibrational relaxation is a competing process with electronic relaxation, the mechanism of valence-vacancy neutralisation in a molecular environment is further complicated by other processes that could emit ultra-low energy electrons (ULEE) rather than photons, such as interatomic Coulombic decay (Cederbaum *et al* 1997, Stumpf *et al* 2016). These processes, which occur as a terminal decay step of vacancy cascades, could emit extra ULEE which would otherwise be energetically forbidden in an atomic environment. Given that the cut-off energy in simulations was set at 50 eV, the emission of these electrons could affect the presently calculated energy deposition only in the first few nanometres

from the source. These processes are environment-dependent and they are beyond the scope of the current version of *BrIccEmis*.

One of the main factors contributing to the large variation of calculated AE and CK yields reported in the literature, is the assumption on whether valence-vacancy neutralisation happens during the vacancy cascade which has a very short lifetime ($\sim 10^{-16}$ – 10^{-13} s). The condensed-phase approximation differs from the isolated-atom approach at radii below the maximum range of M-shell Auger electrons (figure 1 and supplementary figures 8–10). These differences are mainly found within the first 100 nm except in heavy atomic systems such as those involved in the decay of $^{195\text{m}}\text{Pt}$ and ^{201}Tl . Recent ultrafast spectroscopic studies have shown that the electron transfer at certain molecule-nanoparticle interfaces is within a few hundreds of femtoseconds (Anderson and Lian 2004, Furube *et al* 2014). These experiments indicate that the electron transfer rate could be as fast as the vacancy cascade in some circumstances. The reported electron transfer rate is, however, dependent on the properties of the compounds as well as the environment. Better designed experiments using radiopharmaceutical drugs could be crucial in resolving the aforementioned ambiguity. Detection of very low-energy x-rays could be used as an alternative method to distinguish the two approximations since the isolated-atom approximation gives rise to measureable amount of x-rays with energies below 500 eV.

The use of abridged decay data is common in the calculation of *S*-values in the literature (Ftániková and Böhm 2000, Bousis *et al* 2010, Šefl *et al* 2015). Our work shows that the use of averaged data is not critical for the calculation of cellular *S*-values. Nevertheless, unabridged decay data is better as input for the study of sub-micrometre energy deposition since the abridged version could artificially change the dose distribution at this scale.

Although DPKs are shown to be highly sensitive to the differences in input decay spectra, cellular *S*-values are, however, relatively unaffected except for heavy atomic systems within the isolated-atom approach. Differences in the first 100 nm between the DPKs derived from MIRD-RADTABS and *BrIccEmis* spectra are rapidly diminished for larger dimensions. This is evident from a comparison of the energy deposition as a function of DNA dimensions. Figure 4 compares the energy deposition based on spectra from MIRD-RADTABS with *BrIccEmis* in spheres of diameters representing different DNA condensation states. The absorbed doses derived from MIRD-RADTABS spectra are significantly higher in the first 30 nm diameters. However, these differences are considerably reduced as diameters representing DNA condensation states increase.

Taking into consideration that the potential radiation risk of AE emitters used in diagnostic nuclear medicine has been highlighted, especially in the case of intra-nuclear internalisation of the radiopharmaceutical (Howell 2011), dose calculations using MIRD-RADTABS data (Eckerman and Endo 2008a) could potentially lead to an overestimation of radiation risk at sub-cellular dimensions. In contrast, when nuclear targeting with an AE emitter is desired for therapy it is essential to maximise the absorbed dose, thus necessitating the use of accurate decay spectra for dose calculation. Geant4, which has been used to assess the dose enhancement of high-*Z* nanoparticles in radiotherapy and proton therapy (Butterworth *et al* 2012, Yuting *et al* 2014), has recently incorporated a MIRD-RADTABS-esque atomic vacancy cascade approach to generate AE and CK energy spectra for investigations into applications of these nanoparticles in medicine (Incerti *et al* 2016). The dose enhancement in the first few hundred nanometres could be affected by the accuracy of AE and CK energy spectra. In the future, the effect of different AE and CK energy spectra on the dose enhancement of high-*Z* nanoparticles should be carefully investigated.

5. Conclusion

We conclude that the choice of atomic relaxation spectrum after nuclear decay (*BrIccEmis* versus MIRD-RADTABS (Eckerman and Endo 2008a) and *BrIccEmis*-isolated versus *BrIccEmis*-condensed) considerably affects the DPKs of AE emitting radionuclides up to several hundred nanometres. This implies that caution should be employed when evaluating the energy deposition of an AE emitting radionuclide over DNA dimensions. Nevertheless, these differences did not significantly affect *S*-values which are calculated for volumes of larger dimension as most AE and CK electrons deposit their energies within the first micrometre from the decay site. Although the two different approaches concerning the fate of valence vacancies produced during the vacancy cascade (isolated versus condensed) yield significantly different numbers of AE and CK electrons, the discrepancy in DPKs due to different electron yields is mainly restricted to the first 100 nm from a point source.

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Conclusion

8.1 Summary

Atomic relaxation, which is also called vacancy cascade, occurs whenever an atom is ionised and it leads to the emission of Auger and Coster-Kronig electrons as well as X-rays. Accurate modelling of the vacancy cascade is important to many scientists in numerous fields. A new Monte Carlo model of the vacancy cascade, **BrIccEmis**, that accounts for the shake-off process and the effect of spectator vacancies, was developed during the course of the author's PhD research. As a general overarching theme this was the topic and motivation of this thesis, applying the **BrIccEmis** code to study the charge-state distributions and energy spectra resulting from inner-shell ionisations as well as to evaluate the energy deposition by Auger-emitting medical radioisotopes.

The relativistic self-consistent-field Dirac-Fock (DF) method in the implementation of the **RAINE** code [112] was utilised to calculate the transition energies in neutral atoms and ions with arbitrary distributions of spectator vacancies created. For energies of the K-shell Auger electrons and X-rays in the singly-ionised atomic systems, this method deviates from the best calculations and experiments for atomic numbers above $Z = 45$, it overestimates by about 0.8% at $Z = 99$. On the other hand, the level of accuracy was good for transitions from the L shell and above. Moreover, the simulated low-energy electron spectrum (< 4 keV) of the decay of ^{125}I , in which the source was deposited on a thin gold substrate, was demonstrated to agree well with experiment in terms of the overall structure. This agreement has laid the foundation for the quantification of the enhancement of low-energy electron emission from gold due to external irradiation that has been of great interest to researchers in targeted nanotherapy [8, 231, 232]. Furthermore, excellent agreement has been achieved between theory and experiment in multi-electron coincidence spectroscopy for M₅-subshell photoionisation in xenon. This agreement allowed informed speculations to be made about the origins of several components in the measured spectrum, including the shake-photoelectron pairs, in which their formations have been largely neglected in previous studies. The importance of the shake-off process in the vacancy cascade has also been highlighted in this study.

The **BrIccEmis** code was then used to investigate the residual charge-state distribu-

tions following inner-shell ionisations. For the vacancy cascade initiated by M_5 -subshell photoionisation in xenon, the **BrIccEmis** simulation was demonstrated to be in good agreement with experiment. Although the agreement between theory and experiment was worse for the ^{131m}Xe isomeric decay, in which atomic vacancies in deeper shells can be created, **BrIccEmis** was able to reproduce the distribution width and the intensity of highly-ionised ions (Xe^{12+} and onwards). The choice of the propagating vacancy was found to become a contentious issue in the simulation when a vacancy cascade begins in a very deep shell. Ideally, the propagating vacancy should be chosen based on the transition rates but this method was demonstrated to yield worse agreement with experiment than the random selection of the propagating vacancy (see Chapter 5 for details). This can be attributed to the inaccuracy of the calculated transition rates from the Evaluated Atomic Data Library (EADL) that has been demonstrated in previous studies [132, 135]. In fact, the transition rates calculated based on the DF method using the **Grasp2K** [150] and **Ratip** [151] packages, were found to improve the agreement between theory and experiment. The agreement in the charge-state distributions, coupled to the agreement demonstrated for the energy spectra, validate the vacancy-cascade model implemented in the **BrIccEmis** code.

This thesis then investigated applying the **BrIccEmis** code to the charge-state distribution of highly-ionised barium fission fragments that are perturbed due to the internal conversion decays of nanosecond nuclear isomers. The charge-state distribution of these barium fission products is usually Gaussian-like and centred at approximately $21+$ when nanosecond nuclear isomers are not present. The theoretical perturbed charge-state distributions due to the isomeric decays in ^{141}Ba , ^{143}Ba and ^{145}Ba , were derived using the vacancy-cascade simulation and a Bayesian inference technique. The simulation used the ground-state electron configuration for each charge state and the unperturbed distribution was assumed to be Gaussian in the derivation. This allowed deconvolution of the components of experimental charge-state distribution and the derived unperturbed distributions were in fairly good agreement with semi-empirical predictions. It was demonstrated that the agreement could be improved by accounting for secondary effects such as the inclusion of non-ground-state electron configurations. The agreement highlights that the **BrIccEmis** code is applicable for the highly-ionised systems and it will become a valuable tool in the future hunts for low-lying nanosecond nuclear isomers in fission fragments.

The importance of accurate Auger-energy spectra in the dosimetric calculations of Auger-electron emitting radioisotopes was then investigated. It was demonstrated that adopting a rigorous approach to assess transition energies during the vacancy cascade, could change the distribution of dose deposition significantly in the first few hundred nanometres from a point source, compared to an approach where transition energies are derived from neutral binding energies. However, the differences did not significantly affect S -values (absorbed dose per unit cumulated activity), which were calculated for volumes of larger dimension. This study pinpointed the spatial domain where accurate Auger-

energy spectra is essential for future radiobiological studies of Auger-emitting radioactive or irradiated atoms. Furthermore, the impact of the assumption on whether valence-vacancy neutralisation happens during the vacancy cascade (isolated or condensed), which is one of the main factors contributing to the large variation of calculated AE and CK yields reported in the literature, was demonstrated to be mainly restricted to the first 100 nanometres from a point source.

8.2 Outlook

8.2.1 Improvements

Atomic transition rates from EADL are essential in the **BrIccEmis** code as well as all existing vacancy-cascade models. Non-radiative rates from EADL were calculated using the so-called “frozen-orbital” approximation in order to preserve the orthogonality between the initial- and final-state orbitals. However, the frozen-orbital approximation was found to overestimate the transition rates by a few percent up to 14% compared with the calculations using fully relaxed wave functions for the closed-shell atomic systems [206, 233]. While the present simulations based on the EADL rates yield good agreement with experiments, it would be possible to improve the agreement with the transition rates calculated using separately optimised initial- and final-state orbitals. Colleagues at Malmö University, who are the developers of the **Grasp2K** code [149, 150], have been working on the implementation of the biorthogonal transformation [204] for the computation of Auger rates, which will eliminate the need of the frozen-orbital approximation in the future.

In addition, the EADL rates were calculated for the singly-ionised systems but multiply-ionised states are frequently encountered in a vacancy-cascade simulation. In the **BrIccEmis** code, a statistical correction proposed by Krause and Carlson [85] was used to adjust the EADL transition rates for the multiply-ionised states (see Section 3.4.3). This correction was derived based on the fact that Auger rates were found to be fairly constant over a wide range of atomic numbers [146], but the same cannot be said for Coster-Kronig (CK) transitions [148], which are usually low in energy. As a result, the accuracy of the vacancy-cascade simulation could be strongly affected by this statistical correction. Therefore, the **BrIccEmis** code could be improved with an additional correction for the CK transitions, which could be derived from a systematic study of the variation of the CK rates with atomic number.

One of the universal themes running through all the simulations reported here is the importance of the shake-off process in the vacancy cascade. **BrIccEmis** adopted the shake theory from Carlson and Nestor [14, 51] to calculate the shake-off probabilities. Since electron-electron correlation was not accounted, the calculated shake-off probabilities were mainly applicable for the near closed-shell atoms. Recently, a group in the University of

Melbourne [56, 136] demonstrated that the inclusion of the electron-electron correlation in the calculation of shake-off probabilities is essential in achieving good agreement with experiments for transition metals (open- d shell). Such calculations in future studies would help extend the **BrIccEmis** code to include the shake-off process for Auger-emitting medical radioisotopes, in which many come from transition metals and lanthanides.

One improvement that could be done without too much effort is to extend Schönfeld's method of calculating subshell electron-capture probabilities [17] to unique first-forbidden transitions. Strictly speaking, the Schönfeld method is only applicable for allowed and non-unique first-forbidden transitions, but **BrIccEmis** applied it to all types of electron capture. This assumption neglects the contribution of the L_3 subshell in the unique first-forbidden transitions. Capture probabilities of higher-order forbidden unique and non-unique transitions decrease very rapidly with increasing degree of forbiddenness, so accurate calculation of probabilities for these transitions is not critical.

8.2.2 Further applications

This work provides a valuable tool in the form of the **BrIccEmis** code for research into many Auger-related studies. The Monte Carlo model could readily be utilised in a number of interesting topics such as:

- The activity determination of electron-capture radioisotopes by means of liquid scintillation counting (LSC). Realistic modelling of the vacancy cascade was demonstrated to be crucial in improving the computation of the counting efficiency in the LSC method [234, 235, 236, 237]. This application of the **BrIccEmis** code could help extend the activity standardisation to many electron-capture radioisotopes beyond atomic number, $Z = 58$ (Ce), which is the current limit in the LSC technique [236].
- The improvement of the existing radiobiological models. The Geant4-DNA package has recently extended the theoretical electron ionisation cross sections down to 7.4 eV [238] for research into low-energy electron therapy such as the radiosensitisation of gold nanoparticles. However, the vacancy-cascade simulation currently implemented in the Geant4-DNA package [239] is similar to the one in the MIRD-RADTABS program [81] (see Chapter 7 for details). The **BrIccEmis** code could provide a more realistic model of the vacancy cascade to improve the existing radiobiological models for better research outcomes.
- The degradation mechanism of radiopharmaceuticals carrying *in vivo* generators, such as ^{140}Nd and ^{212}Pb , could be studied. ^{212}Pb has recently been used in a human trial for α radioimmunotherapy [240, 241]. Although ^{212}Pb is a β^- -emitter, its decay chain produces α -emitters ^{212}Bi and ^{212}Po . However, ^{212}Pb predominantly decays to

excited states of ^{212}Bi , which then decays by the internal conversion of γ -rays approximately 42% of the time. The emission of conversion electrons and Auger electrons from the subsequent vacancy cascade has been coined as one of the leading causes in freeing the excited ^{212}Bi from the radiopharmaceuticals [242, 243], thus degrading the drugs. The **BrIccEmis** code could help to understand the exact mechanism of the degradation for improving the design of therapeutic radiopharmaceuticals utilising *in vivo* generators.

