Inelastic neutron scattering
Part 1: Triple Axis Spectrometer

NEUTRONS FOR SCIENCE®

Watching the ballet of atoms and magnetic moments

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Milan Klicpera

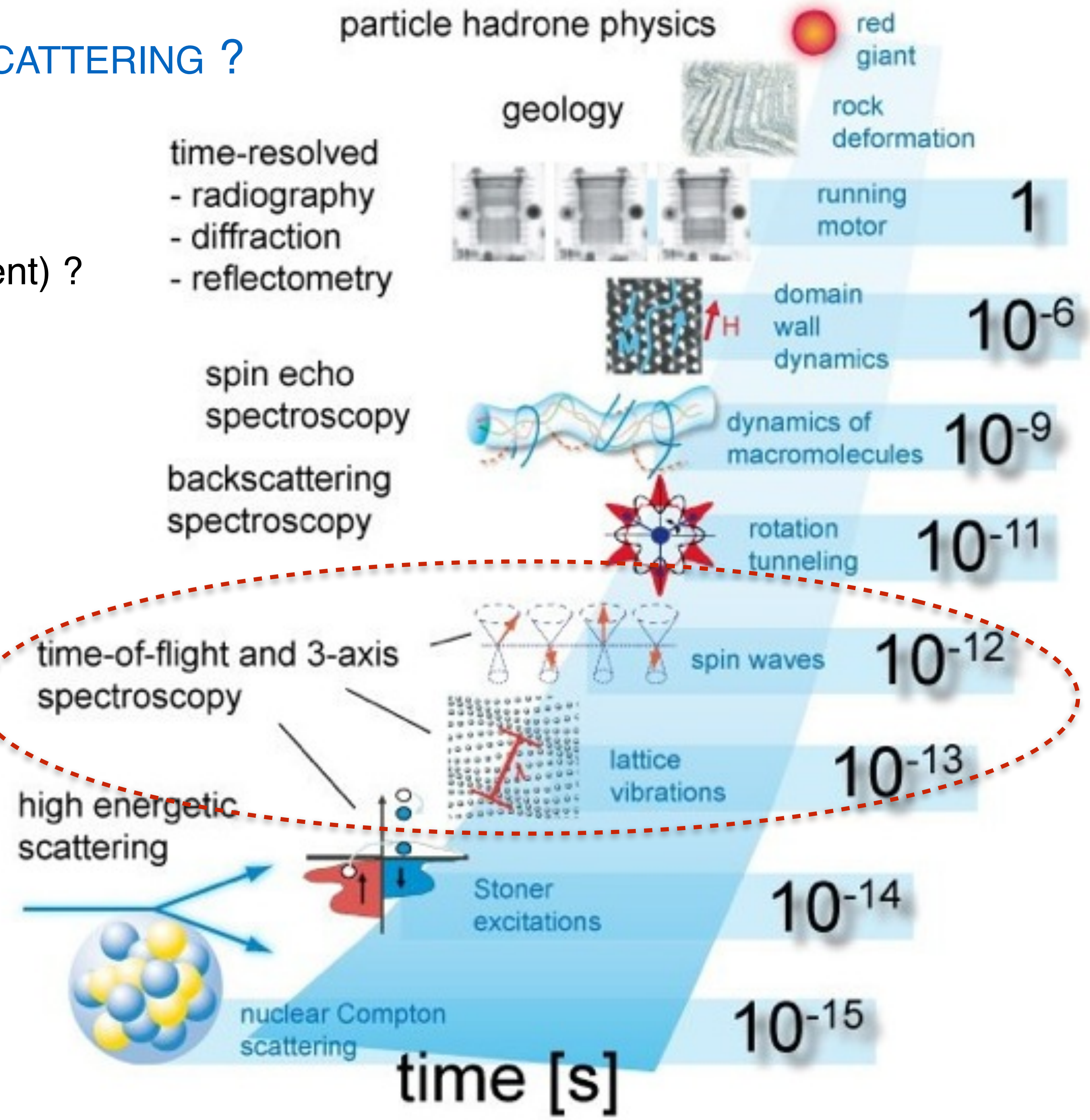
May 12th 2015

Introduction

Neutrons show where atoms are (elastic scattering) and what they do (inelastic scattering)

WHAT DO WE MEASURE IN INELASTIC NEUTRON SCATTERING ?

How Atoms or Magnetic moments are correlated
In Space and Time
With each other (Coherent) or with themselves (Incoherent) ?



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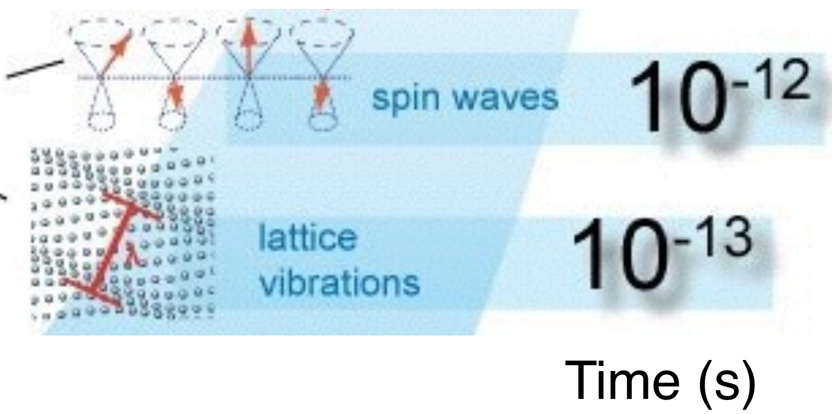
Energy range of the TAS and the TOF spectrometer
=
Energy range of some collective excitations
=
1 to 100 meV (25meV = 300K)

time-of-flight and 3-axis spectroscopy



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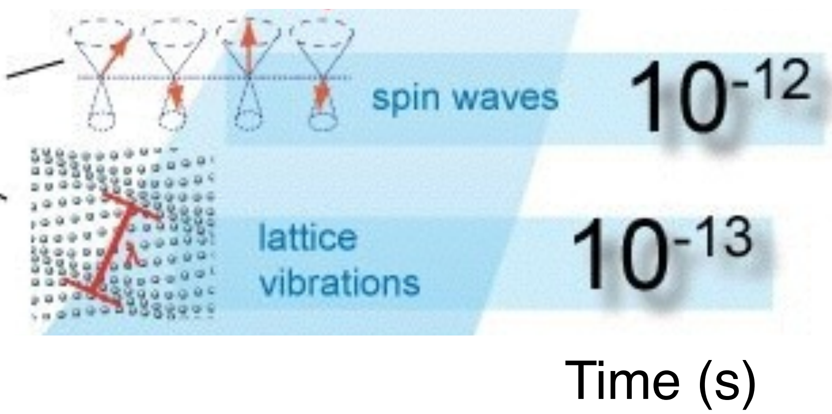
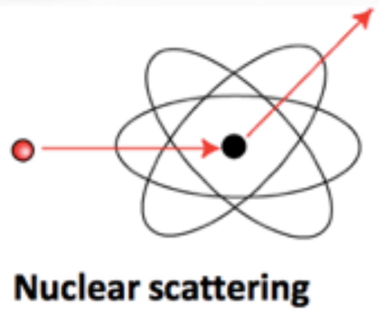
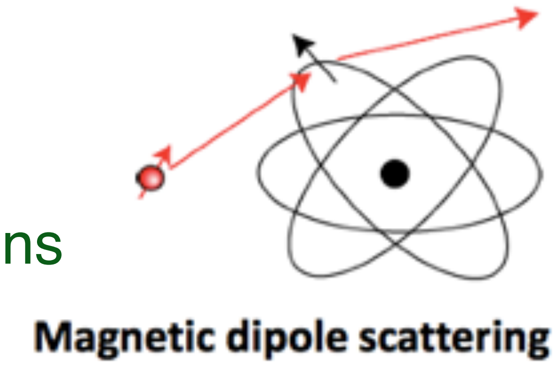




Watching the ballet of atoms / Lattice vibrations / Phonons

Watching the ballet of magnetic moments / Spin waves / Magnons



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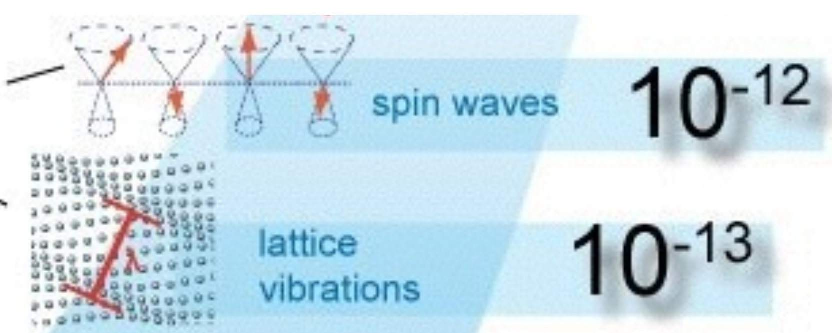


Time (s)