Appendix A

A.1 Nomenclature

ω_{Y_i}	Fluorescence or radiative yield is defined as the number of X-rays for transitions to the subshell Y_i divided by the number of Y_i vacancies.
a_{Y_i}	Auger yield is defined as the number of Auger electrons divided by the number of Y_i vacancies.
f_{ij}	Coster-Kronig yield is defined as the number of transitions shifting vacancies from the Y_i subshell to the Y_j subshell divided by the number of Y_i vacancies.
Auger KLL	K -shell Auger transition where the two new vacancies are in the L -shell.
Auger KLX	K -shell Auger transition where one of the two new vacancies is in the L -shell.
Auger KXY	K -shell Auger transition where neither of the two new vacancies is in the L -shell.
CK LLM	L -shell CK transitions where one of the two new vacancies is in the M -shell.
CK LLX	L -shell CK transitions where neither of the two new vacancies is in the M -shell.
Auger LMM	L -shell Auger transitions where the two new vacancies are in the M -shell.
Auger LMX	L -shell Auger transitions where one of the two new vacancies is in the M -shell.
Auger LXY	L -shell Auger transitions where neither of the two new vacancies is in the M -shell.
Super CK NNN	N -shell Super CK transitions.
CK NNX	N -shell CK transitions where one of the two new vacancies is in the O -shell.
Auger NXY	N -shell Auger transitions where the two new vacancies are in the O -shell.
X-ray K_{α_1}	KL_3 X-ray transition
X-ray K_{α_2}	KL_2 X-ray transition
X-ray K_{β}	KM, N X-ray transitions

A.2 Independent decays of ^{123}I , ^{124}I and ^{125}I .

Table A.1: Examples of Monte Carlo calculated spectra of radiations released in independent decays of ^{123}I in the condensed phase. Coster-Kronig transitions are included in Auger transitions. This table has been published in Ref. [122].

Decay No.	Radiation	No. of rad.	Energy (eV)							
1	γ	1	158970.0							
	X-ray	1	27297.2							
	Auger	17	2829.2	135.4	388.9	84.1	432.2	3.2	8.0	16.8
			2.8	0.2	10.8	12.9	7.3	25.6	26.7	13.6
2			18.5							
	IC	1	127156.2							
	X-ray	2	27571.6	27571.7						
	Auger	20	3280.7	1.7	778.3	26.7	4.9	12.1	26.7	26.7
3			13.6	17.8	2904.9	70.1	317.7	395.5	146.5	26.1
	IC	1	127156.2							
	X-ray	2	3778.0	27297.2						
	Auger	12	547.6	465.8	8.0	14.4	28.7	3412.1	475.7	2.4
4			7.9	478.4	19.1	11.5				
	γ	1	158970.0							
	Auger	5	4325.6	465.8	8.0	14.4	18.5			
5	γ	1	158970.0							
	X-ray	1	27297.2							
	Auger	9	3692.0	16.4	151.3	476.6	20.9	13.2	26.7	13.6
			18.5							
6	γ	1	158970.0							
	X-ray	1	27297.2							
	Auger	11	3127.7	488.2	11.4	28.2	550.9	16.9	7.7	1.5
			83.7	25.4	7.0					
7	γ	1	158970.0							
	X-ray	1	27571.6							
	Auger	12	3376.3	91.0	314.2	2.2	92.6	8.2	19.6	10.3
			7.8	22.5	13.6	18.5				
8	γ	1	158970.0							
	X-ray	1	31098.9							
	Auger	7	99.1	17.6	11.5	463.6	8.0	26.1	19.3	
9	γ	1	158970.0							
	X-ray	2	27571.6	4311.0						
	Auger	1	5.4							
10	γ	1	158970.0							
	X-ray	1	31801.8							
	Auger	3	24.9	14.4	18.5					
11	γ	1	158970.0							
	X-ray	1	31098.9							
	Auger	9	109.8	385.7	6.8	9.9	7.5	8.9	22.3	14.4
			18.5							
12	γ	1	158970.0							
	Auger	13	181.4	3399.9	472.3	5.1	11.7	17.5	11.0	392.1
			8.8	10.5	8.5	25.1	18.5			
13	γ	1	158970.0							
	X-ray	1	27571.6							
	Auger	11	2915.2	395.4	23.9	60.8	18.2	217.0	406.7	16.0
			24.9	15.4	18.5					

Table A.1: Continued.

Decay	No.Radiation	No. of rad.	Energy (eV)							
14	IC	1	127156.2							
	X-ray	1	31046.7							
	Auger	18	49.4	8.4	24.6	1.5	646.2	14.9	24.9	13.8
			19.3	141.2	373.5	0.5	5.8	1.8	7.3	14.9
			7.8	11.5						
15	γ	2	528960.0	158970.0						
	X-ray	1	31791.8							
	Auger	3	34.9	13.8	20.0					

Table A.2: Examples of Monte Carlo calculated spectra of radiations released in independent decays of ^{124}I in the condensed phase. Coster-Kronig transitions are included in Auger transitions. This table has been published in Ref. [122].

Decay	No.Radiation	No. of rad.	Energy (eV)							
1	$\beta+$	1								
2	$\beta+$	1								
3	γ	2	1690960.0	602730.0						
	X-ray	1	27297.2							
	Auger	12	3133.2	692.4	287.2	16.3	2.8	0.2	23.6	9.9
			23.0	59.0	13.0	30.2				
4	$\beta+$	1								
5	γ	2	722780.0	602730.0						
	X-ray	1	27571.6							
	Auger	7	3637.2	6.8	10.6	464.2	22.3	13.8	20.0	
6	X-ray	1	27571.6							
	Auger	9	3148.0	412.1	454.0	0.8	6.3	8.9	8.4	14.4
			18.5							
7	γ	2	1690960.0	602730.0						
	Auger	21	22214.7	3428.0	356.7	477.5	14.6	18.9	12.0	448.5
			2619.2	130.3	101.7	422.9	385.8	112.9	60.3	3.8
			10.6	1.6	8.3	14.3	20.0			
8	Auger	11	2889.6	881.3	60.0	25.9	6.9	269.8	450.9	1.3
			8.5	13.6	17.8					
9	$\beta+$	1								
	γ	1	602730.0							
10	Auger	18	22797.2	3678.2	107.0	2822.4	97.2	455.2	321.9	228.2
			77.1	50.7	4.5	76.8	3.9	10.6	1.5	7.2
			14.3	30.2						
11	X-ray	1	27571.6							
	Auger	11	3148.0	404.2	24.3	7.2	10.9	393.5	7.5	8.9
			7.2	15.1	7.0					
12	γ	1	602730.0							
	X-ray	1	27571.6							
	Auger	14	2915.2	20.0	113.9	389.3	3.2	371.2	0.1	6.8
			9.9	7.5	16.0	24.9	14.4	18.5		
13	γ	2	722780.0	602730.0						
	X-ray	2	27297.2	4041.2						
	Auger	5	406.7	10.5	8.0	0.7	5.4			
14	X-ray	1	27571.6							
	Auger	11	3159.5	404.0	8.5	385.7	6.8	9.9	7.5	14.9
			26.7	26.0	5.4					

Table A.2: Continued.

Decay No.	Radiation	No. of rad.	Energy (eV)							
15	$\beta+$	1								
	IC	1	570916.2							
	X-ray	1	27571.6							
	Auger	14	2653.7	77.5	273.7	22.4	412.2	12.3	6.3	0.3
			57.5	1.4	7.3	13.1	7.8	11.5		

Table A.3: Examples of Monte Carlo calculated spectra of radiations released in independent decays of ^{125}I in the condensed phase. Coster-Kronig transitions are included in Auger transitions. This table has been published in Ref. [122].

Decay No.	Radiation	No. of rad.	Energy (eV)							
1	IC	1	3678.4							
	X-ray	2	31801.8	27297.2						
	Auger	8	26.7	26.0	5.4	3973.7	8.7	478.4	6.1	13.1
2	IC	1	3678.4							
	X-ray	1	31099.0							
	Auger	26	23073.0	3777.1	3102.1	482.9	269.3	321.6	0.4	2.1
			14.5	15.1	34.9	10.8	12.9	7.3	7.8	6.8
			13.8	20.0	28.8	375.5	0.3	5.7	0.5	8.5
			107.4	0.1						
3	IC	1	3678.4							
	X-ray	2	147.3	27571.7						
	Auger	30	23073.0	2647.3	2433.3	765.2	749.4	605.0	95.0	391.3
			16.2	54.2	1.1	5.7	14.9	6.5	12.6	18.5
			2653.6	483.5	17.0	57.7	428.9	0.4	12.1	35.9
			5.6	14.5	5.4	16.8	5.9	10.8		
4	IC	1	3678.4							
	X-ray	2	27297.2	27571.7						
	Auger	18	3422.4	392.3	0.01	4.4	24.8	385.8	21.1	17.1
			7.2	26.8	19.3	3137.7	474.3	3.8	322.2	1.1
			145.6	21.6						
5	IC	1	30553.0							
	X-ray	1	27297.2							
	Auger	23	3176.1	72.3	530.3	11.2	385.2	7.1	9.9	8.3
			19.3	8.5	26.0	18.5	456.1	3443.4	1.7	164.7
			441.8	6.7	11.4	5.4	13.5	19.5	23.2	
6	IC	1	3678.4							
	X-ray	1	27571.7							
	Auger	20	26665.4	3376.3	408.2	466.5	13.1	3.8	459.0	1.9
			8.8	14.9	24.9	15.4	18.5	3149.2	473.3	438.9
			7.3	1.6	5.0	10.8				
7	IC	1	3678.4							
	X-ray	2	3778.1	27571.7						
	Auger	17	26763.6	107.0	532.2	11.9	15.1	477.6	15.4	17.8
			3137.7	482.8	3.1	363.7	0.5	5.6	25.0	6.1
			0.1							
8	IC	1	3678.4							
	X-ray	2	27297.2	31099.0						
	Auger	27	2886.4	22.7	18.0	470.4	4.2	10.9	16.4	105.8
			4.1	380.8	16.3	7.1	9.9	8.9	13.2	34.9
			15.4	18.5	88.8	6.3	358.0	6.8	11.8	8.3

Table A.3: Continued.

Decay No.	Radiation	No. of rad.	Energy (eV)							
			14.9	6.1	23.2					
9	IC	1	3678.4							
	X-ray	2	27297.2	27571.7						
	Auger	17	3983.7	15.6	488.3	26.1	7.0	3137.7	400.9	299.9
			9.1	12.1	16.6	8.4	11.8	8.3	11.8	8.3
			16.8	5.9	11.5					
10	γ	1	35492.2							
	Auger	1	3983.7							
11	IC	1	3678.4							
	X-ray	2	27571.6	27571.7						
	Auger	22	3159.5	486.9	215.5	80.2	2.0	8.7	13.9	33.6
			14.3	20.0	3377.4	20.6	4.4	442.5	7.5	13.4
			8.4	12.7	8.3	14.9	7.8	11.5		
12	IC	1	3678.4							
	X-ray	2	31801.8	27571.7						
	Auger	12	26.7	26.0	17.8	3407.5	20.8	130.8	466.9	13.1
			5.6	25.0	6.8	11.5				
13	γ	1	35492.2							
	Auger	13	549.6	16.7	3159.5	486.9	10.4	244.3	8.1	35.8
			7.3	22.9	14.9	69.5	7.0			
14	IC	1	3678.4							
	X-ray	3	27571.6	3777.5	27571.7					
	Auger	12	479.4	13.6	17.8	3137.7	400.9	4.9	457.7	12.6
			4.5	14.9	7.8	10.8				
15	IC	1	3678.4							
	X-ray	2	27571.6	27279.2						
	Auger	34	2663.8	77.1	383.5	51.3	11.2	441.7	34.0	11.9
			0.1	20.8	9.9	7.5	10.5	9.8	13.6	18.5
			2876.2	129.4	123.5	12.4	290.7	288.0	0.8	16.1
			0.4	7.7	26.3	0.5	8.4	2.1	17.6	13.5
			7.5	0.1						

A.3 Theory of atomic spectroscopy and Auger transitions

- *LS* coupling

For low atomic number ($Z \leq 30$), the electrostatic interaction between electrons dominates over the spin-orbit interaction, making Russel-Saunders *LS* coupling an appropriate approximation [244]. There are only three *KLL* Auger lines since pure *LS* coupling ignores the fine structure splitting:

$$KL_1L_1, \quad KL_1L_{2,3}, \quad KL_{2,3}L_{2,3}. \quad (\text{A.1})$$

Without the fine structure splitting, many weak transitions would not be visible in *LS* coupling. For example, the four transitions of $L_1L_{2,3}M_{4,5}$ form one strong ‘merged’ line instead of four weaker lines $L_1L_2M_4$, $L_1L_2M_5$, $L_1L_3M_4$ and $L_1L_3M_5$.

- *jj* coupling

For large atomic numbers ($Z > 60$ [245]), and for some Coster-Kronig transitions, the spin-orbit interaction in the final state is stronger than the electrostatic interaction between electrons. The theoretical Auger spectrum is more accurately reproduced by *jj* coupling than *LS* coupling [140]. Pure *jj* coupling is only achieved in the limit $Z \rightarrow \infty$ [244]. A complete description of *jj* coupling is explained in [246]. There are six *KLL* Auger lines in *jj* coupling:

$$KL_1L_1, \quad KL_1L_2, \quad KL_1L_3, \quad KL_2L_2, \quad KL_2L_3, \quad KL_3L_3. \quad (\text{A.2})$$

- Intermediate coupling (IC)

For most atomic numbers, the electrostatic and spin-orbit interaction have the same order of magnitude and the intermediate coupling is more appropriate than other coupling schemes. There are nine lines in intermediate coupling for *KLL* transitions:

$$KL_1L_1(^1S_0), \quad KL_1L_2(^3P_0), \quad KL_1L_2(^1P_1), \quad KL_1L_3(^3P_1), \quad KL_1L_3(^3P_2), \\ KL_2L_2(^1S_0), \quad KL_2L_3(^1D_2), \quad KL_3L_3(^3P_0), \quad KL_3L_3(^3P_2). \quad (\text{A.3})$$

In intermediate coupling, only the total angular momentum of the atom J (and M_J) is a valid quantum number; terms with different L and S quantum numbers but the same J become mixed (total angular momentum J is conserved but orbital angular momentum L and spin S are not conserved).

It is easier to resolve all nine *KLL* lines experimentally in the region of $Z = 40$ [247, 248]

- Configuration interaction (CI)

Configuration interaction may be used to account for the mixing between different electronic states. The theory of configuration interaction is explained in [29]. Configuration interactions weaken the $KL_1L_1(^1S_0)$ but strengthen the $KL_2L_2(^1S_0)$ and $KL_3L_3(^3P_0)$ lines for KLL Auger rates [140]. Existing calculations that include configuration interaction are able to explain the discrepancies between the theoretical and experimental transition rates, especially in atoms with low atomic number Z [247]. Nonetheless, the configuration interaction does not affect the total transition rates for principal atomic shells; the total transition rates for KLL , KLM , KMM , LMM , etc., are unaffected.

A.4 Shake-off probabilities in tellurium

Table A.4: Probabilities (in percent) for electron shake-off from the various subshell, n_s , in tellurium as a result of a sudden vacancy in orbital, n_0 calculated using the methodology proposed by Krause and Carlson [85].