Nuclear scattering amplitude

Magnetic scattering amplitude

Interferences between scattering amplitude → Polarized TAS

Outline

I. Instrumentation

- Inelastic neutron scattering
- Triple Axis Spectrometer
- Polarized TAS

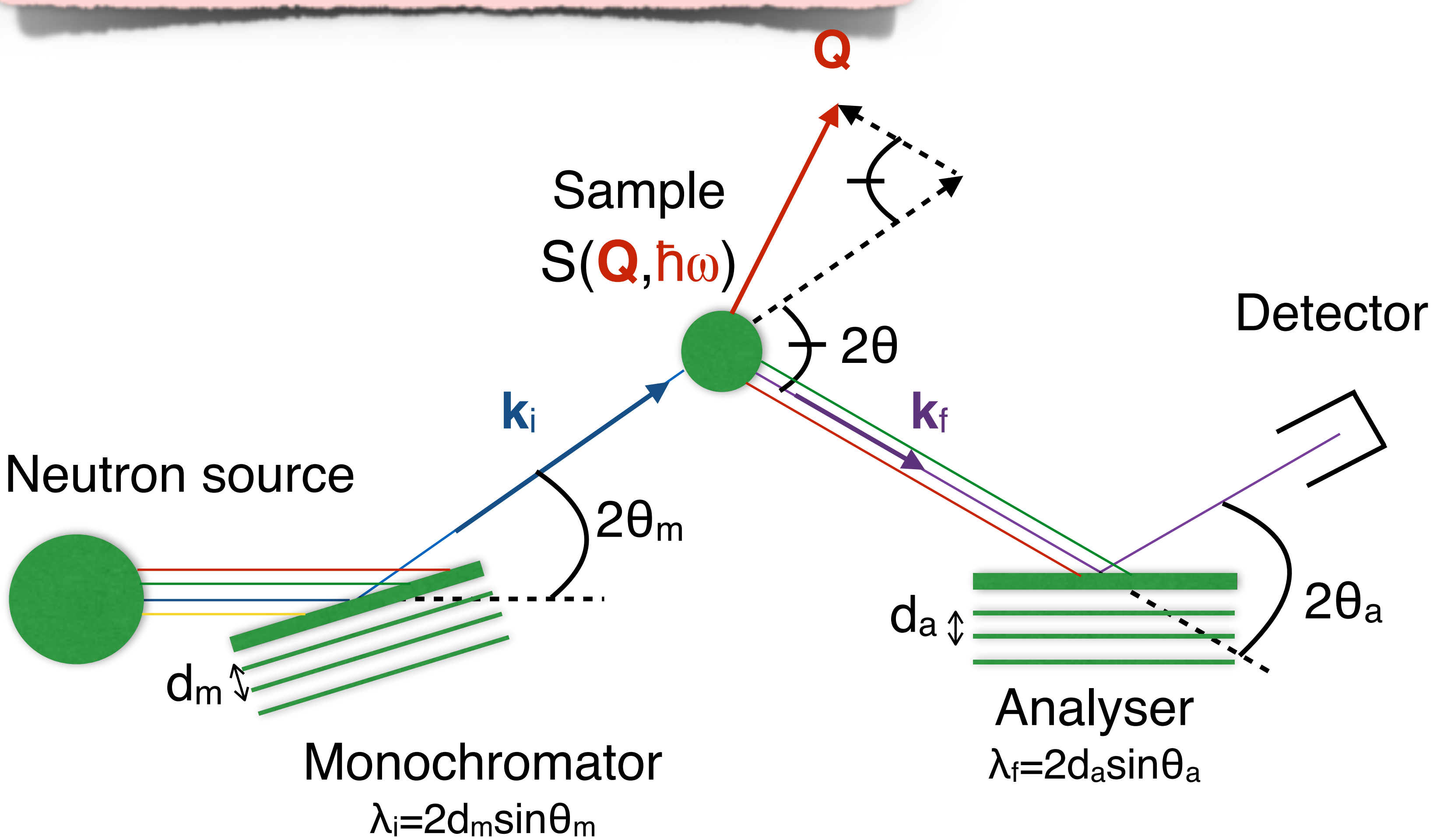
II. Phonons and magnons

- Physical description
- How do we measure them with neutrons ?
- Examples

Inelastic neutron scattering

Triple Axis Spectrometer

$$\left(\frac{d^2\sigma}{d\Omega dE_f}\right)_{k_i \to k_f} = \mathbf{k}_f/\mathbf{k}_i\, S(\mathbf{Q},\hbar\omega)$$



The conservation laws:

- Momentum conservation:

$\mathbf{k}_i - \mathbf{k}_f = \mathbf{Q}$

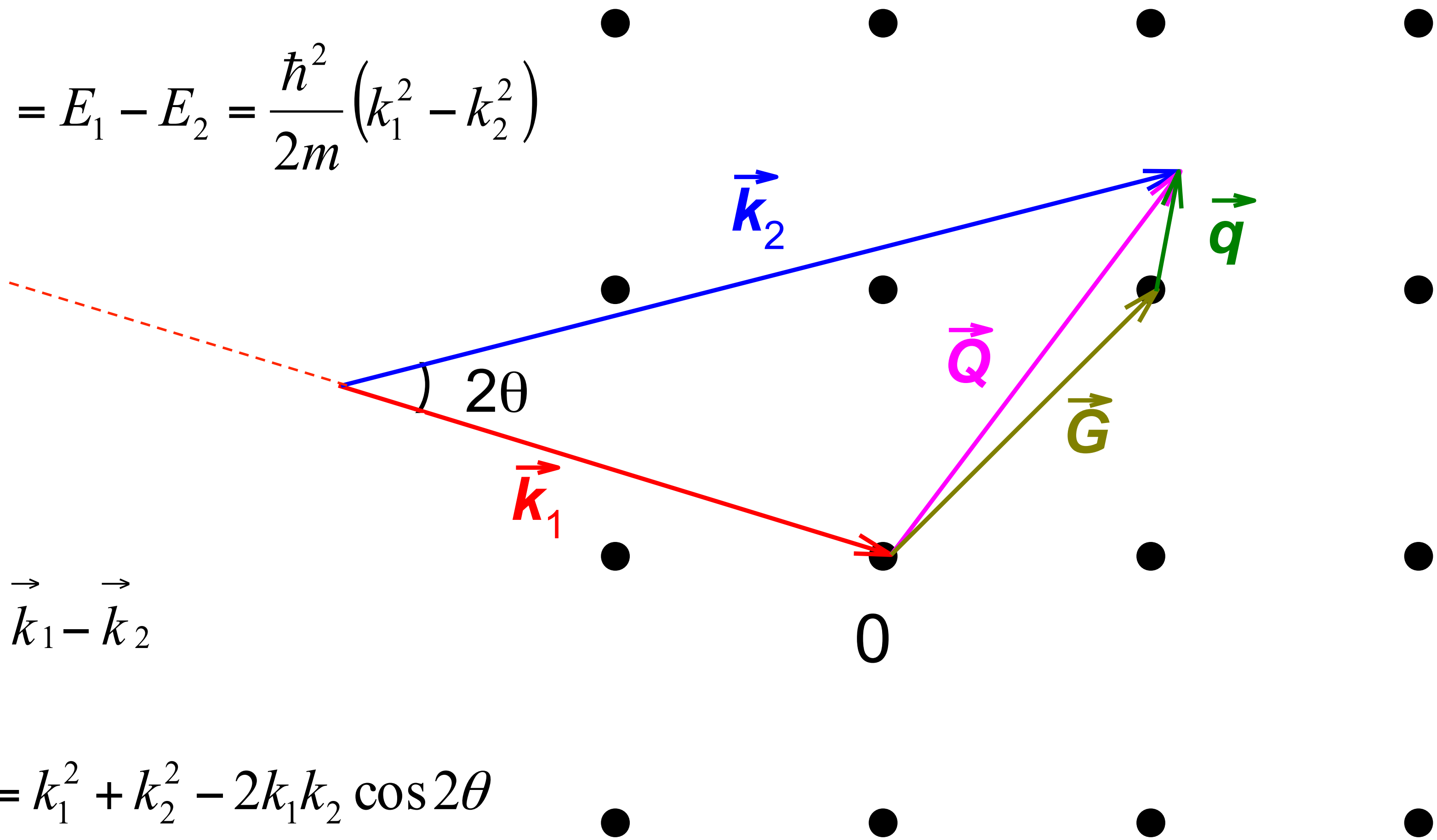
$Q^2 = k_i^2 + k_f^2 - 2\mathbf{k}_i\mathbf{k}_f\cos(2\theta)$

- Energy conservation:

$$\hbar\omega = E_i - E_f = \frac{\hbar^2 k_i^2}{2m_n} - \frac{\hbar^2 k_f^2}{2m_n}$$

Scattering triangle

$$\hbar\omega = E_1 - E_2 = \frac{\hbar^2}{2m}\left(k_1^2 - k_2^2\right)$$



$$\vec{Q} = \vec{k}_1 - \vec{k}_2$$

$$Q^2 = k_1^2 + k_2^2 - 2k_1 k_2 \cos 2\theta$$

$$\cos 2\theta = \frac{k_1^2 + k_2^2 - Q^2}{2k_1 k_2}$$

→ Scattering function: $S(\vec{Q}, \hbar\omega)$

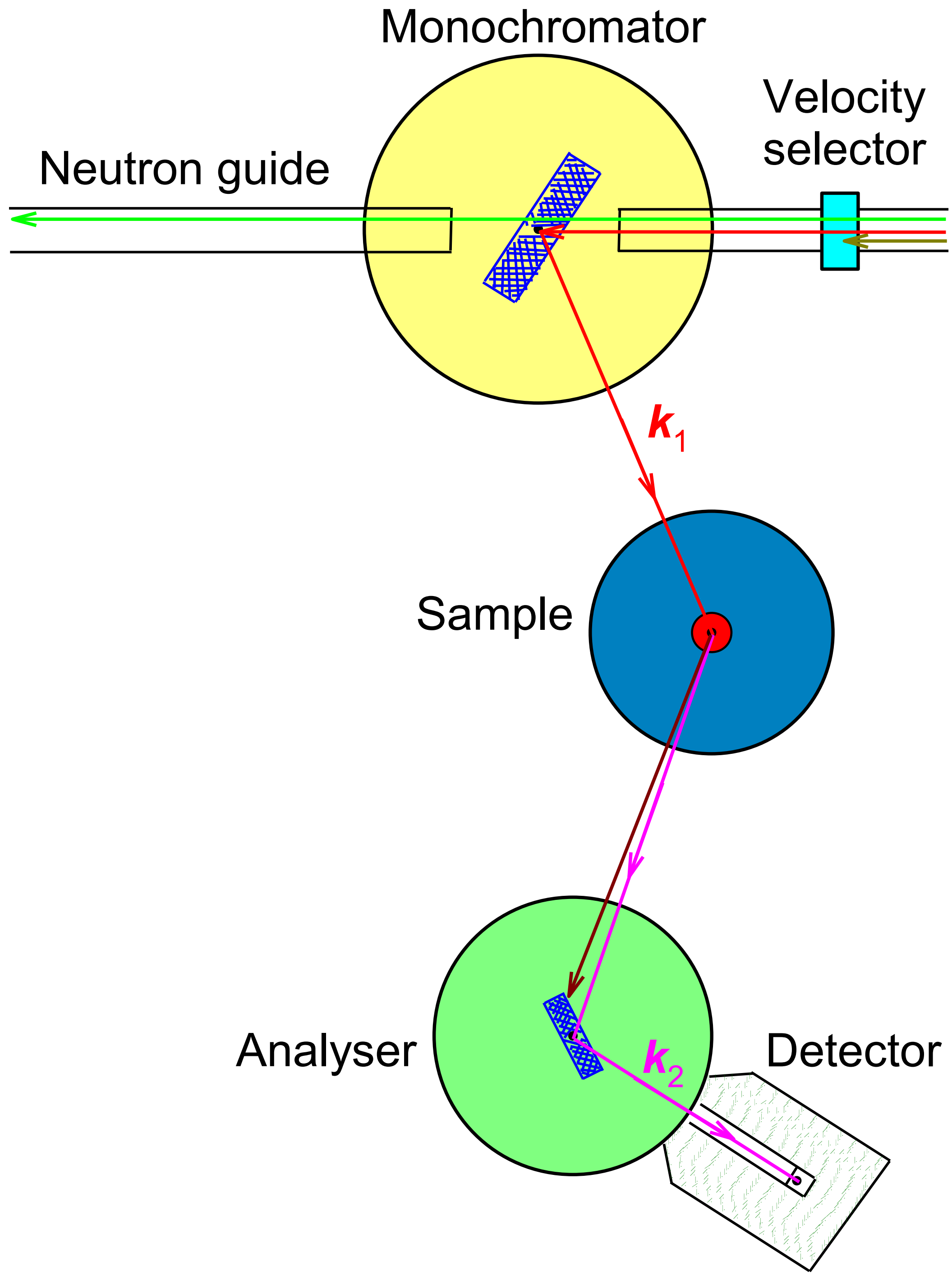
Theory

$$S(\vec{Q},\omega) = \frac{1}{2\pi\hbar}\sum_{\lambda_i} p(\lambda_i) \sum_{j,j'=1}^{N} b_j b_{j'}^{*} \int_{-\infty}^{\infty} \mathrm{d}t \langle\lambda_i|\mathrm{e}^{-\mathrm{i}\vec{Q}\cdot\vec{R}_{j'}^{0}}\mathrm{e}^{\mathrm{i}\vec{Q}\cdot\vec{R}_j(t)}|\lambda_i\rangle \mathrm{e}^{-\mathrm{i}\omega t}$$

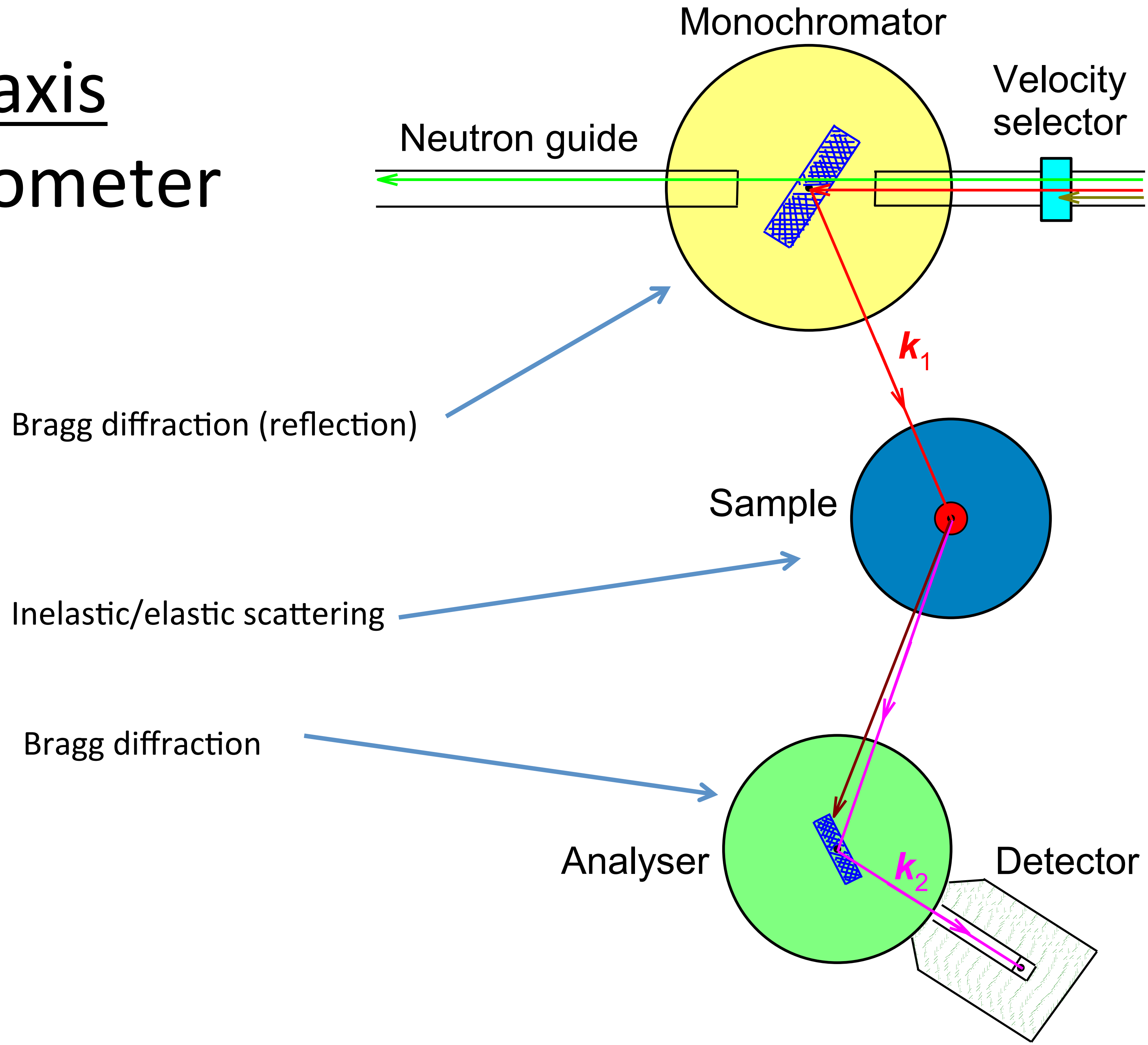
$S(\vec{Q},\hbar\omega)$ - defined everywhere in reciprocal space (= $(\vec{Q},\hbar\omega)$ space)

- if Born approximation is valid

Triple axis spectrometer



Triple axis spectrometer



Triple-axis spectrometer - animation

http://www.ill.eu/about/movies/animations/instrument-animations/three-axis-neutron-spectrometer/

Experiment

$S(\vec{Q},\hbar\omega)$ - defined everywhere in reciprocal space (= $(\vec{Q},\hbar\omega)$ space)

- if Born approximation is valid

Typically - experimental configuration with $\vec{k}_1$ (or $\vec{k}_2$) fixed

$$\hbar\omega = E_1 - E_2 = \frac{\hbar^2}{2m}\left(k_1^2 - k_2^2\right)$$

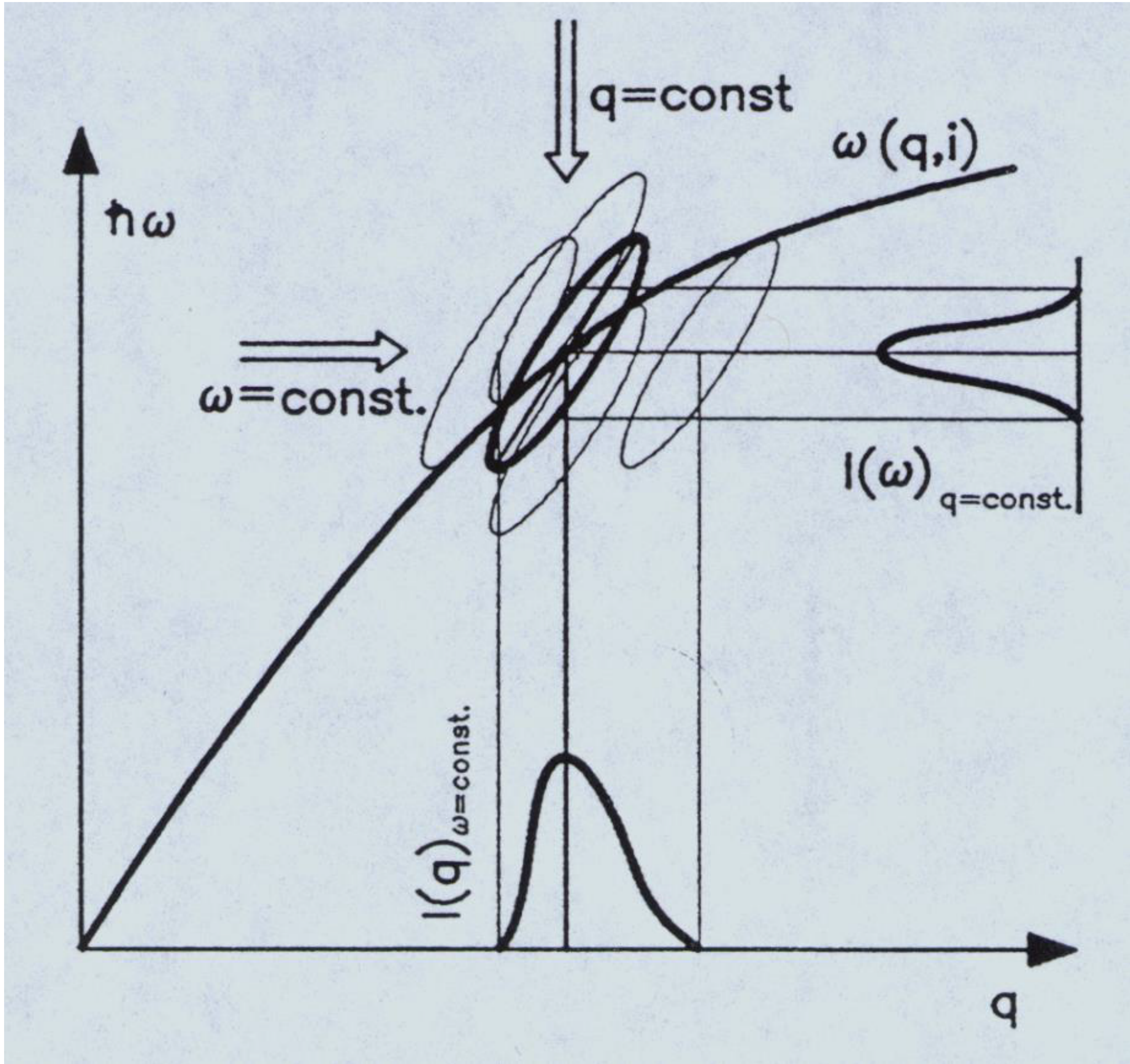
$$\cos 2\theta = \frac{k_1^2 + k_2^2 - Q^2}{2k_1 k_2}$$

$\Downarrow$

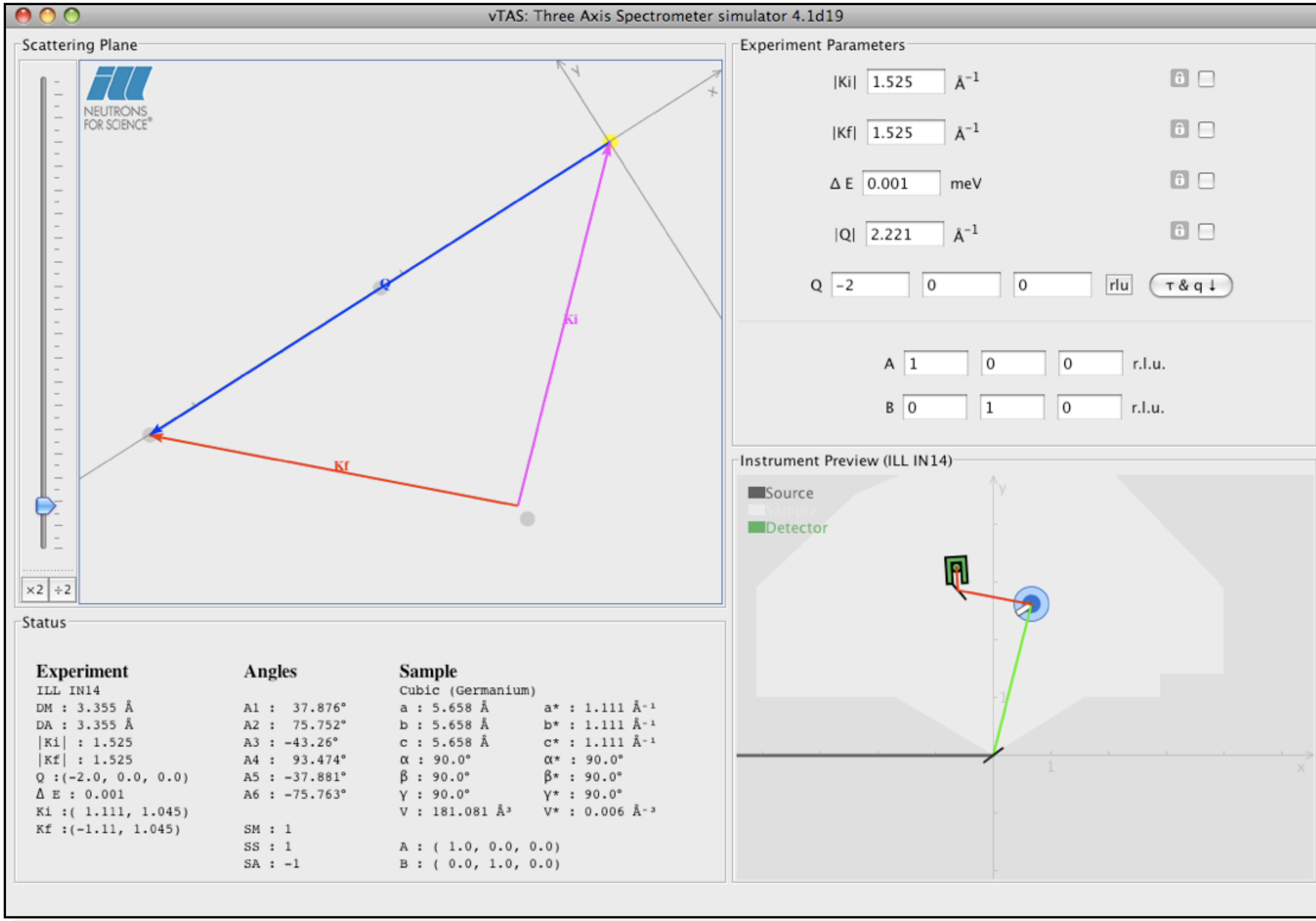
Restricted places in reciprocal space

Example: elastic scattering → $k_1 = k_2$ → if 2θ = π (backscattering) → $Q = 2\,k_1$ = maximal