$n_s \backslash n_0$	K	L ₁	L ₂	L ₃	M ₁	M ₂	M ₃	M ₄	M ₅	N ₁	N ₂	N ₃	N ₄	N ₅	O ₁	O ₂	O ₃	Total
K	0.00	0.05	0.09	0.16	0.19	0.30	0.57	0.46	0.67	0.50	0.76	1.49	2.26	3.38	2.95	6.39	7.01	27.21
L ₁	0.00	0.00	0.01	0.02	0.06	0.10	0.22	0.36	0.54	0.30	0.45	0.97	2.30	3.31	2.67	6.15	6.05	23.51
L ₂	0.00	0.01	0.00	0.02	0.08	0.11	0.23	0.38	0.56	0.35	0.46	1.01	2.32	3.33	2.78	6.17	6.08	23.89
L ₃	0.00	0.01	0.00	0.01	0.08	0.11	0.23	0.38	0.56	0.35	0.46	1.01	2.32	3.33	2.78	6.17	6.08	23.89
M ₁	0.00	0.00	0.00	0.02	0.00	0.01	0.02	0.06	0.08	0.14	0.23	0.53	2.32	2.24	2.45	5.81	5.72	19.64
M ₂	0.00	0.00	0.00	0.02	0.01	0.01	0.02	0.06	0.10	0.18	0.22	0.50	1.55	2.23	2.56	5.77	5.68	18.90
M ₃	0.00	0.00	0.00	0.02	0.01	0.01	0.02	0.06	0.10	0.18	0.22	0.50	1.55	2.23	2.56	5.77	5.68	18.90
M ₄	0.00	0.00	0.00	0.02	0.02	0.02	0.04	0.04	0.10	0.20	0.26	0.58	1.56	2.26	2.61	5.86	5.77	19.32
M ₅	0.00	0.00	0.00	0.02	0.02	0.02	0.04	0.06	0.08	0.20	0.26	0.58	1.56	2.26	2.61	5.86	5.77	19.32
N ₁	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.03	0.08	0.52	0.73	1.97	5.22	5.10	13.66
N ₂	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.03	0.05	0.25	0.52	0.59	2.12	5.15	5.05	13.75
N ₃	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.03	0.10	0.19	0.52	0.59	2.12	5.15	5.05	13.73
N ₄	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.02	0.06	0.26	0.48	1.95	5.15	4.81	12.76
N ₅	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.02	0.06	0.35	0.40	1.95	5.15	4.81	12.77
O ₁	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.01	0.40	1.89	1.74	4.07
O ₂	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.01	0.80	0.87	1.65	3.34
O ₃	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.01	0.80	1.72	0.83	3.38

A.5 Initial-vacancy distributions of ^{123}I and ^{125}I decays

Tables A.5 and A.6 compare the initial vacancy distributions calculated for two iodine isotopes to values in the literature. Pomplun [249] and Humm [250] included the contributions from L_3 and M_3 subshells in ^{123}I even though only allowed and first non-unique forbidden EC transitions are present in the radionuclide. The calculated initial-vacancy distributions due to IC processes are consistent with the literature values.

Table A.5: Initial-vacancy distributions for ^{123}I . This table has been published in Ref. [122].

Atomic Subshell	Pomplun et al. [249]		Humm [250]		This work	
	EC	IC	EC	IC	EC	IC
K	0.797	0.135	0.818	0.147	0.854	0.134
L1	0.104	0.016	0.106	0.017	0.1127	0.016
L2	0.003	0.0011	0.003	0.002	0.0029	0.0011
L3	0.007	0.0004	0.007	0.001	–	0.0003
M1	0.022	0.0035	0.022	0.003	0.024	0.003
M2	0.001	0.00	0.001	0.00	0.0007	0.0002
M3	0.038	0.00	0.038	0.00	–	0.00
M4	0.00	0.00	0.00	0.00	–	0.00
M5	0.00	0.00	0.00	0.00	–	0.00
N1	0.005	0.0008	0.005	0.00	0.0052	0.0006
N2	0.00	–	–	–	0.0002	0.00
N3	0.00	–	–	–	–	0.00
O1	–	–	–	–	0.0003	0.00
Sum	0.977	0.1568	1.0	0.17	1.0	0.1552

Table A.6: Initial-vacancy distributions for the decay of ^{125}I . This table has been published in Ref. [122].

Radionuclide	^{125}I			
	Charlton and Booz ([118])		This work	
Atomic Subshell	EC	IC	EC	IC
K	0.806	0.795	0.8024	0.776
L1	0.154	0.095	0.1512	0.0936
L2	0.004	0.007	0.0039	0.0172
L3	–	0.004	–	0.0158
M1	0.0035	0.019	0.0336	0.0186
M2	0.001	0.004	0.0009	0.0036
M3	–	0.001	–	0.0033
M4	–	–	–	0.00
M5	–	–	–	0.00
N1	–	0.004	0.0074	0.0037
N2	–	0.001	0.0002	0.0007
N3	–	–	–	0.0006
O1	–	–	0.0004	0.0004
Sum	1.0	0.930	1.0	0.9335

A.6 Dirac-Hartree-Fock (or Dirac-Fock) calculation of atomic transition rates

Wave functions describing the initial and final-ion states of a radiative or non-radiative transition in the singly-ionised system are calculated using the relativistic atomic structure package **Grasp2K** [149, 150], relying on the multiconfiguration Dirac-Hartree-Fock (MCDHF) approach and the jj coupling scheme. A natural starting point in the fully relativistic calculations is the Dirac-Coulomb Hamiltonian, which is given by:

$$H_{DC} = \sum_{i=1}^N [c\alpha_i \cdot p_i + (\beta_i - 1)c^2 + V_N(r_i)] + \sum_{i>k}^N \frac{1}{r_{ij}}, \quad (\text{A.4})$$

where $V_N(r)$ is the monopole part of the electron-nucleus interaction, r_{ij} is the distance between electrons i and j , and α and β are the Dirac matrices. In MCDHF theory, approximate eigenfunctions to the Dirac-Coulomb Hamiltonian above are the so-called atomic state functions (ASFs), $\Psi(\gamma P J M)$, which are expansions over configuration state functions (CSFs), $\phi(\gamma_i P J M)$, with total angular momentum J symmetry and parity P , and are given by:

$$\Psi(\gamma P J M) = \sum_{i=1}^{N_{CSFs}} c_i \phi(\gamma_i P J M). \quad (\text{A.5})$$

In the expansion above, γ_i represents the configuration, coupling and other quantum numbers necessary to uniquely describe the state, M is the projection of J on the z -axis and c_i is the expansion coefficient. Since electron-electron correlation is not taken into account in the present work, the calculations are performed using the *Dirac-Hartree-Fock (DHF)* approach. However, since the CSFs in **Grasp2K** are generated using non-relativistic configurations as input, an ASF may also in this case be expanded over several interacting CSFs, representing different relativistic configurations and couplings. The advantage of this approach is that the intermediate coupling is taken into account and thus a good description of the resulting ASFs is provided also when mixing of the jj -coupled states is expected to be significant [134]. The CSFs are constructed from the one-electron Dirac orbitals, where the radial wave functions together with the expansion coefficients are obtained in a relativistic self-consistent-field procedure [251]. The transverse photon or Breit interaction as well as the leading quantum electrodynamic (QED) corrections are accounted for in the subsequent relativistic configuration interaction (RCI) calculations [252].

Once a set of ASFs is obtained, the non-radiative transition probability is computed using the **AUGER** component of the **Ratip** package [151]. In **AUGER**, the so-called “frozen-orbital” (FO) approximation must be used, and thus a common orthonormal basis with one-electron Dirac orbitals describing the initial state is used for the computation of the non-radiative transition probability. The energy of the outgoing electron, however, is

obtained as the energy difference of the initial state and the final-ion state using separate orbital basis sets and is referenced to the vacuum level.

For the radiative transition, the **REOS** component of the **Ratip** package is used to compute the probability. In **REOS**, the FO approximation is not required and the computation of transition probability is performed using two sets of independently-optimised orbitals, i.e. non-orthogonal, representing the initial and final-ion states by exploiting the Löwdin expression [201]. The energy of the outgoing photon is obtained by a similar method to the non-radiative transition.

Acknowledgements

The author is indebted to Dr. Jörgen Ekman of Malmö University for his input to this section.

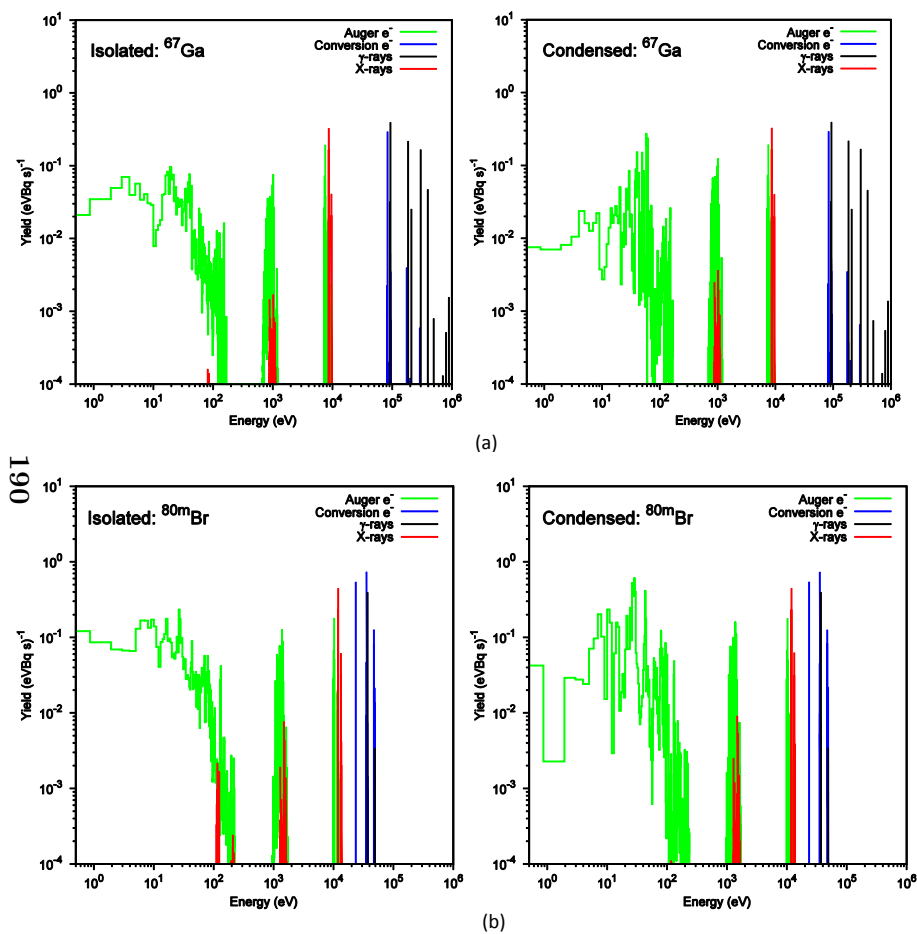
A.7 Atomic states in Kr-like barium

Table A.7: Lowest 37 atomic states in Kr-like barium and their associated energies (relative to the ground state) and lifetimes calculated using the **Grasp2K** package within the Dirac-Fock physics framework (see description described in A.6). No correlation is included in the calculation and only $E1$ and $M2$ transitions are considered. When there is no possible $E1$ or $M2$ transition from an atomic state, the lifetime of the state is left unassigned. It is expected to be long lived.

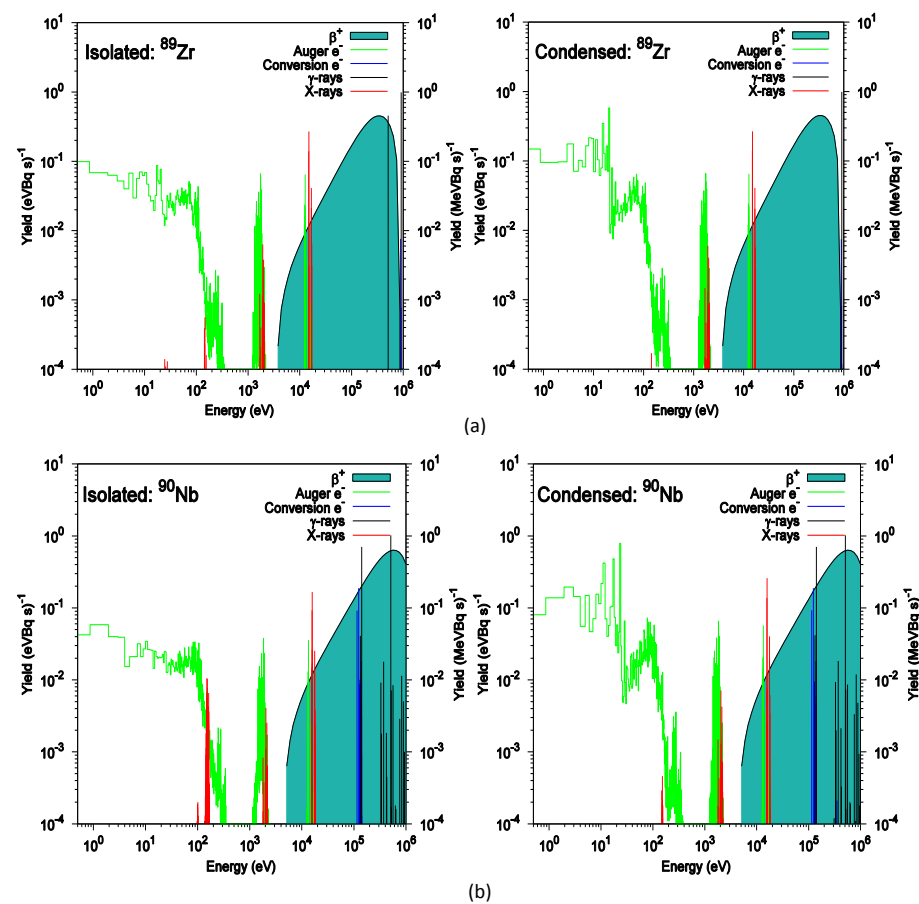
Atomic state	E (eV)	τ (s)
$4s^2 4p^6 {}^1S_0$	0.0000	
$4s^2 4p^5 {}^2P 4d {}^3P_0^o$	85.9874	
$4s^2 4p^5 {}^2P 4d {}^3P_1^o$	87.8635	3.3441E-08
$4s^2 4p^5 {}^2P 4d {}^3F_3^o$	90.9011	
$4s^2 4p^5 {}^2P 4d {}^3D_2^o$	91.1997	2.6814E-03
$4s^2 4p^5 {}^2P 4d {}^3F_4^o$	92.4362	
$4s^2 4p^5 {}^2P 4d {}^1D_2^o$	93.9084	8.5194E-04
$4s^2 4p^5 {}^2P 4d {}^3D_3^o$	97.5568	
$4s^2 4p^5 {}^2P 4d {}^3D_1^o$	103.5233	1.4804E-11
$4s^2 4p^5 {}^2P 4d {}^3F_2^o$	109.1667	5.4957E-03
$4s^2 4p^5 {}^2P 4d {}^3D_2^o$	113.3159	4.4071E-03
$4s^2 4p^5 {}^2P 4d {}^1F_3^o$	114.8097	
$4s^2 4p^5 {}^2P 4d {}^1P_1^o$	129.3735	8.4924E-13
$4s^2 4p^5 {}^2P 5s {}^3P_2^o$	258.1201	7.1825E-05
$4s^2 4p^5 {}^2P 5s {}^1P_1^o$	259.2535	2.3043E-12
$4s^2 4p^5 {}^2P 5s {}^3P_0^o$	277.5273	
$4s^2 4p^5 {}^2P 5s {}^3P_1^o$	278.1454	4.4670E-12
$4s^2 4p^5 {}^2P 6p {}^3S_1$	282.4742	2.7654E-12
$4s^2 4p^5 {}^2P 6p {}^3D_2$	283.3554	2.7861E-12
$4s^2 4p^5 {}^2P 6p {}^3D_3$	289.9302	2.9583E-12
$4s^2 4p^5 {}^2P 6p {}^1P_1$	290.1184	3.1609E-12
$4s^2 4p^5 {}^2P 6p {}^3P_2$	291.7428	2.9966E-12
$4s^2 4p^5 {}^2P 6p {}^3P_0$	295.2618	3.3679E-12
$4s^2 4p^5 {}^2P 6p {}^3D_1$	302.4590	2.8323E-12
$4s^2 4p^5 {}^2P 6p {}^1S_0$	306.9655	3.3015E-12
$4s^2 4p^5 {}^2P 6p {}^3P_1$	309.8890	2.9894E-12
$4s^2 4p^5 {}^2P 6p {}^3D_2$	310.0924	2.9936E-12
$4s^2 4p^5 {}^2P 5p {}^1P_1$	417.2476	5.9658E-12
$4s^2 4p^5 {}^2P 5p {}^3D_3$	417.2689	5.7841E-12
$4s^2 4p^5 {}^2P 5p {}^3P_2$	418.1010	5.8093E-12
$4s^2 4p^5 {}^2P 5p {}^1S_0$	419.4450	6.8728E-12
$4s^2 4p^5 {}^2P 5p {}^3P_1$	436.9260	5.8022E-12
$4s^2 4p^5 {}^2P 5p {}^3D_2$	437.0544	5.8222E-12
$4s^2 4p^5 {}^2P 5p {}^3P_1$	484.0243	8.1602E-12
$4s^2 4p^5 {}^2P 5p {}^3D_2$	484.0964	8.2070E-12
$4s^2 4p^5 {}^2P 5p {}^3D_1$	503.5007	8.1710E-12
$4s^2 4p^5 {}^2P 5p {}^3P_0$	504.2156	8.9518E-12