Dispersion



TAS simulator

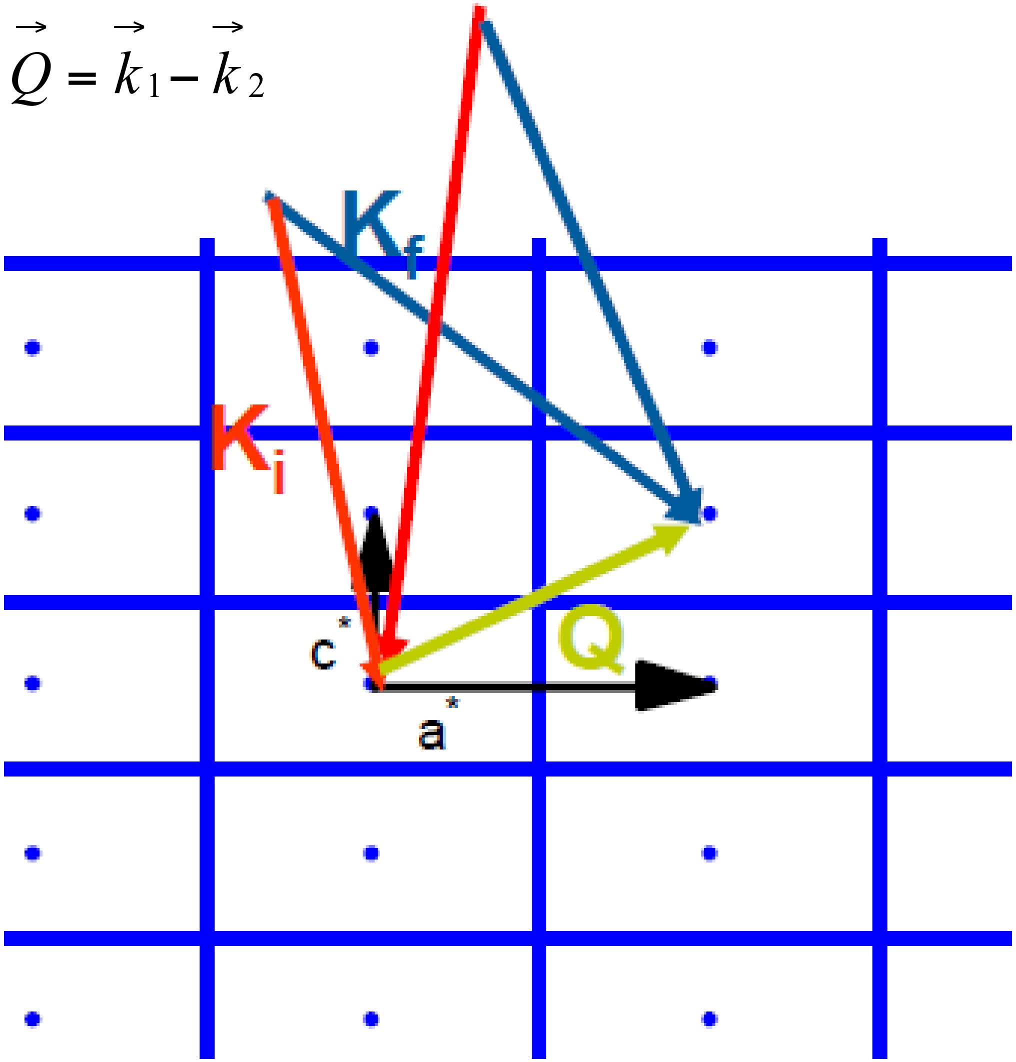


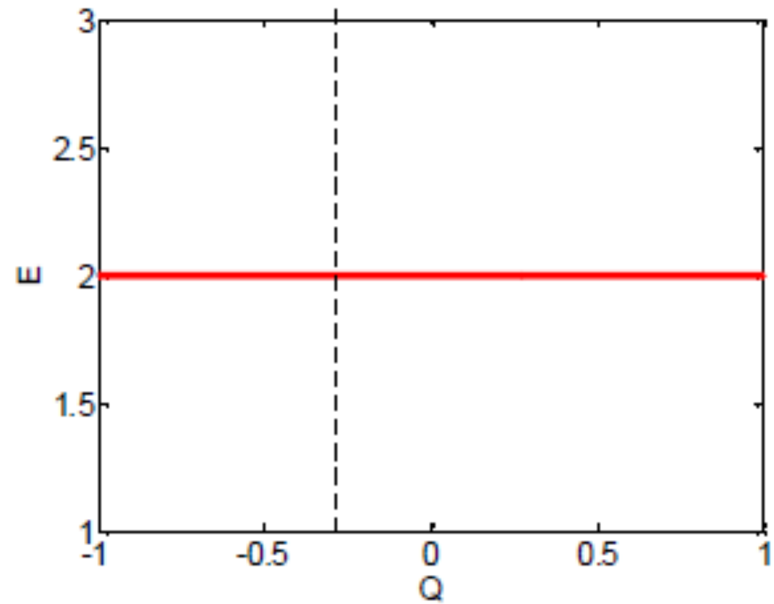
http://www.ill.eu/instruments-support/computing-for-science/cs-software/all-software/vtas/

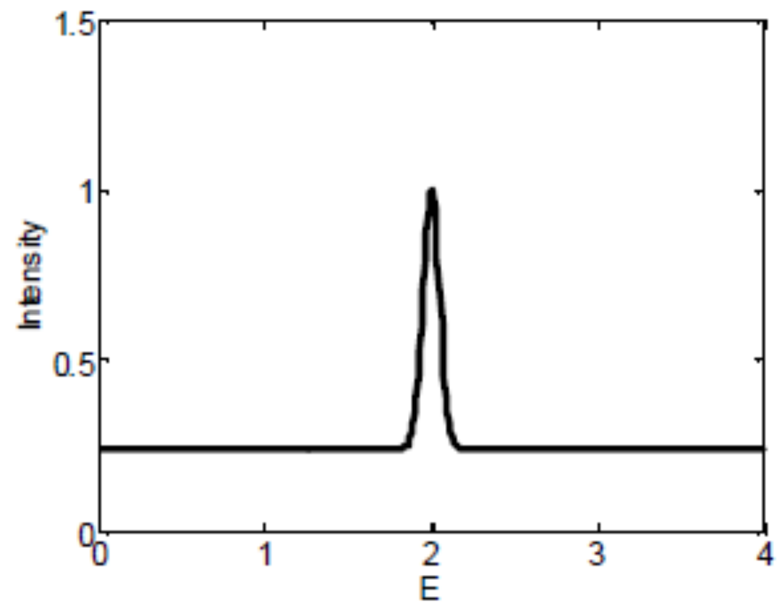
Conventional measurements, **constant-*Q***

$$\hbar\omega = E_1 - E_2 = \frac{\hbar^2}{2m}\left(k_1^2 - k_2^2\right)$$

$$\vec{Q} = \vec{k}_1 - \vec{k}_2$$



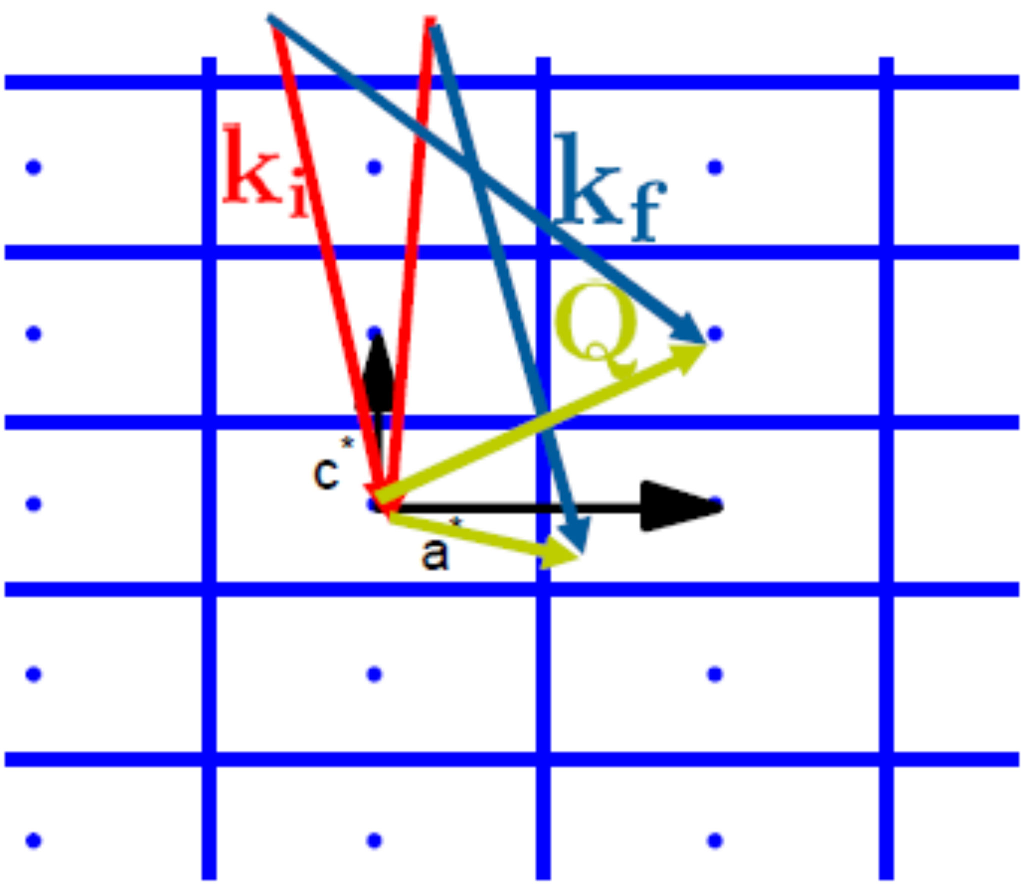


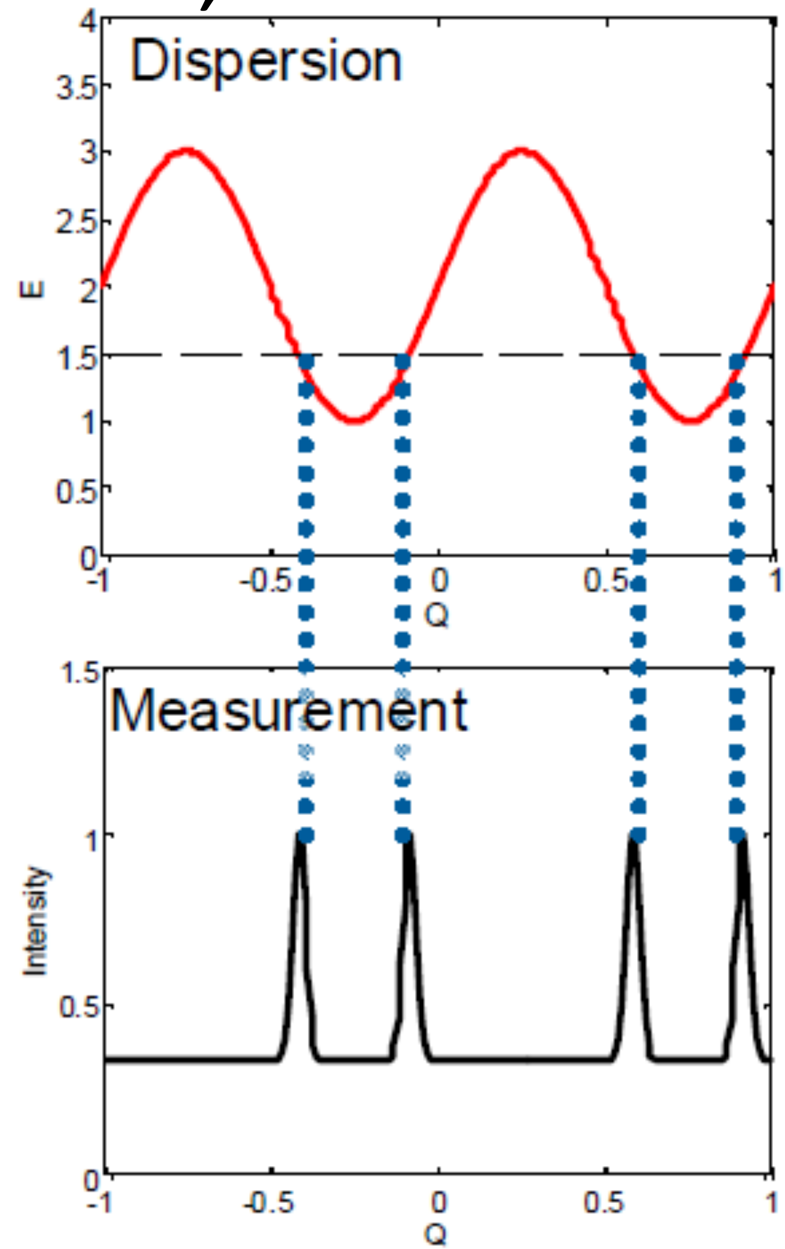


Conventional measurements, **constant-*E***

$$\hbar\omega = E_1 - E_2 = \frac{\hbar^2}{2m}\left(k_1^2 - k_2^2\right)$$

$$\vec{Q} = \vec{k}_1 - \vec{k}_2$$





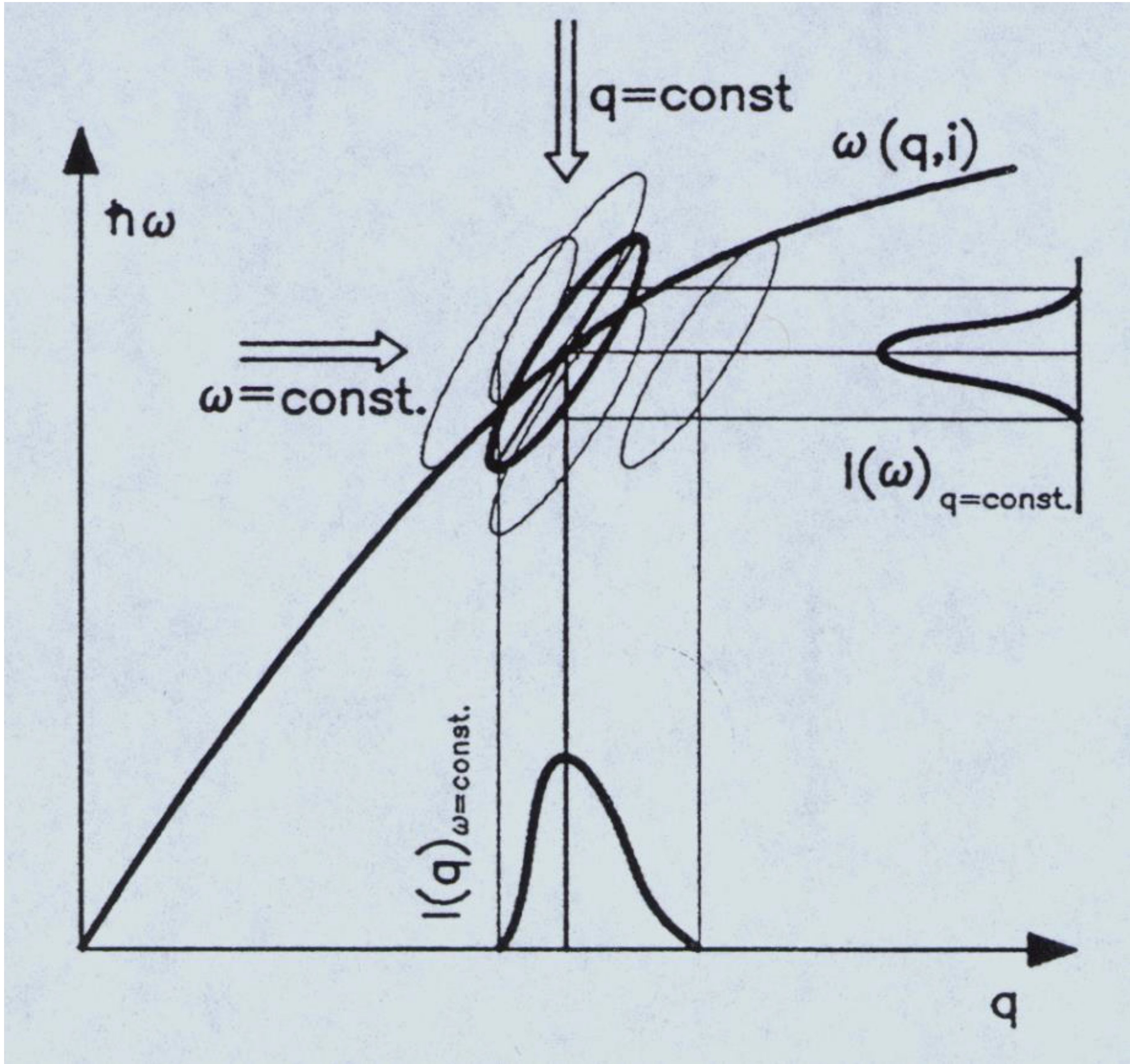
Resolution

Q and ***ω*** *defined only to a certain level of precision*

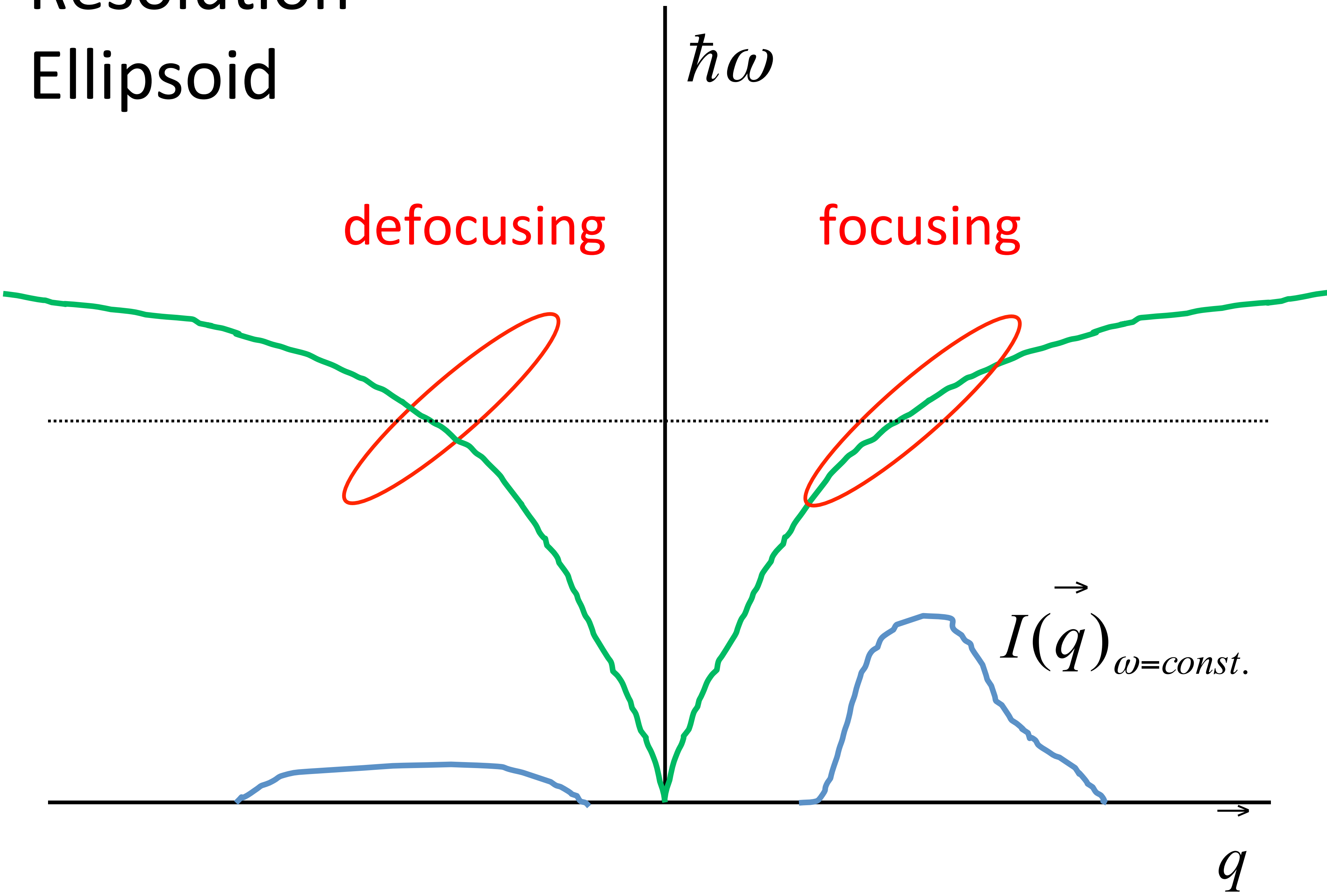
Reducing these uncertainties leads to a **better resolution**
BUT
it also leads to **lower counts**

→ resolution volumes, with distinct orientations
in $(\vec{Q}, \hbar\omega)$ space → ***resolution ellipsoid***

Resolution Ellipsoid



Resolution Ellipsoid



Convolution of resolution function with scattering function $S(\vec{Q},\hbar\omega)$

The measured intensity is the convolution:

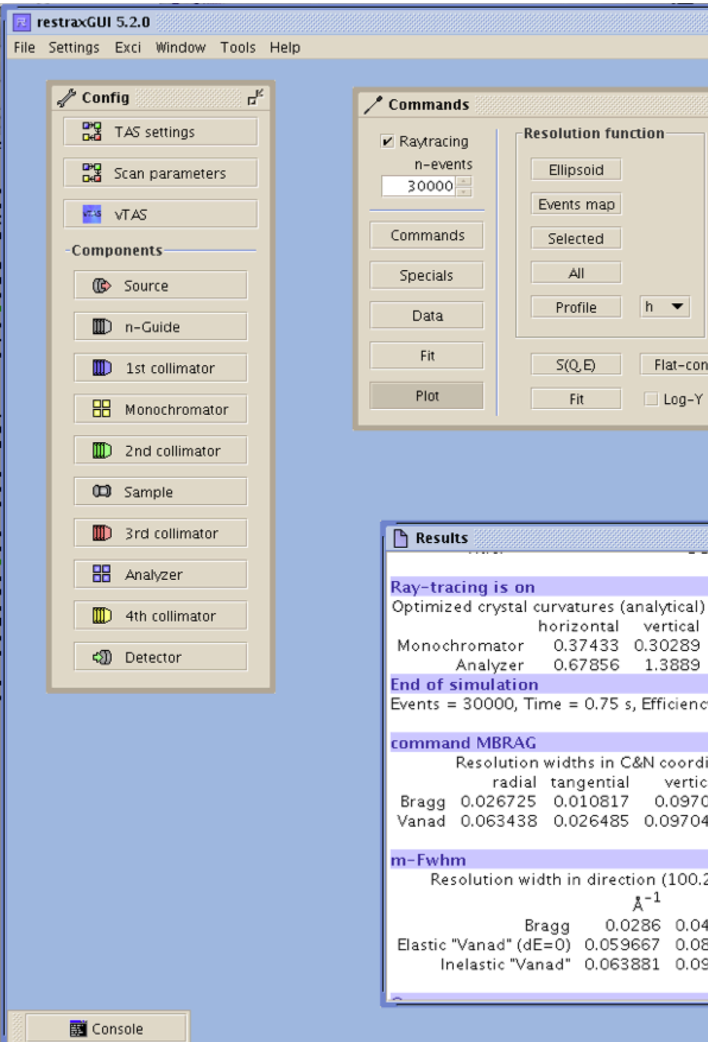
$$I_{measured} = \int S(\vec{Q},\omega) R(\vec{Q}-\vec{Q}_0, \omega-\omega_0) d\vec{Q}\, d\omega$$

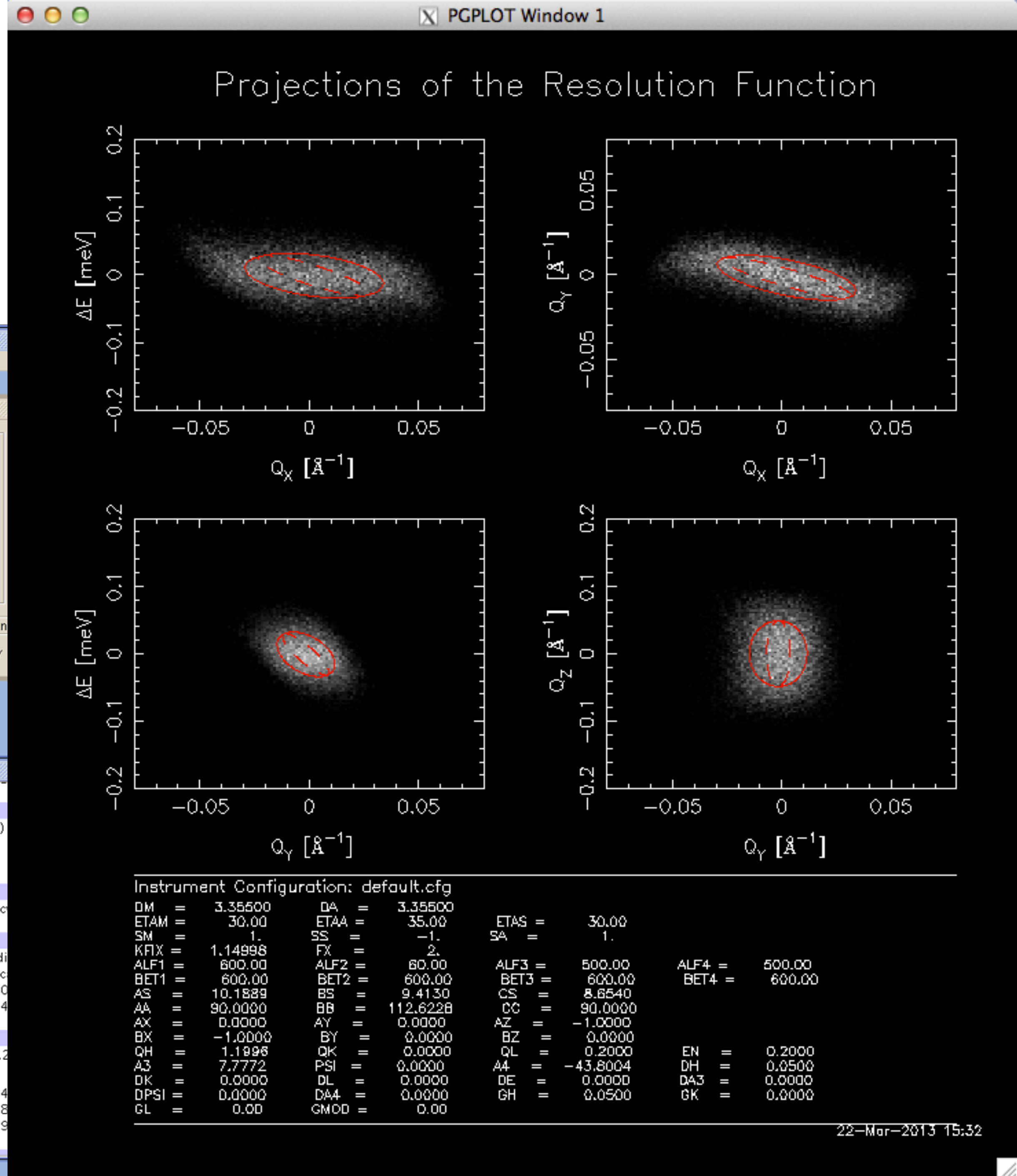
peak at $(\vec{Q}_0, \omega_0)$ instrument position
I decreases with $(\Delta Q_0, \Delta\omega_0)$
purely instrumental property

Restrax

http://neutron.ujf.cas.cz/restrax

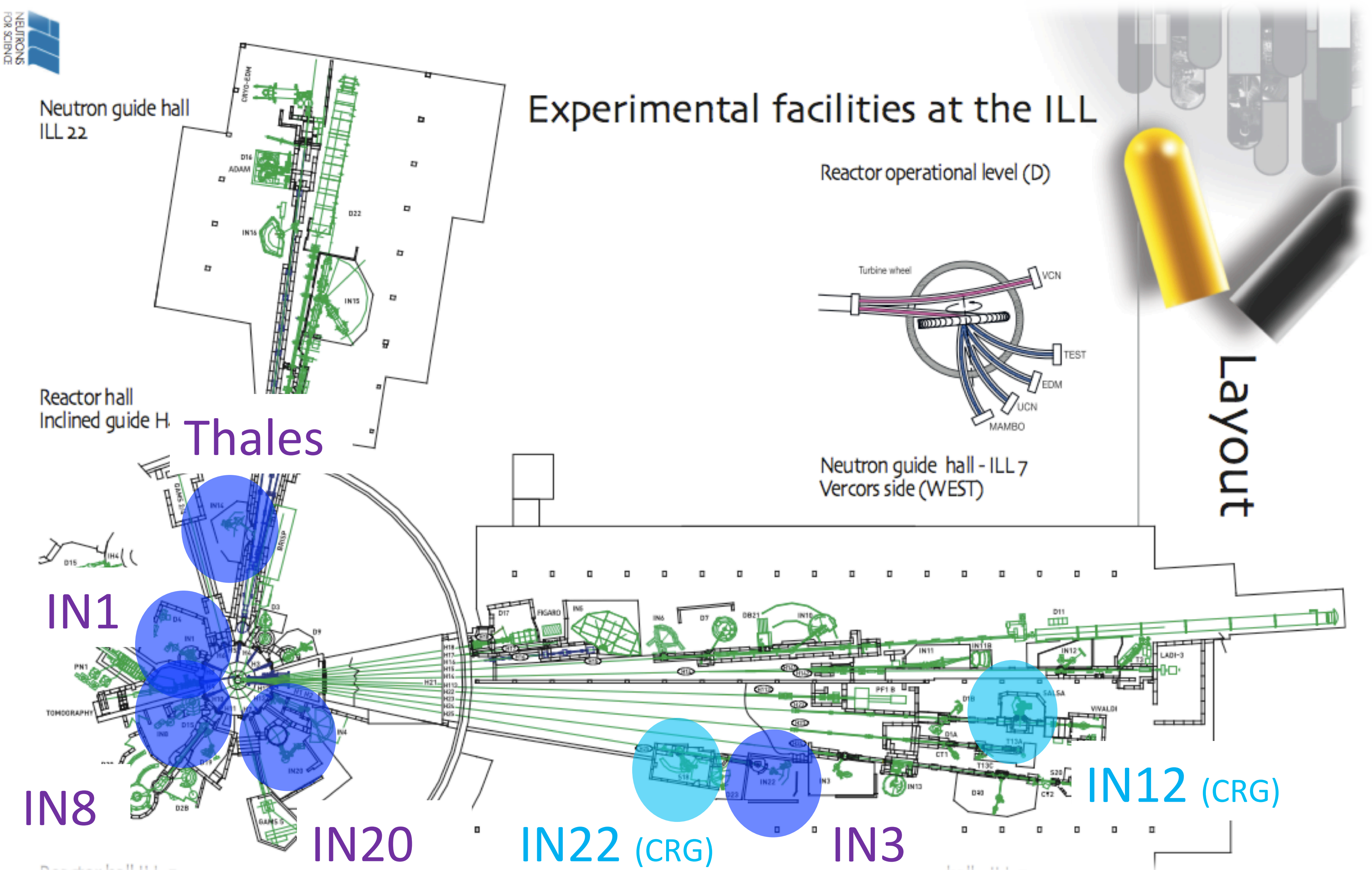
- By J. Saroun and J. Kulda





TAS - ILL

Cold – ThALES, IN12 (0.05 – 10 meV), IN22 (0.5 – 25 meV)
Thermal – IN20, IN8 (2 – 100 meV)
Hot – IN1 (few 100 meV)