Appendix B

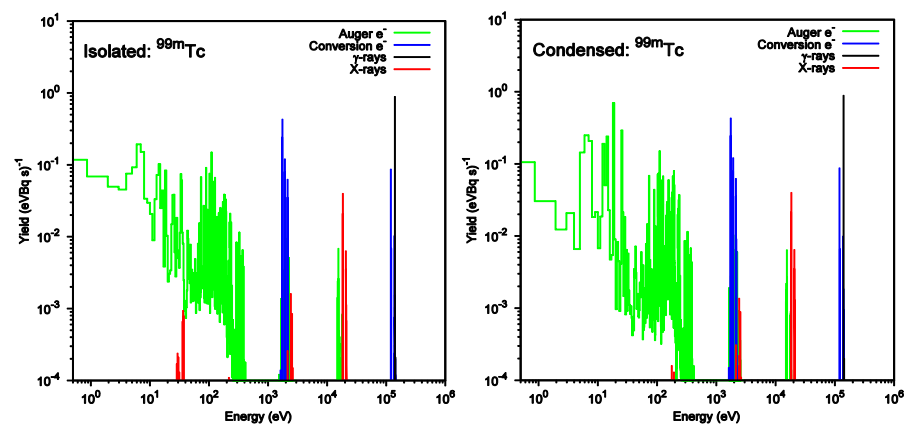
B.1 Supplementary figures of Chapter 7



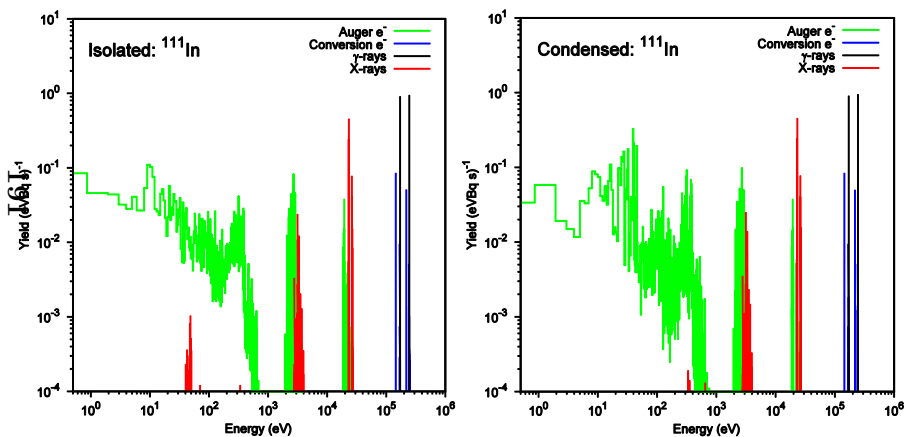
Supplementary Figure 1. Energy spectra of nuclear and atomic radiations of (a) ^{67}Ga and (b) $^{80\text{m}}\text{Br}$ in isolated-atom and condensed-phase approximations within 0.5 and 10^6 eV range. All spectra are binned using 1 eV bin size.



Supplementary Figure 2. Energy spectra of nuclear and atomic radiations of (a) ^{89}Zr and (b) ^{90}Nb in isolated-atom and condensed-phase approximations within 0.5 and 10^6 eV range. All spectra are binned using 1 eV bin size except β^+ spectra. β^+ spectra are extracted from MIRD-RADTABS program (Eckerman and Endo, 2008a) and plotted on the right y-axis.

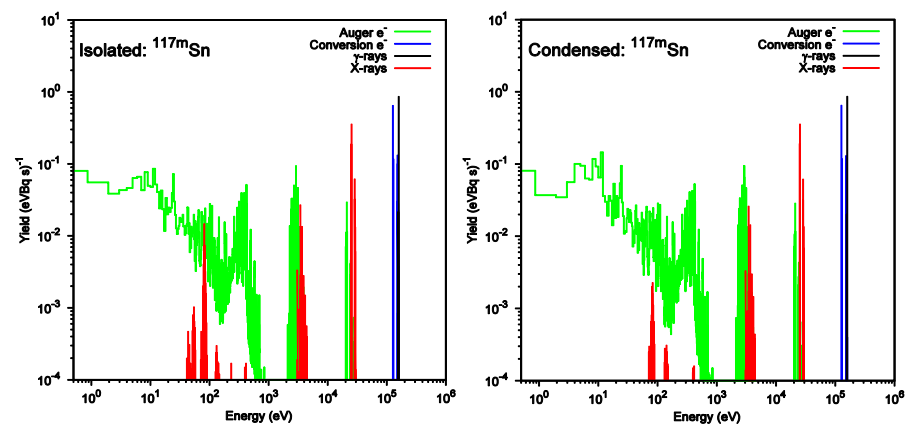


(a)

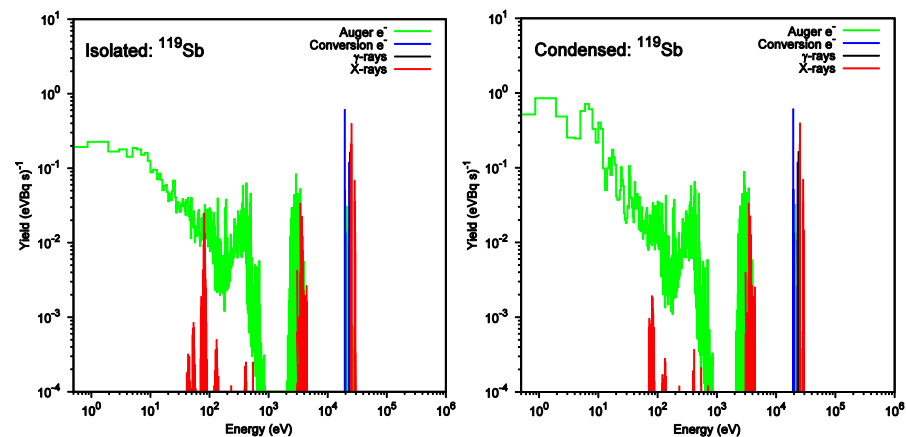


(b)

Supplementary Figure 3. Energy spectra of nuclear and atomic radiations of (a) ^{99m}Tc and (b) ^{111}In in isolated-atom and condensed-phase approximations within 0.5 and 10^6 eV range. All spectra are binned using 1 eV bin size.

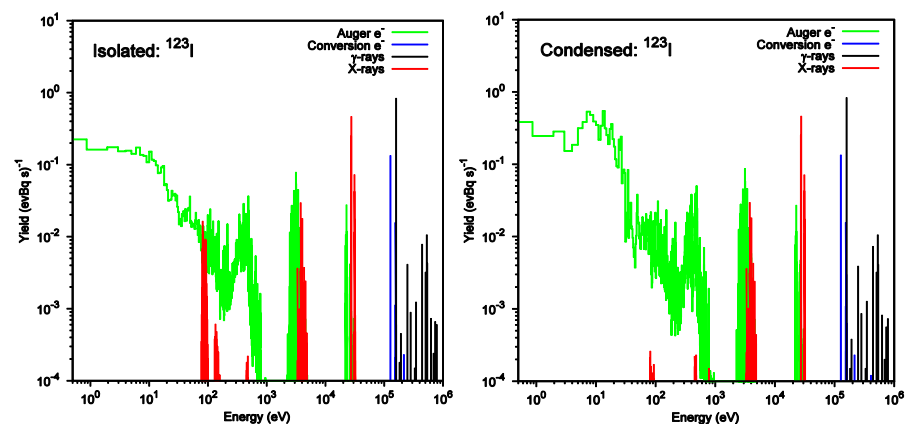


(a)

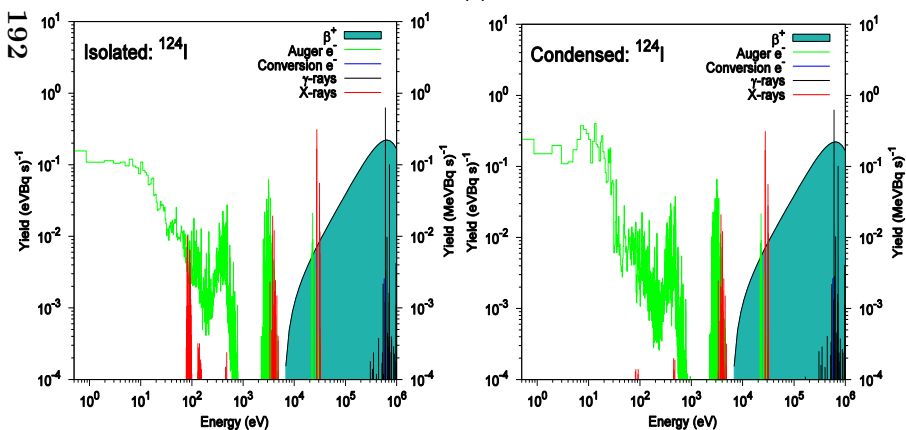


(b)

Supplementary Figure 4. Energy spectra of nuclear and atomic radiations of (a) ^{117m}Sn and (b) ^{119}Sb in isolated-atom and condensed-phase approximations within 0.5 and 10^6 eV range. All spectra are binned using 1 eV bin size.

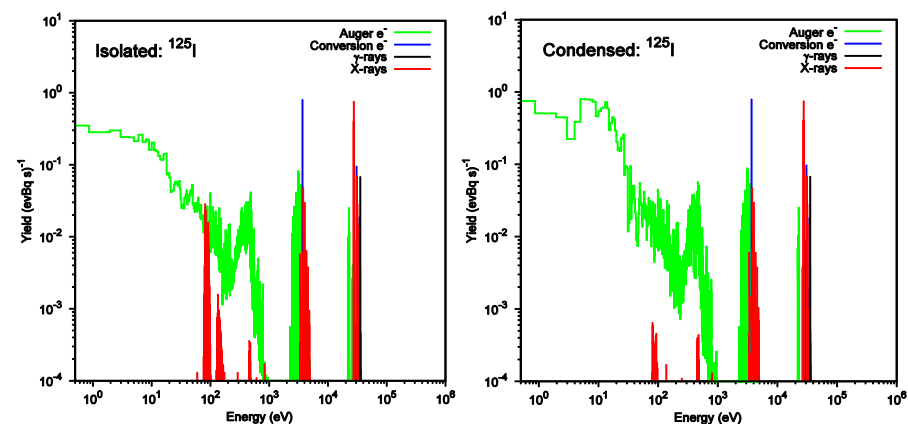


(a)

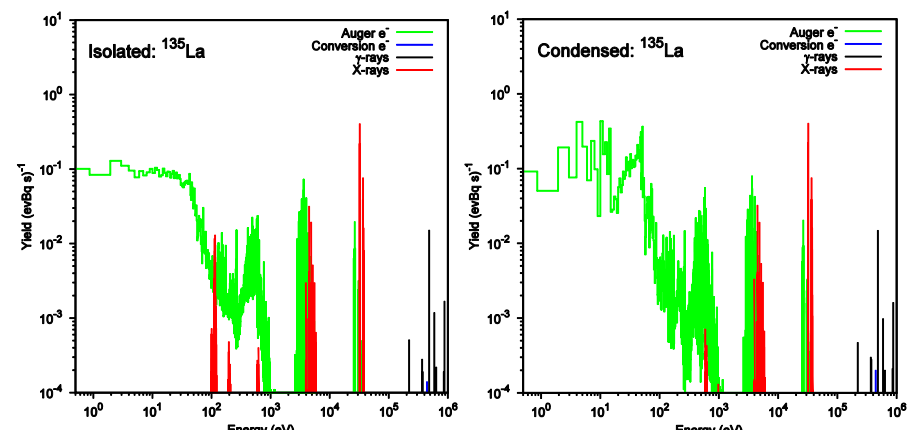


(b)

Supplementary Figure 5. Energy spectra of nuclear and atomic radiations of (a) ^{123}I and (b) ^{124}I in isolated-atom and condensed-phase approximations within 0.5 and 10^6 eV range. All spectra are binned using 1 eV bin size. β^+ spectra are extracted from MIRD-RADTABS program (Eckerman and Endo, 2008a) and plotted on the right y-axis.

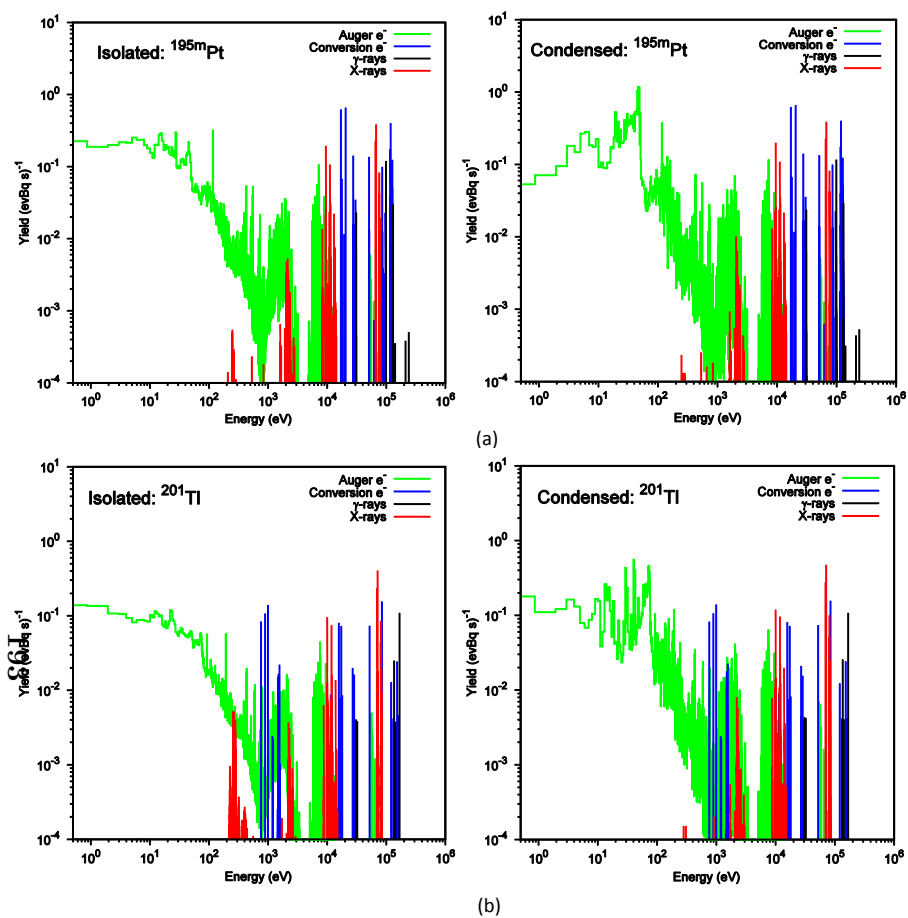


(a)

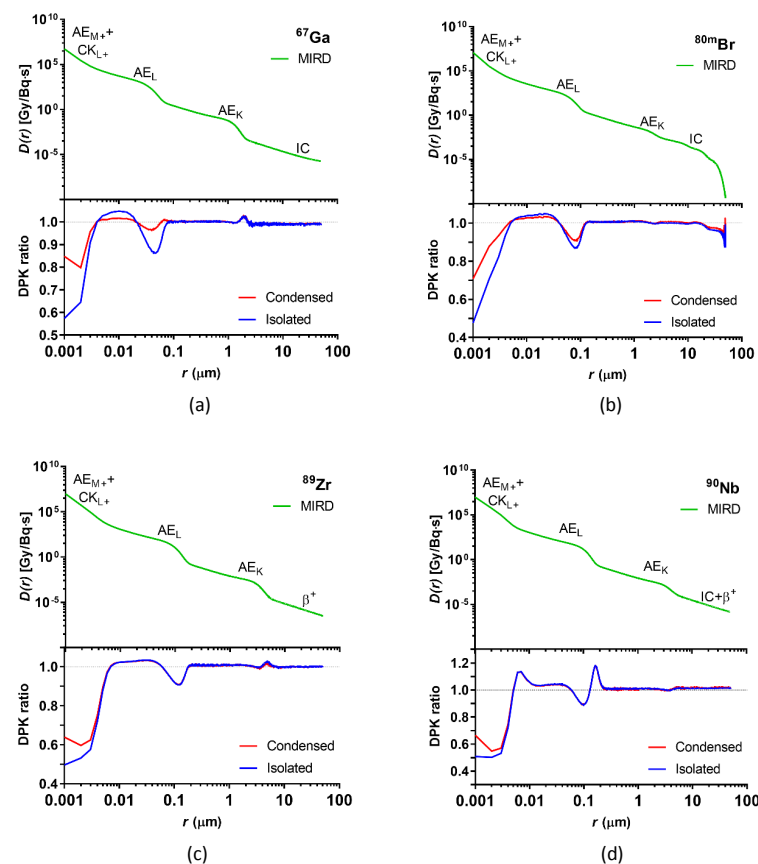


(b)

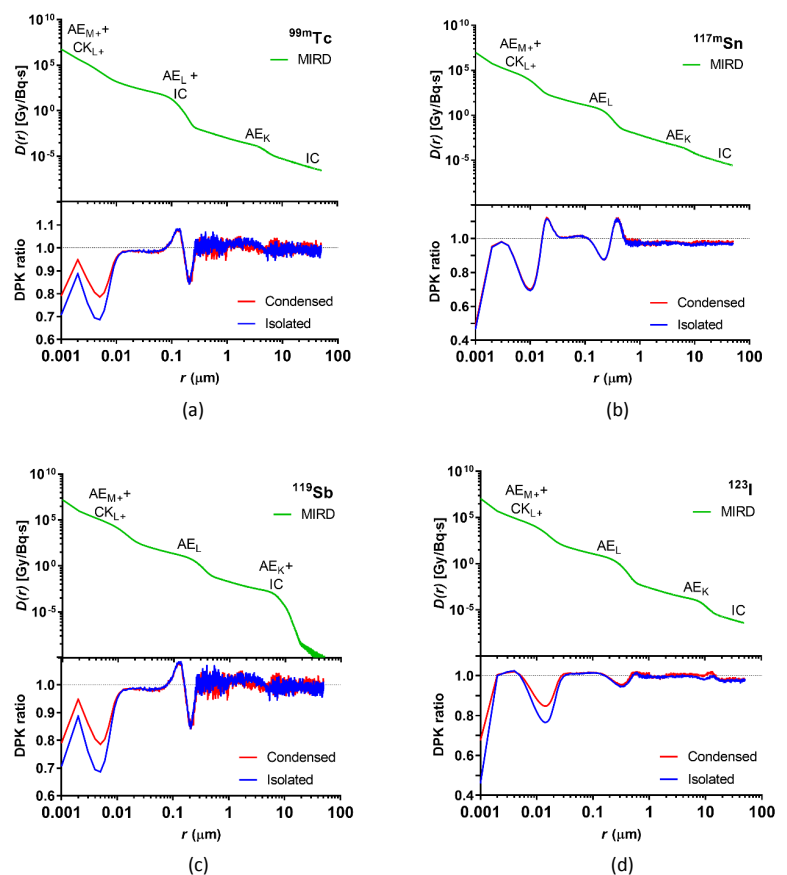
Supplementary Figure 6. Energy spectra of nuclear and atomic radiations of (a) ^{125}I and (b) ^{135}La in isolated-atom and condensed-phase approximations within 0.5 and 10^6 eV range. All spectra are binned using 1 eV bin size.



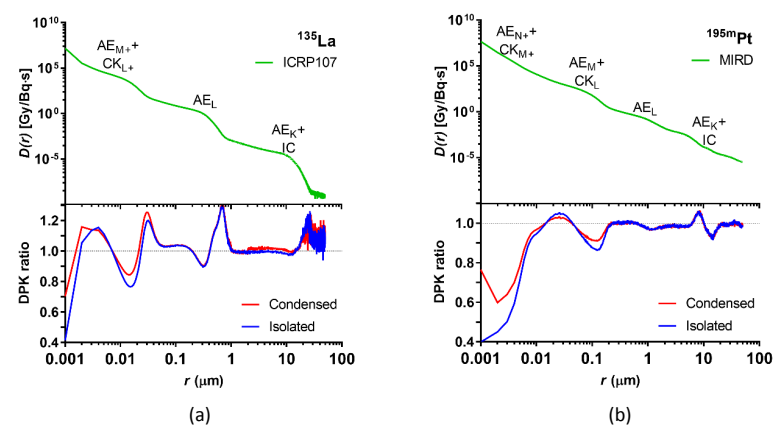
Supplementary Figure 7. Energy spectra of nuclear and atomic radiations of (a) ^{195m}Pt and (b) ^{201}Tl in isolated-atom and condensed-phase approximations within 0.5 and 10^6 eV range. All spectra are binned using 1 eV bin size.



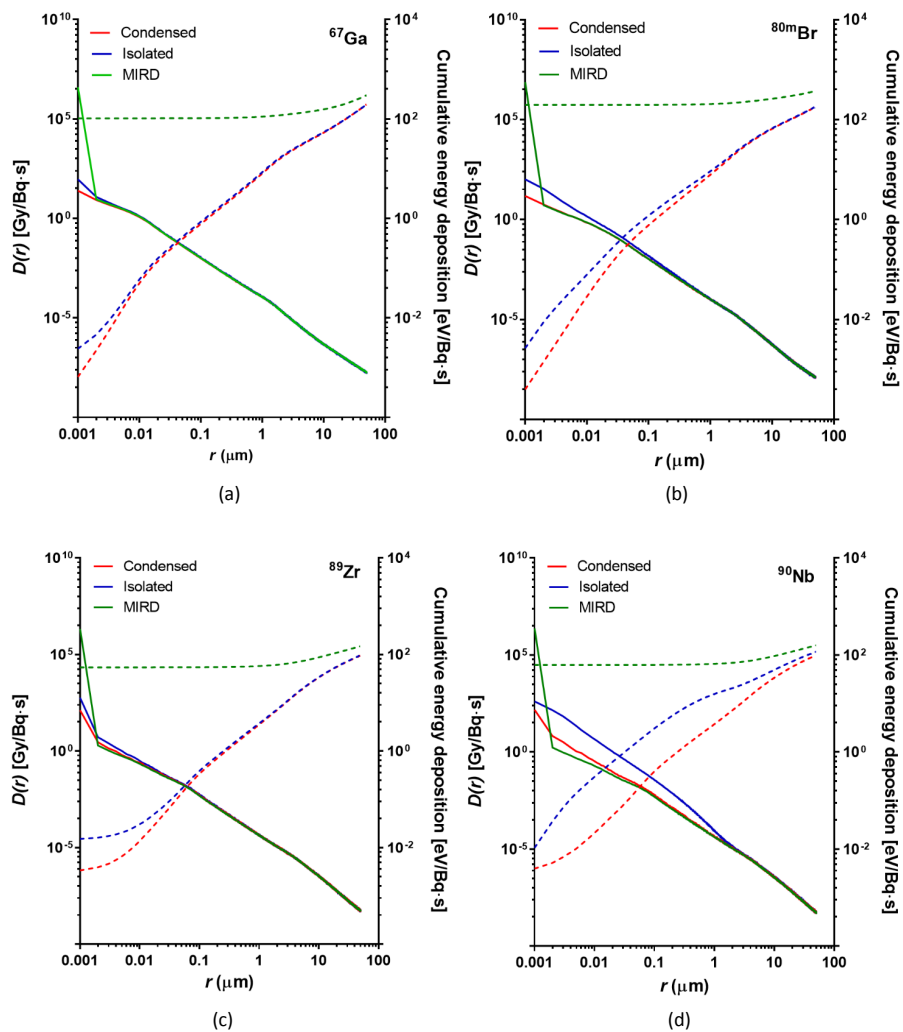
Supplementary Figure 8. DPKs (Gy/Bq·s) for the AE+CK+IC+ β^+ spectra from MIRD-RADTABS (Eckerman and Endo, 2008a) of (a) ^{67}Ga , (b) ^{80m}Br , (c) ^{89}Zr and (d) ^{90}Nb plotted on the top panel as a function of distance from a point source (solid lines). DPK ratios, which are defined as $\text{DPK}_{\text{BrIccEmis}}/\text{DPK}_{\text{MIRD-RADTABS}}$, comparing dose distribution from MIRD-RADTABS data with spectra generated from *BrIccEmis* using condensed-phase and isolated-atom approximations plotted on the bottom panel.



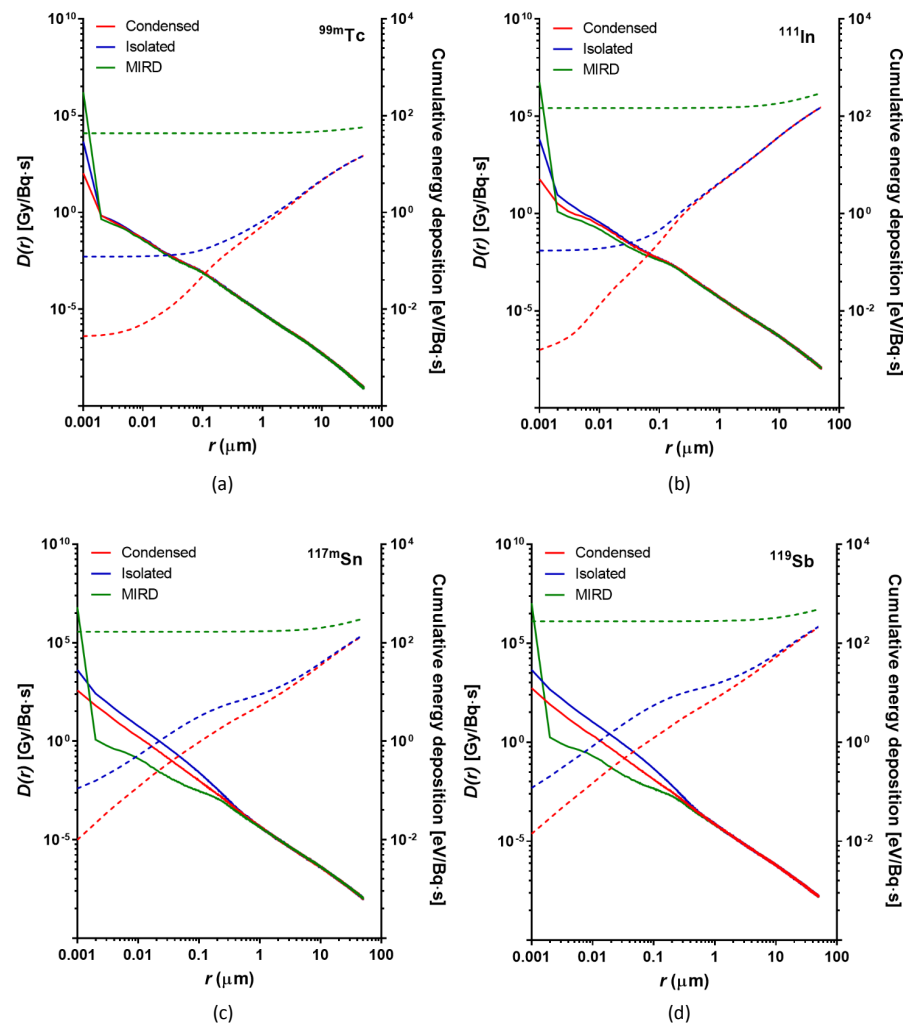
Supplementary Figure 9. DPKs (Gy/Bq·s) for the AE+CK+IC spectra from MIRD-RADTABS (Eckerman and Endo, 2008a) of (a) $^{99\text{m}}\text{Tc}$, (b) $^{117\text{m}}\text{Sn}$, (c) ^{119}Sb and (d) ^{123}I plotted on the top panel as a function of distance from a point source. DPK ratios, which are defined as $\text{DPK}_{\text{BrIccEmis}} / \text{DPK}_{\text{MIRD-RADTABS}}$, comparing dose distribution from MIRD-RADTABS data with spectra generated from *BrIccEmis* using condensed-phase and isolated-atom approximations plotted on the bottom panel.