Multiplex-detectors in ILL

FlatCone

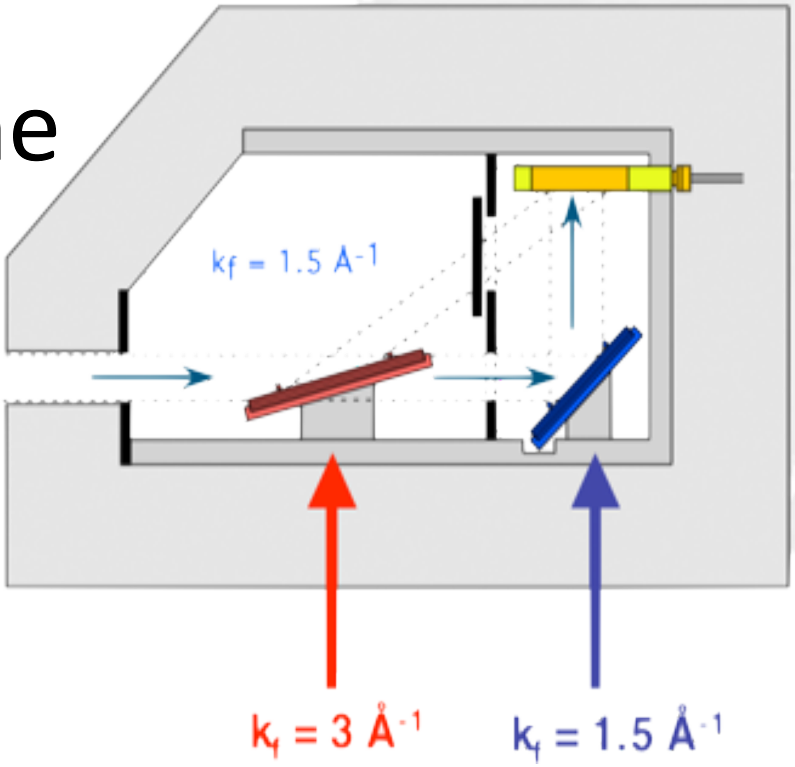
Lagrange

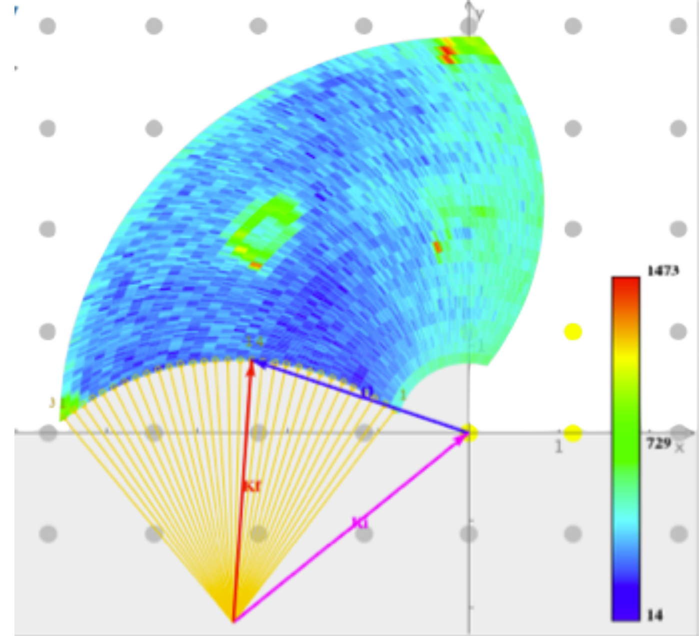
IMPS

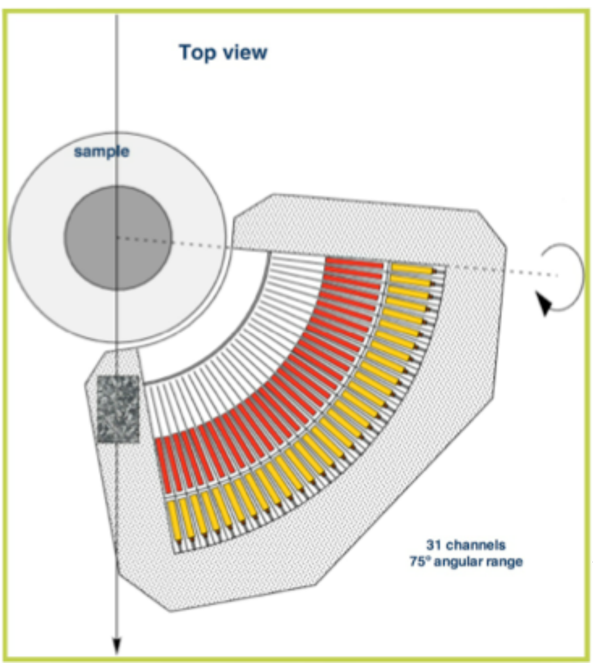
UFO

- ‘Mapping’
- Broad **Q** regions at constant energy transfers

FlatCone





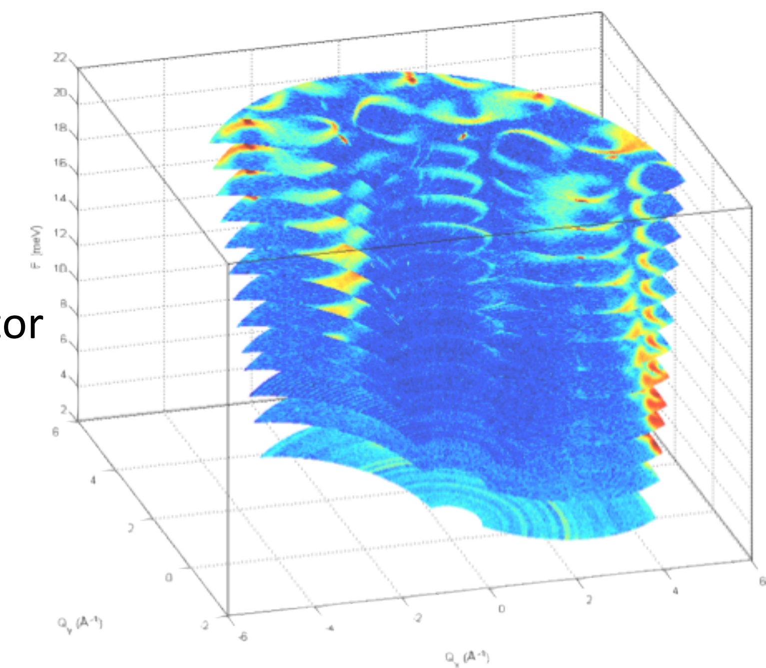


31 individual analyser-detector channels

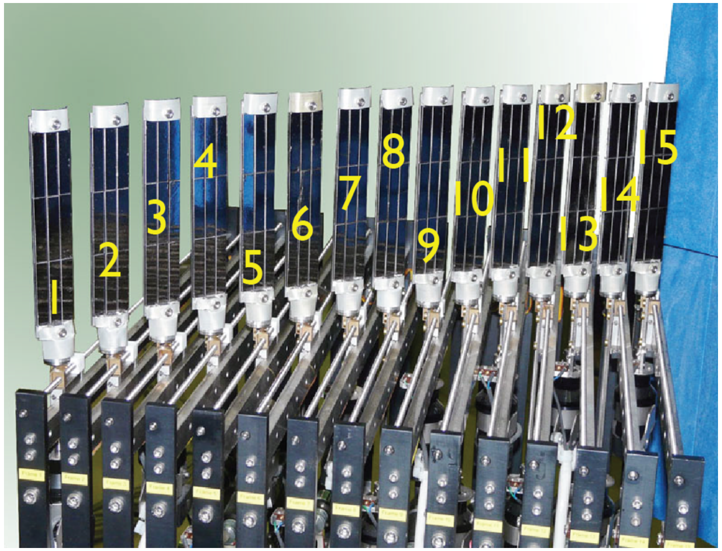
Fixed $\boldsymbol{k}_f$ (1.5 and 3 A^{-1})

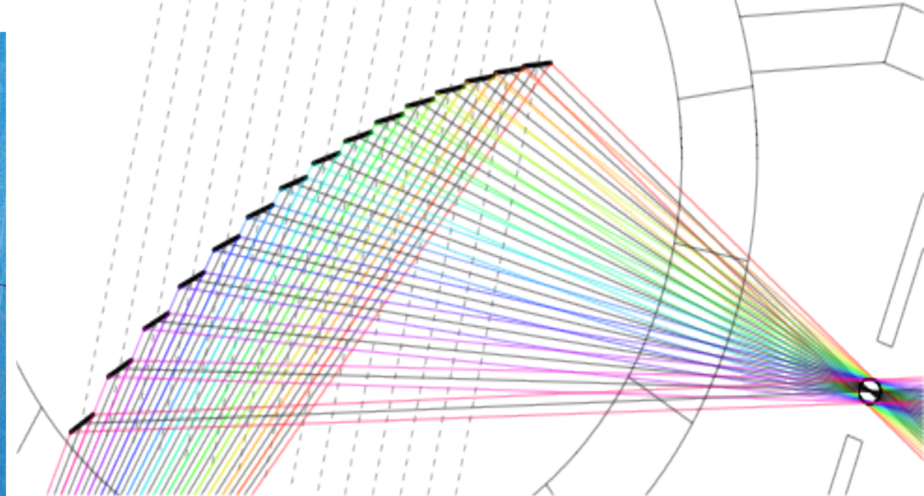
Solid angle: 0.0042

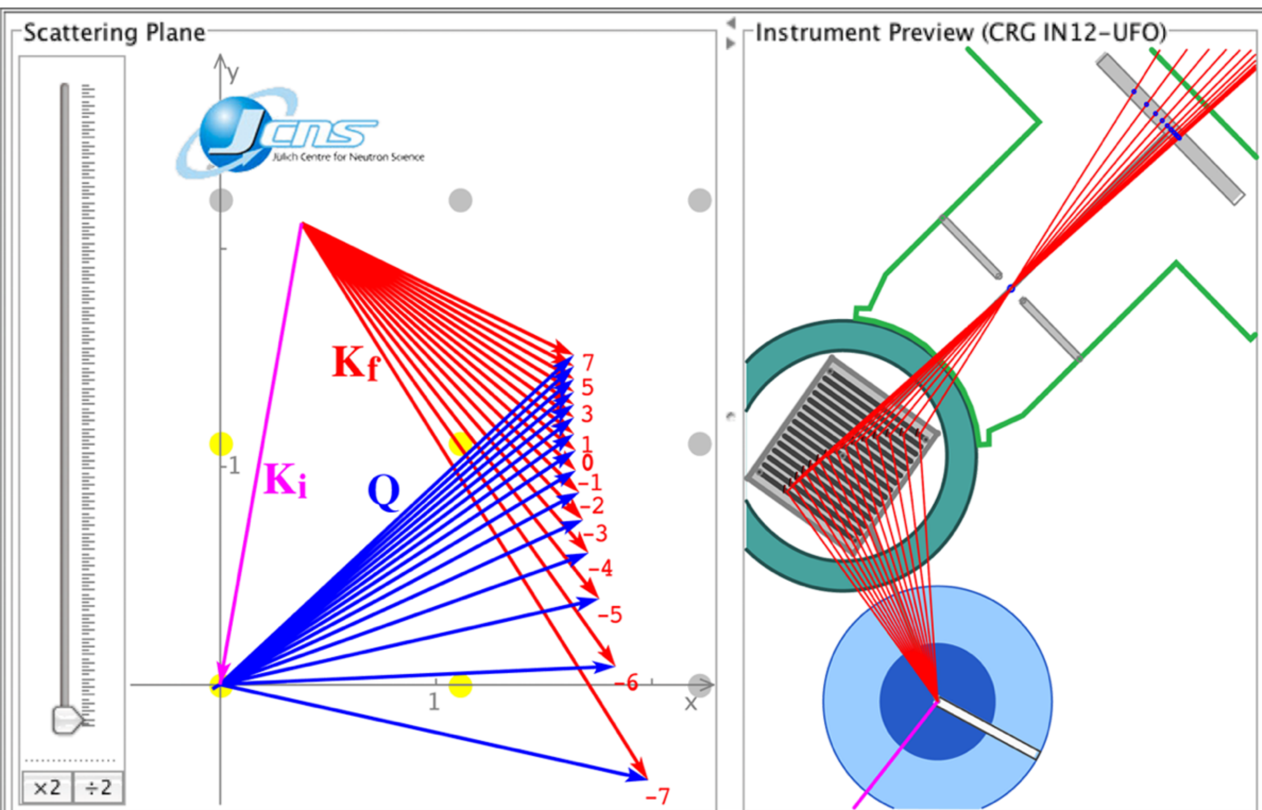
Angular coverage single channel: 2.5 degrees