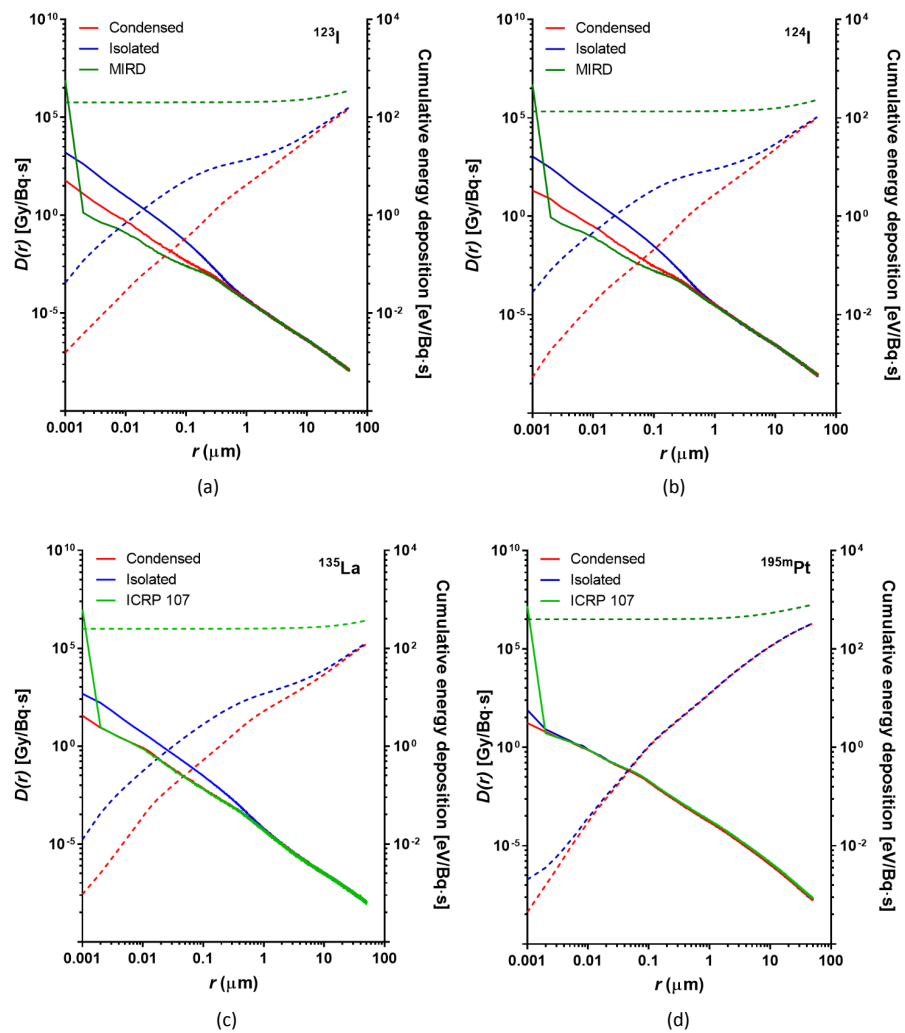
Supplementary Figure 10. DPKs (Gy/Bq·s) for the AE+CK+IC spectra from ICRP 107 (Eckerman and Endo, 2008b) and MIRD-RADTABS (Eckerman and Endo, 2008a) of (a) ^{135}La and (b) $^{195\text{m}}\text{Pt}$ plotted on the top panel as a function of distance from a point source. DPK ratios, which are defined as $\text{DPK}_{\text{BrIccEmis}} / \text{DPK}_{\text{MIRD-RADTABS}}$, comparing dose distribution from MIRD-RADTABS and ICRP 107 data with spectra generated from *BrIccEmis* using condensed-phase and isolated-atom approximations plotted on the bottom panel.



Supplementary Figure 11. DPKs ($\text{Gy/Bq}\cdot\text{s}$) for the X-ray spectra of (a) ^{67}Ga , (b) $^{80\text{m}}\text{Br}$, (c) ^{89}Zr and (d) ^{90}Nb from *BrIccEmis* and MIRD-RADTABS (Eckerman and Endo, 2008a) plotted on the left y-axis as a function of distance from a



Supplementary Figure 12. DPKs ($\text{Gy/Bq}\cdot\text{s}$) for the X-ray spectra of (a) $^{99\text{m}}\text{Tc}$, (b) ^{111}In , (c) $^{117\text{m}}\text{Sn}$ and (d) ^{119}Sb from *BrIccEmis* and MIRD-RADTABS (Eckerman and Endo, 2008a) plotted on the left y-axis as a function of distance from a



Supplementary Figure 13. DPKs (Gy/Bq·s) for the x-ray spectra of (a) ^{123}I , (b) ^{124}I , (c) ^{135}La and (d) $^{195\text{m}}\text{Pt}$ from *BrIccEmis*, MIRD-RADTABS (Eckerman and Endo, 2008a) and ICRP 107 (Eckerman and Endo, 2008b) plotted on the left y-axis as

B.2 Supplementary tables of Chapter 7

Supplementary Table 1-6. Cellular S-values (Gy/Bq·s) for ^{67}Ga , $^{80\text{m}}\text{Br}$, ^{89}Zr , ^{90}Nb , $^{99\text{m}}\text{Tc}$, ^{111}In , $^{117\text{m}}\text{Sn}$, ^{119}Sb , ^{123}I , ^{125}I , $^{195\text{m}}\text{Pt}$ and ^{201}Tl calculated from event-by-event MC simulation: Comparison of S-values derived from *BrIccEmis* and MIRD-RADTABS (Falzone *et al.*, 2015) decay data.

^{67}Ga									$^{80\text{m}}\text{Br}$					
R_c	R_N	S _{Condensed spectra} / S _{MIRD}			S _{Isolated spectra} / S _{MIRD}				S _{Condensed spectra} / S _{MIRD}			S _{Isolated spectra} / S _{MIRD}		
(μm)	(μm)	S(N←N)	S(N←Cy)	S(N←CS)	S(N←N)	S(N←Cy)	S(N←CS)		S(N←N)	S(N←Cy)	S(N←CS)	S(N←N)	S(N←Cy)	S(N←CS)
3	1	0.99	1.00	0.99	0.97	1.00	0.99		0.97	1.01	1.00	0.95	1.01	1.00
3	2	0.99	1.00	1.00	0.97	1.00	1.00		0.99	1.00	1.00	0.97	1.00	1.00
4	2	0.99	1.00	0.99	0.97	1.00	0.99		0.99	1.00	1.00	0.97	1.00	1.00
4	3	0.99	1.00	1.00	0.98	1.00	1.00		0.99	1.01	1.00	0.98	1.00	1.00
5	2	0.99	1.00	0.99	0.97	1.00	0.99		0.99	1.00	1.00	0.97	1.00	1.00
5	3	0.99	1.00	0.99	0.98	1.00	0.99		0.99	1.00	1.01	0.98	1.00	1.00
5	4	0.99	1.00	1.00	0.98	1.00	0.99		0.99	1.01	1.01	0.98	1.00	1.00
6	3	0.99	1.00	0.99	0.98	1.00	0.99		0.99	1.01	1.00	0.98	1.01	1.00
6	4	0.99	1.00	0.99	0.98	1.00	0.99		0.99	1.00	1.01	0.98	1.00	1.00
6	5	0.99	1.00	1.00	0.98	1.00	1.00		1.00	1.00	1.01	0.99	1.00	1.00
7	3	0.99	1.00	0.99	0.98	1.00	0.99		0.99	1.01	1.01	0.98	1.00	1.00
7	4	0.99	1.00	0.99	0.98	1.00	0.99		0.99	1.00	1.01	0.98	1.00	1.00
7	5	0.99	1.00	0.99	0.98	1.00	0.99		1.00	1.00	1.01	0.99	0.99	1.00
7	6	0.99	1.00	1.00	0.98	0.99	0.99		1.00	1.01	1.00	0.99	1.01	1.00
8	4	0.99	1.00	0.99	0.98	1.00	0.99		0.99	1.01	1.01	0.98	1.00	1.00
8	5	0.99	1.00	0.99	0.98	0.99	0.99		1.00	1.01	1.00	0.99	1.00	1.00
8	6	0.99	0.99	0.99	0.98	0.99	0.99		1.00	1.01	1.01	0.99	1.00	1.00
8	7	0.99	1.00	1.00	0.98	1.00	0.99		0.99	1.01	1.01	0.99	1.00	1.00
9	5	0.99	1.00	0.99	0.98	1.00	0.99		1.00	1.01	1.00	0.99	1.00	1.00
9	6	0.99	1.00	0.99	0.98	0.99	0.99		1.00	1.00	1.00	0.99	1.00	1.00
9	7	0.99	1.00	0.99	0.98	1.00	0.99		0.99	1.01	1.01	0.99	1.00	1.00
9	8	0.99	1.00	1.00	0.98	1.00	0.99		1.00	1.00	1.00	0.99	1.00	1.00
10	5	0.99	0.99	0.99	0.98	0.99	0.99		1.00	1.00	1.01	0.99	1.00	1.00
10	6	0.99	1.00	0.99	0.98	0.99	0.99		1.00	1.00	1.00	0.99	1.00	1.00
10	7	0.99	1.00	0.99	0.98	0.99	0.99		0.99	1.00	1.01	0.99	1.00	1.00
10	8	0.99	1.00	0.99	0.98	0.99	0.99		1.00	1.01	1.01	0.99	1.00	1.00
10	9	0.99	1.00	0.99	0.98	1.00	0.99		1.00	1.00	1.00	0.99	1.00	1.00

Supplementary Table 2

⁸⁹ Zr								⁹⁰ Nb					
<i>R</i> _C	<i>R</i> _N	<i>S</i> _{Condensed spectra} / <i>S</i> _{MIRD}			<i>S</i> _{Isolated spectra} / <i>S</i> _{MIRD}			<i>S</i> _{Condensed spectra} / <i>S</i> _{MIRD}			<i>S</i> _{Isolated spectra} / <i>S</i> _{MIRD}		
(μm)	(μm)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)
3	1	0.92	1.00	1.00	0.90	1.01	1.00	0.92	1.00	0.99	0.90	1.01	0.99
3	2	0.95	1.01	1.00	0.93	1.01	1.00	0.94	1.00	0.98	0.92	1.00	0.98
4	2	0.95	1.01	1.00	0.93	1.01	1.00	0.94	1.00	0.99	0.92	1.00	1.00
4	3	0.95	1.00	1.00	0.94	1.00	1.01	0.95	1.00	0.99	0.94	1.00	0.99
5	2	0.95	1.00	1.00	0.93	1.00	1.01	0.94	1.01	1.00	0.92	1.02	1.00
5	3	0.95	1.00	1.00	0.94	1.00	1.00	0.95	1.01	0.99	0.94	1.01	1.00
5	4	0.95	1.00	1.00	0.94	1.01	1.01	0.95	1.00	0.99	0.94	1.00	0.99
6	3	0.95	1.00	1.00	0.94	1.00	1.01	0.95	1.00	1.00	0.94	1.00	1.00
6	4	0.95	1.00	1.00	0.94	1.00	1.00	0.95	1.00	1.00	0.94	1.00	1.00
6	5	0.96	1.00	1.00	0.95	1.01	1.00	0.95	1.00	0.99	0.94	1.00	1.00
7	3	0.95	1.00	1.00	0.94	1.00	1.01	0.95	1.00	1.01	0.94	1.01	1.00
7	4	0.95	1.00	1.00	0.94	1.00	1.00	0.95	1.00	1.00	0.94	1.00	1.00
7	5	0.96	1.00	1.00	0.95	1.00	1.00	0.95	0.99	1.00	0.94	0.99	1.00
7	6	0.96	1.00	1.00	0.95	1.00	1.00	0.96	1.00	1.00	0.95	1.01	1.00
8	4	0.95	1.00	1.00	0.94	1.01	1.01	0.95	1.00	1.01	0.94	1.00	1.00
8	5	0.96	1.00	1.00	0.95	1.00	1.01	0.95	0.99	1.00	0.94	1.00	1.00
8	6	0.96	1.00	1.00	0.95	1.00	1.00	0.96	1.00	1.00	0.95	1.00	1.00
8	7	0.96	1.00	1.00	0.95	1.00	1.00	0.96	1.00	1.00	0.95	1.00	1.00
9	5	0.96	1.00	1.00	0.95	1.00	1.00	0.95	1.01	1.00	0.94	1.01	1.00
9	6	0.96	1.00	1.00	0.95	1.00	1.01	0.96	1.00	1.01	0.95	1.00	1.01
9	7	0.96	1.00	1.00	0.95	1.00	1.00	0.96	1.00	1.00	0.95	1.00	1.00
9	8	0.96	1.00	1.00	0.95	1.00	1.01	0.96	1.01	1.00	0.95	1.01	1.00
10	5	0.96	1.00	1.00	0.95	1.01	1.00	0.95	1.00	1.00	0.94	1.00	1.00
10	6	0.96	1.00	1.00	0.95	1.00	1.00	0.96	1.00	1.00	0.95	1.00	1.00
10	7	0.96	1.00	1.00	0.95	1.00	1.00	0.96	1.00	1.00	0.95	1.00	1.00
10	8	0.96	1.00	1.00	0.95	1.00	1.01	0.96	1.00	1.00	0.95	1.01	1.00
10	9	0.96	1.00	1.00	0.95	1.00	1.01	0.98	1.01	1.01	0.94	1.00	1.00

Supplementary Table 3

^{99m} Tc								¹¹¹ In					
<i>R_C</i>	<i>R_N</i>	<i>S</i> _{Condensed spectra / <i>S</i>_{MIRD}}			<i>S</i> _{Isolated spectra / <i>S</i>_{MIRD}}			<i>S</i> _{Condensed spectra / <i>S</i>_{MIRD}}			<i>S</i> _{Isolated spectra / <i>S</i>_{MIRD}}		
(μm)	(μm)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)
3	1	0.97	1.01	1.00	0.96	1.02	1.02	0.97	0.99	1.00	0.94	0.99	1.00
3	2	0.97	1.01	1.00	0.96	1.02	1.02	0.97	0.99	1.00	0.94	1.00	1.00
4	2	0.97	1.00	1.00	0.96	1.02	1.01	0.97	0.99	0.99	0.94	1.00	1.00
4	3	0.98	1.00	1.00	0.96	1.02	1.01	0.97	0.99	0.99	0.95	1.00	1.00
5	2	0.97	1.00	1.00	0.96	1.01	1.00	0.97	0.99	0.99	0.94	1.00	0.99
5	3	0.98	1.00	1.00	0.96	1.01	1.00	0.97	0.99	0.99	0.95	0.99	1.00
5	4	0.97	1.00	1.00	0.96	1.01	1.01	0.97	0.99	0.99	0.95	0.99	0.99
6	3	0.98	1.00	1.00	0.96	1.01	1.00	0.97	0.99	0.99	0.95	0.99	1.00
6	4	0.97	1.00	1.00	0.96	1.01	1.00	0.97	0.99	0.99	0.95	1.00	0.99
6	5	0.98	1.00	1.00	0.97	1.01	1.01	0.97	0.99	0.99	0.96	1.00	1.00
7	3	0.98	1.00	1.00	0.96	1.01	0.99	0.97	0.99	0.98	0.95	1.00	0.99
7	4	0.97	1.00	1.00	0.96	1.01	0.99	0.97	0.99	0.99	0.95	1.00	1.00
7	5	0.98	1.00	1.00	0.97	1.01	1.00	0.97	0.99	0.99	0.96	0.99	0.99
7	6	0.98	1.00	1.00	0.97	1.01	1.00	0.98	0.99	0.99	0.96	1.00	1.00
8	4	0.97	1.00	1.00	0.96	1.01	0.99	0.97	0.99	0.99	0.95	0.99	0.99
8	5	0.98	1.00	1.00	0.97	1.00	1.00	0.97	0.99	0.99	0.96	1.00	0.99
8	6	0.98	1.00	1.00	0.97	1.01	1.00	0.98	0.99	0.99	0.96	0.99	0.99
8	7	0.98	1.00	1.00	0.97	1.00	1.00	0.98	0.99	0.99	0.96	1.00	0.99
9	5	0.98	1.00	1.00	0.97	1.00	0.99	0.97	0.99	0.99	0.96	0.99	0.99
9	6	0.98	1.00	1.00	0.97	1.00	0.99	0.98	0.99	0.99	0.96	0.99	0.99
9	7	0.98	1.00	1.00	0.97	1.00	1.00	0.98	0.99	0.99	0.96	0.99	0.99
9	8	0.98	1.00	1.00	0.97	1.01	1.00	0.98	0.99	0.99	0.96	0.99	0.99
10	5	0.98	1.00	1.00	0.97	1.00	0.99	0.97	0.99	0.98	0.96	0.99	0.99
10	6	0.98	1.00	1.00	0.97	1.00	0.99	0.98	0.99	0.98	0.96	0.99	0.99
10	7	0.98	1.00	1.00	0.97	1.00	0.99	0.98	0.99	0.99	0.96	0.99	0.99
10	8	0.98	1.00	1.00	0.97	1.00	1.00	0.98	0.99	0.99	0.96	0.99	0.99
10	9	0.98	1.00	1.00	0.97	1.00	1.00	0.98	0.99	0.99	0.96	0.99	0.99

Supplementary Table 4

^{117m} Sn								¹¹⁹ Sb					
<i>R_C</i>	<i>R_N</i>	<i>S</i> _{Condensed spectra / <i>S</i>_{MIRD}}			<i>S</i> _{Isolated spectra / <i>S</i>_{MIRD}}			<i>S</i> _{Condensed spectra / <i>S</i>_{MIRD}}			<i>S</i> _{Isolated spectra / <i>S</i>_{MIRD}}		
(μm)	(μm)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)	<i>S</i> (N←N)	<i>S</i> (N←Cy)	<i>S</i> (N←CS)
3	1	0.90	0.97	0.98	0.90	0.97	0.97	0.95	1.00	1.01	0.94	1.00	1.00
3	2	0.91	0.97	0.98	0.91	0.97	0.97	0.96	1.00	1.00	0.95	1.00	1.00
4	2	0.91	0.98	0.98	0.91	0.97	0.97	0.96	1.00	1.00	0.95	1.00	1.00
4	3	0.92	0.97	0.98	0.92	0.97	0.97	0.97	1.00	1.00	0.96	1.00	1.00
5	2	0.91	0.98	0.98	0.91	0.97	0.97	0.96	1.00	1.00	0.95	1.00	1.00
5	3	0.92	0.97	0.97	0.92	0.97	0.97	0.97	1.00	1.01	0.96	0.99	1.00
5	4	0.93	0.97	0.98	0.92	0.97	0.97	0.97	1.00	1.00	0.96	1.00	0.99
6	3	0.92	0.97	0.97	0.92	0.97	0.97	0.97	1.01	1.00	0.96	1.00	0.99
6	4	0.93	0.97	0.97	0.92	0.97	0.97	0.97	1.00	1.00	0.96	1.00	1.00
6	5	0.93	0.98	0.97	0.92	0.97	0.97	0.98	1.00	1.00	0.97	0.99	0.99
7	3	0.92	0.98	0.97	0.92	0.97	0.97	0.97	1.00	0.99	0.96	0.99	0.99
7	4	0.93	0.97	0.97	0.92	0.97	0.97	0.97	1.00	1.00	0.96	1.00	0.99
7	5	0.93	0.97	0.97	0.92	0.97	0.97	0.98	1.00	1.00	0.97	0.99	0.99
7	6	0.93	0.97	0.98	0.93	0.97	0.97	0.98	1.00	1.00	0.97	0.99	0.99
8	4	0.93	0.97	0.97	0.92	0.97	0.96	0.97	1.00	0.99	0.96	0.99	0.99
8	5	0.93	0.97	0.97	0.92	0.97	0.97	0.98	1.00	0.99	0.97	0.99	0.99
8	6	0.93	0.98	0.97	0.93	0.97	0.96	0.98	1.00	1.00	0.97	0.99	0.99
8	7	0.94	0.97	0.98	0.93	0.97	0.97	0.98	1.00	1.00	0.97	0.99	0.99
9	5	0.93	0.97	0.97	0.92	0.97	0.97	0.98	1.00	0.99	0.97	0.99	0.99
9	6	0.93	0.97	0.97	0.93	0.97	0.96	0.98	1.00	0.99	0.97	0.99	0.99
9	7	0.94	0.97	0.98	0.93	0.97	0.97	0.98	1.00	1.00	0.97	0.99	0.99
9	8	0.94	0.97	0.97	0.93	0.97	0.97	0.98	1.00	0.99	0.97	0.99	0.99
10	5	0.93	0.97	0.97	0.92	0.97	0.97	0.98	1.00	0.98	0.97	0.99	0.98
10	6	0.93	0.97	0.97	0.93	0.97	0.97	0.98	1.00	0.99	0.97	0.99	0.98
10	7	0.94	0.98	0.97	0.93	0.97	0.97	0.98	1.00	0.99	0.97	0.99	0.99
10	8	0.94	0.97	0.97	0.93	0.97	0.97	0.98	1.00	0.99	0.97	0.99	0.99
10	9	0.98	1.00	1.00	0.97	1.00	1.00	0.98	1.00	1.00	0.97	0.99	0.99

Supplementary Table 5

123I								125I					
R_C	R_N	$S_{\text{Condensed spectra}}/S_{\text{MIRD}}$			$S_{\text{Isolated spectra}}/S_{\text{MIRD}}$			$S_{\text{Condensed spectra}}/S_{\text{MIRD}}$			$S_{\text{Isolated spectra}}/S_{\text{MIRD}}$		
(μm)	(μm)	$S(N \leftarrow N)$	$S(N \leftarrow Cy)$	$S(N \leftarrow CS)$	$S(N \leftarrow N)$	$S(N \leftarrow Cy)$	$S(N \leftarrow CS)$	$S(N \leftarrow N)$	$S(N \leftarrow Cy)$	$S(N \leftarrow CS)$	$S(N \leftarrow N)$	$S(N \leftarrow Cy)$	$S(N \leftarrow CS)$
3	1	0.96	0.99	1.01	0.92	0.98	0.99	0.96	0.98	1.00	0.94	0.98	0.99
3	2	0.96	1.00	1.01	0.93	0.99	1.00	0.96	0.98	1.00	0.94	0.98	0.99
4	2	0.96	1.00	1.01	0.93	0.99	1.00	0.96	0.99	1.00	0.94	0.99	0.99
4	3	0.96	1.00	1.01	0.93	0.99	1.00	0.96	0.99	1.00	0.95	0.98	0.99
5	2	0.96	1.00	1.01	0.93	0.99	0.99	0.96	0.99	0.99	0.94	0.98	0.99
5	3	0.96	1.00	1.01	0.93	0.99	1.00	0.96	0.99	1.00	0.95	0.98	0.99
5	4	0.96	1.00	1.00	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.98	0.99
6	3	0.96	1.00	1.01	0.93	0.99	0.99	0.96	0.99	0.99	0.95	0.99	0.99
6	4	0.96	1.00	1.01	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.98	0.99
6	5	0.96	1.00	1.01	0.94	0.99	1.00	0.96	0.99	0.99	0.95	0.98	0.99
7	3	0.96	1.00	1.01	0.93	0.99	1.00	0.96	0.99	0.99	0.95	0.99	0.99
7	4	0.96	1.00	1.01	0.94	1.00	1.00	0.96	0.99	0.99	0.95	0.99	0.99
7	5	0.96	1.01	1.01	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.99	0.99
7	6	0.97	1.00	1.01	0.94	0.99	1.00	0.96	0.99	1.00	0.95	0.99	0.99
8	4	0.96	1.01	1.01	0.94	1.00	0.99	0.96	0.99	0.99	0.95	0.99	0.99
8	5	0.96	1.01	1.00	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.99	0.99
8	6	0.97	1.00	1.01	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.98	0.99
8	7	0.97	1.01	1.01	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.98	0.98
9	5	0.96	1.00	1.00	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.99	0.98
9	6	0.97	1.00	1.00	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.99	0.98
9	7	0.97	1.00	1.01	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.99	0.98
9	8	0.97	1.00	1.01	0.95	0.99	0.99	0.96	0.99	0.99	0.95	0.98	0.98
10	5	0.96	1.00	1.01	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.99	0.98
10	6	0.97	1.01	1.00	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.98	0.98
10	7	0.97	1.00	1.00	0.94	0.99	0.99	0.96	0.99	0.99	0.95	0.98	0.98
10	8	0.97	1.00	1.01	0.95	0.99	0.99	0.96	0.99	0.99	0.95	0.98	0.98
10	9	0.97	1.00	1.01	0.95	0.99	0.99	0.97	0.99	0.99	0.95	0.98	0.99

Supplementary Table 6

195mPt								201Tl					
R_C	R_N	$S_{\text{Condensed spectra}}/S_{\text{MIRD}}$			$S_{\text{Isolated spectra}}/S_{\text{MIRD}}$			$S_{\text{Condensed spectra}}/S_{\text{MIRD}}$			$S_{\text{Isolated spectra}}/S_{\text{MIRD}}$		
(μm)	(μm)	$S(N \leftarrow N)$	$S(N \leftarrow Cy)$	$S(N \leftarrow CS)$	$S(N \leftarrow N)$	$S(N \leftarrow Cy)$	$S(N \leftarrow CS)$	$S(N \leftarrow N)$	$S(N \leftarrow Cy)$	$S(N \leftarrow CS)$	$S(N \leftarrow N)$	$S(N \leftarrow Cy)$	$S(N \leftarrow CS)$
3	1	0.93	0.98	0.98	0.89	0.98	0.98	0.92	0.98	0.96	0.82	0.97	0.96
3	2	0.94	0.98	0.98	0.91	0.98	0.98	0.94	0.97	0.97	0.85	0.97	0.96
4	2	0.94	0.98	0.98	0.91	0.98	0.98	0.94	0.97	0.95	0.85	0.96	0.95
4	3	0.95	0.98	0.98	0.92	0.98	0.98	0.94	0.97	0.96	0.86	0.96	0.96
5	2	0.94	0.98	0.98	0.91	0.98	0.99	0.94	0.97	0.94	0.85	0.96	0.94
5	3	0.95	0.98	0.99	0.92	0.98	0.99	0.94	0.97	0.95	0.86	0.96	0.95
5	4	0.96	0.98	0.98	0.93	0.98	0.99	0.94	0.97	0.96	0.87	0.96	0.96
6	3	0.95	0.98	0.99	0.92	0.99	0.99	0.94	0.96	0.95	0.86	0.96	0.94
6	4	0.96	0.98	0.99	0.93	0.98	0.99	0.94	0.96	0.95	0.87	0.96	0.95
6	5	0.95	0.99	0.99	0.93	0.99	0.99	0.94	0.97	0.96	0.87	0.96	0.96
7	3	0.95	0.98	1.00	0.92	0.99	1.00	0.94	0.96	0.95	0.86	0.96	0.94
7	4	0.96	0.99	1.00	0.93	0.99	1.00	0.94	0.96	0.95	0.87	0.96	0.94
7	5	0.95	0.99	0.99	0.93	0.99	0.99	0.94	0.96	0.95	0.87	0.96	0.94
7	6	0.96	0.99	0.98	0.94	0.99	0.99	0.94	0.97	0.96	0.88	0.96	0.95
8	4	0.96	0.99	1.01	0.93	0.99	1.01	0.94	0.96	0.95	0.87	0.95	0.94
8	5	0.95	0.99	1.00	0.93	1.00	1.00	0.94	0.96	0.94	0.87	0.95	0.94
8	6	0.96	0.99	0.99	0.94	0.99	1.00	0.94	0.96	0.95	0.88	0.95	0.94
8	7	0.96	0.99	0.99	0.94	0.99	0.99	0.94	0.96	0.95	0.88	0.96	0.95
9	5	0.95	0.99	1.00	0.93	0.99	1.00	0.94	0.96	0.94	0.87	0.95	0.93
9	6	0.96	1.00	1.00	0.94	1.00	1.00	0.94	0.96	0.94	0.88	0.96	0.93
9	7	0.96	0.99	0.99	0.94	0.99	1.00	0.94	0.96	0.94	0.88	0.95	0.94
9	8	0.96	0.99	0.99	0.95	0.99	0.99	0.94	0.96	0.95	0.88	0.95	0.95
10	5	0.95	0.99	1.01	0.93	0.99	1.01	0.94	0.96	0.94	0.87	0.95	0.93
10	6	0.96	0.99	1.00	0.94	0.99	1.00	0.94	0.96	0.94	0.88	0.95	0.93
10	7	0.96	0.99	0.99	0.94	0.99	0.99	0.94	0.96	0.94	0.88	0.95	0.93
10	8	0.96	0.99	0.99	0.95	0.99	0.99	0.94	0.96	0.94	0.88	0.95	0.93
10	9	0.96	0.99	0.99	0.95	0.99	0.99	0.94	0.96	0.95	0.88	0.95	0.95

Supplementary Table 7. Cellular S-values (Gy/Bq·s) for ^{124}I for both condensed-phase and isolated-atom assumptions of the *BrIccEmis* code.

R_N	R_C	$S_{\text{Condensed spectra}}$			$S_{\text{Isolated spectra}}$			Condensed spectra			Isolated spectra		
(μm)	(μm)	$S(N \leftarrow N)$	$S(N \leftarrow \text{Cy})$	$S(N \leftarrow \text{CS})$	$S(N \leftarrow N)$	$S(N \leftarrow \text{Cy})$	$S(N \leftarrow \text{CS})$	S_{AE}/S_{TOT}	S_{IC}/S_{TOT}	S_{BET}/S_{TOT}	S_{AE}/S_{TOT}	S_{IC}/S_{TOT}	S_{BET}/S_{TOT}
1	3	1.00E-01	9.39E-04	2.70E-04	9.70E-02	9.34E-04	2.69E-04	98.75%	0.02%	1.23%	98.71%	0.02%	1.27%
2	3	1.38E-02	6.99E-04	2.87E-04	1.34E-02	6.95E-04	2.86E-04	97.04%	0.04%	2.92%	96.95%	0.05%	3.00%
2	4	1.38E-02	3.82E-04	1.81E-04	1.34E-02	3.81E-04	1.80E-04	97.39%	0.04%	2.57%	97.32%	0.04%	2.64%
3	4	4.33E-03	3.43E-04	1.88E-04	4.21E-03	3.41E-04	1.87E-04	95.59%	0.07%	4.34%	95.48%	0.07%	4.46%
2	5	1.38E-02	2.62E-04	1.31E-04	1.34E-02	2.60E-04	1.30E-04	97.59%	0.04%	2.38%	97.51%	0.04%	2.45%
3	5	4.33E-03	2.26E-04	1.31E-04	4.21E-03	2.25E-04	1.31E-04	96.05%	0.06%	3.89%	95.95%	0.06%	3.99%
4	5	1.94E-03	2.11E-04	1.34E-04	1.89E-03	2.10E-04	1.34E-04	94.38%	0.08%	5.54%	94.25%	0.08%	5.66%
3	6	4.33E-03	1.70E-04	9.66E-05	4.21E-03	1.70E-04	9.62E-05	96.33%	0.05%	3.61%	96.23%	0.06%	3.71%
4	6	1.94E-03	1.52E-04	9.56E-05	1.89E-03	1.51E-04	9.52E-05	94.87%	0.08%	5.05%	94.75%	0.08%	5.18%
5	6	1.05E-03	1.41E-04	9.66E-05	1.03E-03	1.40E-04	9.62E-05	93.31%	0.10%	6.59%	93.20%	0.10%	6.70%
3	7	4.33E-03	1.35E-04	7.12E-05	4.21E-03	1.35E-04	7.09E-05	96.51%	0.05%	3.44%	96.42%	0.05%	3.53%
4	7	1.94E-03	1.20E-04	6.99E-05	1.89E-03	1.19E-04	6.96E-05	95.19%	0.07%	4.74%	95.08%	0.07%	4.85%
5	7	1.05E-03	1.06E-04	6.93E-05	1.03E-03	1.06E-04	6.90E-05	93.78%	0.09%	6.12%	93.68%	0.09%	6.23%
6	7	6.40E-04	9.88E-05	7.01E-05	6.26E-04	9.84E-05	6.99E-05	92.38%	0.11%	7.50%	92.24%	0.11%	7.64%
4	8	1.94E-03	9.68E-05	5.03E-05	1.89E-03	9.64E-05	5.01E-05	95.41%	0.07%	4.53%	95.29%	0.07%	4.64%
5	8	1.05E-03	8.56E-05	5.02E-05	1.03E-03	8.52E-05	5.00E-05	94.12%	0.09%	5.80%	94.01%	0.09%	5.90%
6	8	6.40E-04	7.62E-05	5.05E-05	6.26E-04	7.59E-05	5.04E-05	92.83%	0.11%	7.06%	92.69%	0.11%	7.20%
7	8	4.22E-04	7.13E-05	5.19E-05	4.13E-04	7.10E-05	5.17E-05	91.47%	0.13%	8.41%	91.32%	0.13%	8.56%
5	9	1.05E-03	7.01E-05	3.59E-05	1.03E-03	6.99E-05	3.58E-05	94.34%	0.08%	5.57%	94.24%	0.09%	5.67%
6	9	6.40E-04	6.22E-05	3.66E-05	6.26E-04	6.20E-05	3.65E-05	93.15%	0.10%	6.75%	93.02%	0.10%	6.88%
7	9	4.22E-04	5.60E-05	3.75E-05	4.13E-04	5.58E-05	3.74E-05	91.88%	0.12%	8.00%	91.73%	0.12%	8.15%
8	9	2.94E-04	5.28E-05	3.91E-05	2.87E-04	5.26E-05	3.90E-05	90.60%	0.14%	9.26%	90.42%	0.14%	9.44%
5	10	1.05E-03	5.77E-05	2.49E-05	1.03E-03	5.75E-05	2.49E-05	94.51%	0.08%	5.41%	94.41%	0.08%	5.51%
6	10	6.40E-04	5.14E-05	2.61E-05	6.26E-04	5.12E-05	2.61E-05	93.39%	0.10%	6.51%	93.26%	0.10%	6.65%
7	10	4.22E-04	4.60E-05	2.73E-05	4.13E-04	4.59E-05	2.72E-05	92.19%	0.12%	7.69%	92.05%	0.12%	7.84%
8	10	2.94E-04	4.20E-05	2.86E-05	2.87E-04	4.19E-05	2.85E-05	90.98%	0.13%	8.89%	90.80%	0.14%	9.07%
9	10	2.13E-04	4.02E-05	3.01E-05	2.08E-04	4.01E-05	3.00E-05	89.71%	0.15%	10.13%	89.52%	0.15%	10.32%
5	11	1.05E-03	4.75E-05	1.68E-05	1.03E-03	4.73E-05	1.68E-05	94.65%	0.08%	5.27%	94.55%	0.08%	5.37%
6	11	6.40E-04	4.26E-05	1.83E-05	6.26E-04	4.25E-05	1.83E-05	93.57%	0.10%	6.34%	93.44%	0.10%	6.47%
7	11	4.22E-04	3.84E-05	1.96E-05	4.13E-04	3.83E-05	1.96E-05	92.43%	0.11%	7.46%	92.28%	0.11%	7.60%
8	11	2.94E-04	3.49E-05	2.09E-05	2.87E-04	3.48E-05	2.09E-05	91.28%	0.13%	8.59%	91.10%	0.13%	8.77%
9	11	2.13E-04	3.23E-05	2.22E-05	2.08E-04	3.22E-05	2.21E-05	90.07%	0.15%	9.78%	89.87%	0.15%	9.98%
10	11	1.60E-04	3.13E-05	2.37E-05	1.57E-04	3.12E-05	2.36E-05	88.80%	0.17%	11.03%	88.63%	0.17%	11.20%
6	12	6.40E-04	3.54E-05	1.24E-05	6.26E-04	3.53E-05	1.24E-05	93.71%	0.09%	6.19%	93.58%	0.09%	6.32%
7	12	4.22E-04	3.21E-05	1.38E-05	4.13E-04	3.20E-05	1.38E-05	92.61%	0.11%	7.28%	92.47%	0.11%	7.42%
8	12	2.94E-04	2.93E-05	1.51E-05	2.87E-04	2.92E-05	1.51E-05	91.51%	0.13%	8.37%	91.33%	0.13%	8.54%
9	12	2.13E-04	2.70E-05	1.63E-05	2.08E-04	2.69E-05	1.63E-05	90.35%	0.14%	9.51%	90.16%	0.15%	9.70%
10	12	1.60E-04	2.53E-05	1.76E-05	1.57E-04	2.52E-05	1.75E-05	89.13%	0.16%	10.71%	88.95%	0.16%	10.88%
11	12	1.23E-04	2.48E-05	1.90E-05	1.21E-04	2.47E-05	1.89E-05	87.95%	0.18%	11.87%	87.79%	0.18%	12.03%

Supplementary Table 8. S-value (Gy/Bq·s) contribution of MIRD-RADTABS X-ray spectra (Eckerman and Endo, 2008a) to the total dose from radiative (X-rays) and non-radiative (AE, CK, IC and β^+) processes deposited in the nucleus per cumulated activity from contributions from the nucleus, cytoplasm and cell surface i.e. $S_{TOT} = S(N \leftarrow N) + S(N \leftarrow Cy) + S(N \leftarrow Cs)$.

R_C	R_N	^{67}Ga	^{80m}Br	^{89}Zr	^{90}Nb	^{99m}Tc	^{111}In	^{117m}Sn	^{119}Sb	^{123}I	^{124}I	^{125}I	^{195}Pt	^{201}Tl
(μm)	(μm)	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$
3	1	2.60%	3.72%	2.58%	2.73%	1.82%	3.92%	4.06%	3.88%	4.75%	5.19%	3.53%	2.36%	3.57%
3	2	1.91%	2.05%	1.86%	1.92%	1.70%	3.42%	3.34%	2.79%	4.24%	4.56%	3.14%	1.57%	2.62%
4	2	2.04%	2.31%	2.03%	2.08%	1.72%	3.49%	3.46%	2.94%	4.33%	4.67%	3.21%	1.70%	2.81%
4	3	1.81%	1.55%	1.63%	1.60%	1.62%	3.07%	2.90%	2.10%	3.94%	4.19%	2.94%	1.26%	2.33%
5	2	2.07%	2.42%	2.12%	2.18%	1.74%	3.54%	3.53%	3.03%	4.37%	4.71%	3.23%	1.76%	2.88%
5	3	1.92%	1.73%	1.78%	1.75%	1.66%	3.17%	3.03%	2.25%	4.04%	4.31%	3.01%	1.38%	2.51%
5	4	1.78%	1.25%	1.56%	1.47%	1.56%	2.79%	2.56%	1.68%	3.66%	3.86%	2.76%	1.10%	2.18%
6	3	1.95%	1.82%	1.86%	1.84%	1.68%	3.24%	3.11%	2.38%	4.09%	4.38%	3.05%	1.45%	2.59%
6	4	1.89%	1.38%	1.70%	1.60%	1.60%	2.91%	2.69%	1.83%	3.77%	3.99%	2.83%	1.20%	2.35%
6	5	1.78%	1.05%	1.55%	1.40%	1.53%	2.62%	2.29%	1.46%	3.40%	3.60%	2.59%	0.99%	2.10%
7	3	1.96%	1.88%	1.90%	1.89%	1.69%	3.30%	3.17%	2.49%	4.13%	4.42%	3.07%	1.50%	2.63%
7	4	1.91%	1.45%	1.77%	1.68%	1.62%	3.00%	2.79%	1.96%	3.84%	4.07%	2.88%	1.26%	2.43%
7	5	1.87%	1.15%	1.68%	1.52%	1.57%	2.73%	2.41%	1.59%	3.52%	3.74%	2.67%	1.08%	2.26%
7	6	1.77%	0.91%	1.56%	1.35%	1.50%	2.51%	2.10%	1.33%	3.24%	3.39%	2.47%	0.94%	2.05%
8	4	1.91%	1.51%	1.80%	1.72%	1.63%	3.07%	2.86%	2.07%	3.90%	4.14%	2.91%	1.31%	2.47%
8	5	1.89%	1.22%	1.75%	1.59%	1.59%	2.83%	2.51%	1.71%	3.60%	3.83%	2.72%	1.14%	2.33%
8	6	1.86%	0.99%	1.68%	1.46%	1.54%	2.63%	2.22%	1.45%	3.36%	3.53%	2.55%	1.02%	2.20%
8	7	1.78%	0.82%	1.58%	1.32%	1.48%	2.46%	1.97%	1.25%	3.12%	3.24%	2.36%	0.91%	2.01%
9	5	1.90%	1.27%	1.77%	1.63%	1.61%	2.90%	2.58%	1.81%	3.67%	3.91%	2.76%	1.19%	2.38%
9	6	1.88%	1.05%	1.75%	1.53%	1.56%	2.72%	2.31%	1.56%	3.44%	3.63%	2.60%	1.08%	2.27%
9	7	1.87%	0.88%	1.70%	1.42%	1.52%	2.57%	2.08%	1.36%	3.23%	3.38%	2.44%	0.98%	2.16%
9	8	1.78%	0.75%	1.60%	1.30%	1.46%	2.42%	1.86%	1.20%	3.03%	3.13%	2.28%	0.88%	1.98%
10	5	1.90%	1.31%	1.78%	1.65%	1.62%	2.96%	2.64%	1.89%	3.72%	3.98%	2.79%	1.22%	2.40%
10	6	1.89%	1.09%	1.77%	1.56%	1.58%	2.79%	2.38%	1.65%	3.52%	3.71%	2.65%	1.12%	2.31%
10	7	1.88%	0.93%	1.76%	1.48%	1.54%	2.66%	2.16%	1.46%	3.32%	3.48%	2.50%	1.03%	2.22%
10	8	1.86%	0.80%	1.72%	1.39%	1.49%	2.53%	1.96%	1.31%	3.14%	3.26%	2.36%	0.95%	2.12%
10	9	1.79%	0.69%	1.63%	1.28%	1.43%	2.40%	1.77%	1.17%	2.97%	3.07%	2.23%	0.86%	1.96%

Supplementary Table 9. S-value (Gy/Bq·s) contribution of *BrIccEmis* isolated assumption X-ray spectra to the total dose from radiative (X-rays) and non-radiative (AE, CK, IC and β^+) processes deposited in the nucleus per cumulated activity from contributions from the nucleus, cytoplasm and cell surface i.e. $S_{TOT} = S(N \leftarrow N) + S(N \leftarrow Cy) + S(N \leftarrow Cs)$.

R_C	R_N	^{67}Ga	^{80m}Br	^{89}Zr	^{90}Nb	^{99m}Tc	^{111}In	^{117m}Sn	^{119}Sb	^{123}I	^{124}I	^{125}I	^{195}Pt	^{201}Tl
(μm)	(μm)	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$	$S_{X\text{-ray}}/S_{TOT}$
3	1	0.18%	0.16%	0.17%	0.63%	0.03%	0.11%	0.22%	0.20%	0.31%	0.31%	0.24%	0.07%	0.21%
3	2	0.30%	0.20%	0.28%	0.63%	0.05%	0.22%	0.28%	0.22%	0.41%	0.40%	0.31%	0.12%	0.31%
4	2	0.28%	0.20%	0.27%	0.64%	0.05%	0.20%	0.27%	0.22%	0.39%	0.38%	0.29%	0.11%	0.30%
4	3	0.40%	0.22%	0.36%	0.64%	0.07%	0.30%	0.32%	0.22%	0.47%	0.46%	0.36%	0.14%	0.38%
5	2	0.27%	0.19%	0.26%	0.65%	0.04%	0.19%	0.26%	0.21%	0.38%	0.37%	0.29%	0.11%	0.28%
5	3	0.37%	0.22%	0.35%	0.65%	0.07%	0.28%	0.31%	0.21%	0.45%	0.44%	0.34%	0.14%	0.36%
5	4	0.48%	0.23%	0.44%	0.66%	0.10%	0.37%	0.35%	0.21%	0.52%	0.51%	0.40%	0.16%	0.44%
6	3	0.35%	0.21%	0.34%	0.66%	0.06%	0.26%	0.30%	0.21%	0.43%	0.43%	0.33%	0.13%	0.35%
6	4	0.45%	0.22%	0.43%	0.67%	0.09%	0.34%	0.34%	0.21%	0.49%	0.49%	0.38%	0.16%	0.42%
6	5	0.54%	0.23%	0.52%	0.69%	0.12%	0.43%	0.38%	0.22%	0.56%	0.55%	0.43%	0.18%	0.49%
7	3	0.34%	0.21%	0.33%	0.66%	0.06%	0.25%	0.30%	0.22%	0.42%	0.42%	0.32%	0.13%	0.34%
7	4	0.42%	0.22%	0.42%	0.68%	0.08%	0.33%	0.33%	0.22%	0.48%	0.47%	0.37%	0.15%	0.41%
7	5	0.52%	0.22%	0.51%	0.70%	0.11%	0.40%	0.36%	0.22%	0.54%	0.53%	0.41%	0.17%	0.47%
7	6	0.61%	0.22%	0.59%	0.71%	0.14%	0.49%	0.40%	0.23%	0.60%	0.59%	0.46%	0.19%	0.53%
8	4	0.41%	0.21%	0.41%	0.68%	0.08%	0.32%	0.33%	0.22%	0.47%	0.46%	0.36%	0.15%	0.39%
8	5	0.49%	0.22%	0.50%	0.70%	0.10%	0.39%	0.36%	0.22%	0.52%	0.51%	0.40%	0.17%	0.46%
8	6	0.58%	0.22%	0.59%	0.72%	0.13%	0.47%	0.39%	0.23%	0.58%	0.57%	0.44%	0.19%	0.52%
8	7	0.66%	0.22%	0.65%	0.74%	0.15%	0.54%	0.41%	0.24%	0.64%	0.63%	0.49%	0.21%	0.57%
9	5	0.47%	0.21%	0.48%	0.70%	0.10%	0.38%	0.35%	0.23%	0.52%	0.51%	0.39%	0.17%	0.44%
9	6	0.55%	0.22%	0.58%	0.73%	0.12%	0.45%	0.38%	0.23%	0.57%	0.55%	0.43%	0.19%	0.50%
9	7	0.64%	0.22%	0.66%	0.75%	0.14%	0.53%	0.41%	0.24%	0.62%	0.61%	0.47%	0.21%	0.56%
9	8	0.71%	0.22%	0.71%	0.76%	0.17%	0.60%	0.43%	0.25%	0.68%	0.66%	0.52%	0.22%	0.61%
10	5	0.46%	0.21%	0.47%	0.70%	0.09%	0.37%	0.35%	0.23%	0.51%	0.50%	0.39%	0.17%	0.43%
10	6	0.53%	0.21%	0.56%	0.73%	0.11%	0.44%	0.38%	0.24%	0.56%	0.55%	0.42%	0.18%	0.49%
10	7	0.61%	0.21%	0.65%	0.75%	0.14%	0.51%	0.40%	0.25%	0.61%	0.59%	0.46%	0.20%	0.54%
10	8	0.69%	0.21%	0.72%	0.77%	0.16%	0.58%	0.42%	0.26%	0.66%	0.65%	0.50%	0.22%	0.60%
10	9	0.75%	0.21%	0.77%	0.77%	0.19%	0.66%	0.45%	0.27%	0.72%	0.70%	0.54%	0.23%	0.64%

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