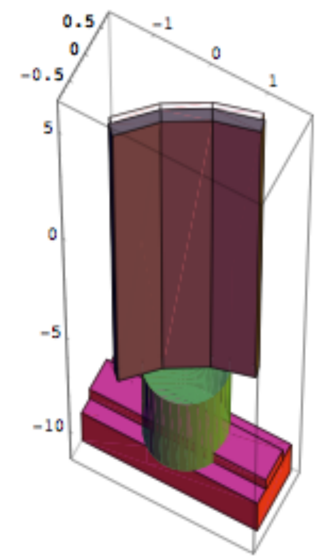
UFO


1
2
3
4
5
6
7
8
9
10
11
12
13
14
15




Scattering Plane
Instrument Preview (CRG IN12-UFO)
Jülich Centre for Neutron Science
y
x
1
1
K_f
K_i
Q
7
5
3
1
0
-1
-2
-3
-4
-5
-6
-7
×2
÷2


20
10
0
-10


0.5
0
-0.5
-1
0
1
5
0
-5
-10

Polarized neutron scattering

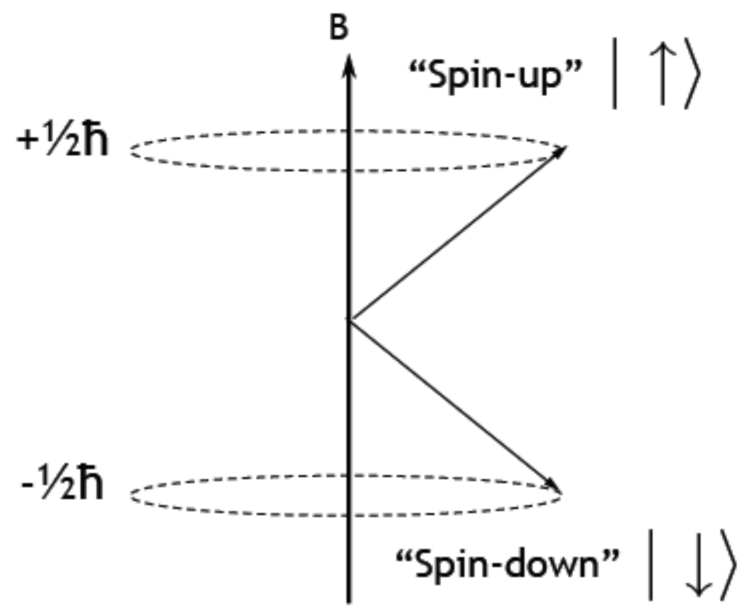
Polarized neutron beam

- spin **s** = ½
- angular momentum ± ½ ħ
- spin vector $\vec{S}_n$

Polarization of a neutron beam = ensemble average over all the neutron spin vectors, normalized to their modulus:

$$P = \left\langle \vec{S}_n \right\rangle / \frac{1}{2} = 2\left\langle \vec{S}_n \right\rangle$$

Application of external field ***B***:
- ONE quantization axis
→ spin-up X spin-down
→ polarization: $P = \dfrac{N_+ - N_-}{N_+ + N_-}$



Polarized neutron beam

$$P = \frac{N_+ - N_-}{N_+ + N_-} = \frac{\frac{N_+}{N_-} - 1}{\frac{N_+}{N_-} + 1} = \frac{F - 1}{F + 1}$$

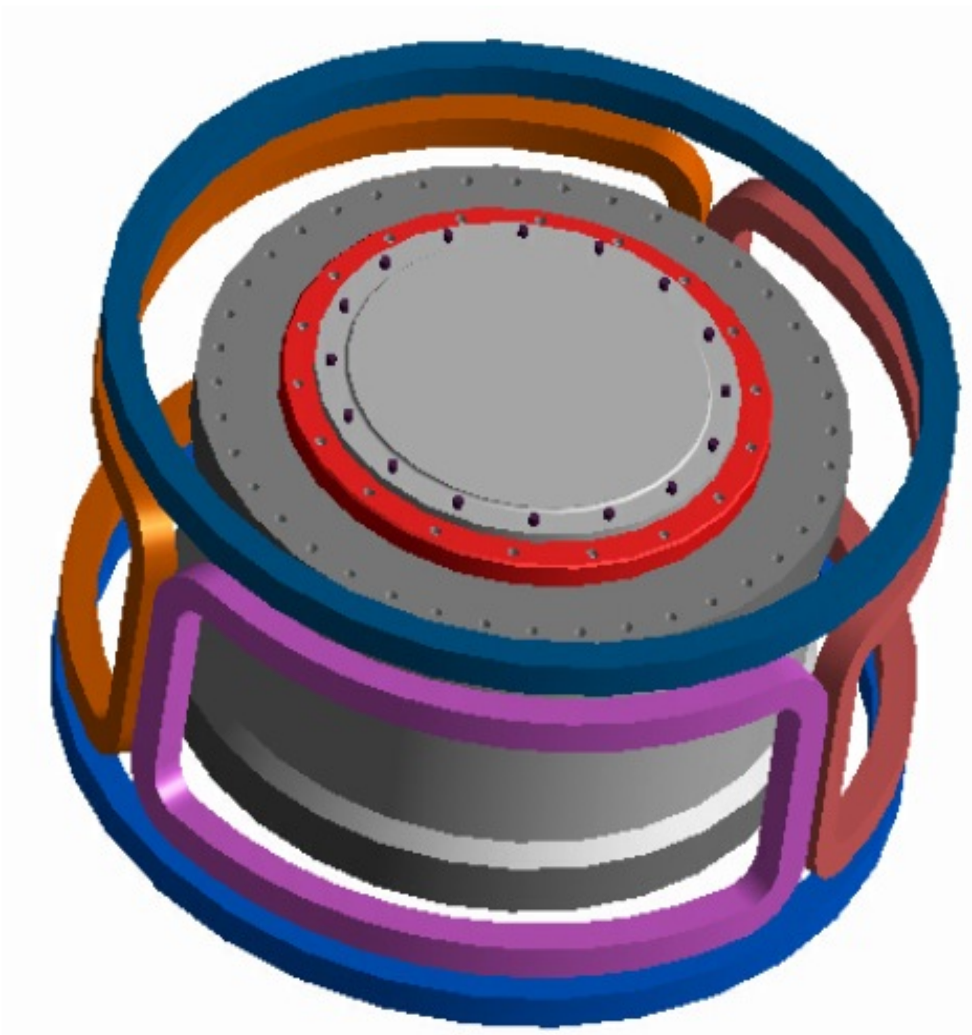
Flipping ratio $F = \frac{N_+}{N_-}$ is measureable quantity

- description for experiments with only 1 quantization axis
  - *Longitudinal polarization analysis*

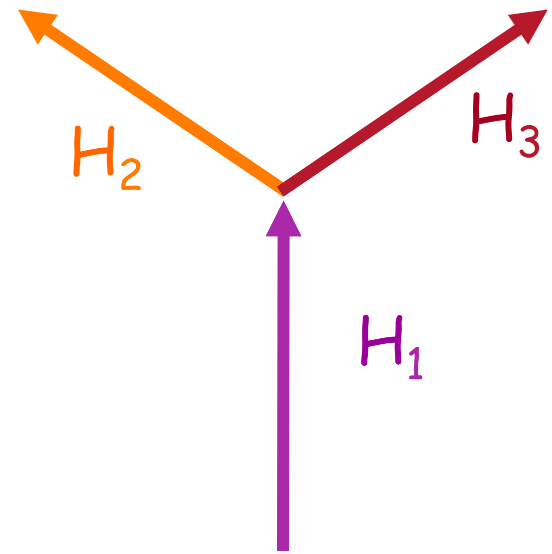
- in case of 3 dimensions
  - *Vector (spherical) polarization analysis*

Define a neutron spin quantization axis

- small magnetic fields

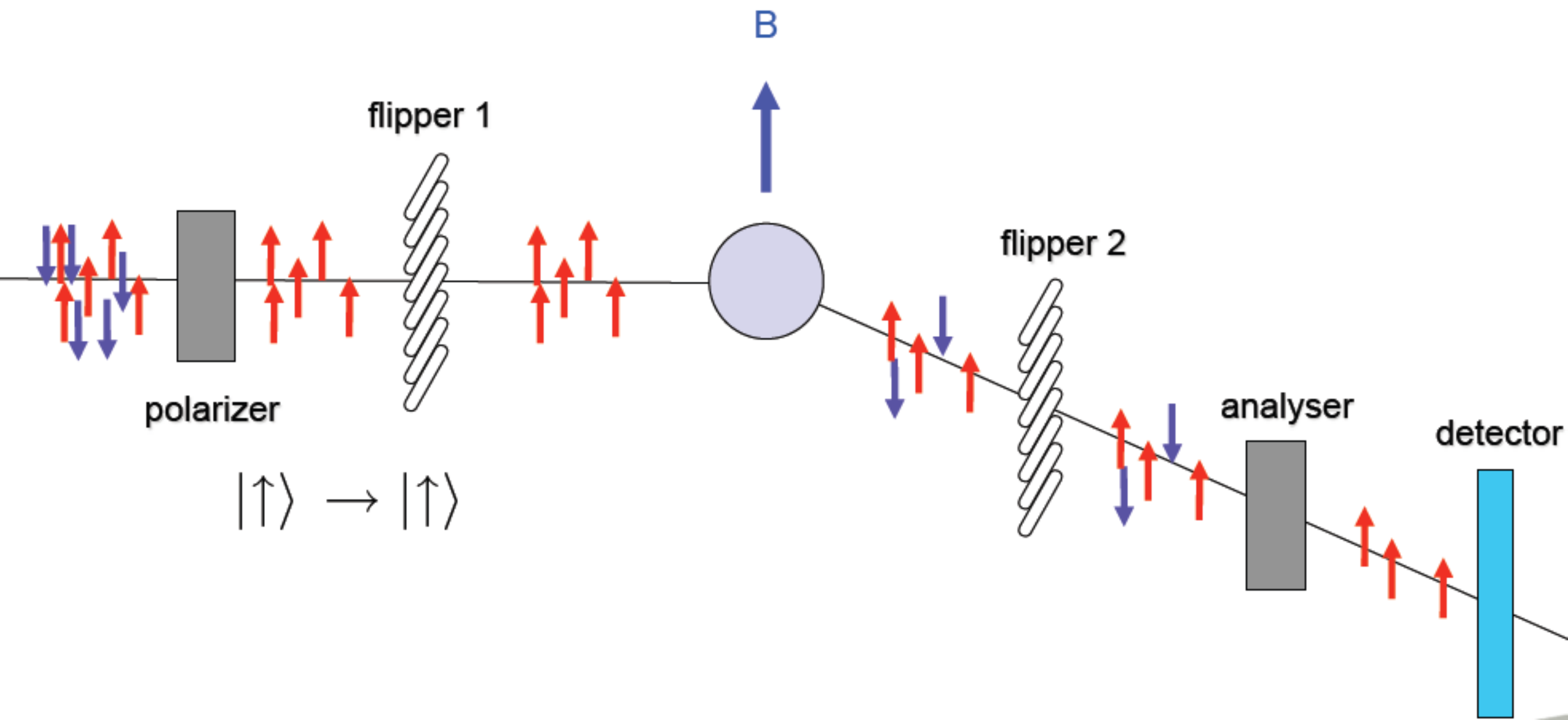


Horizontal field:

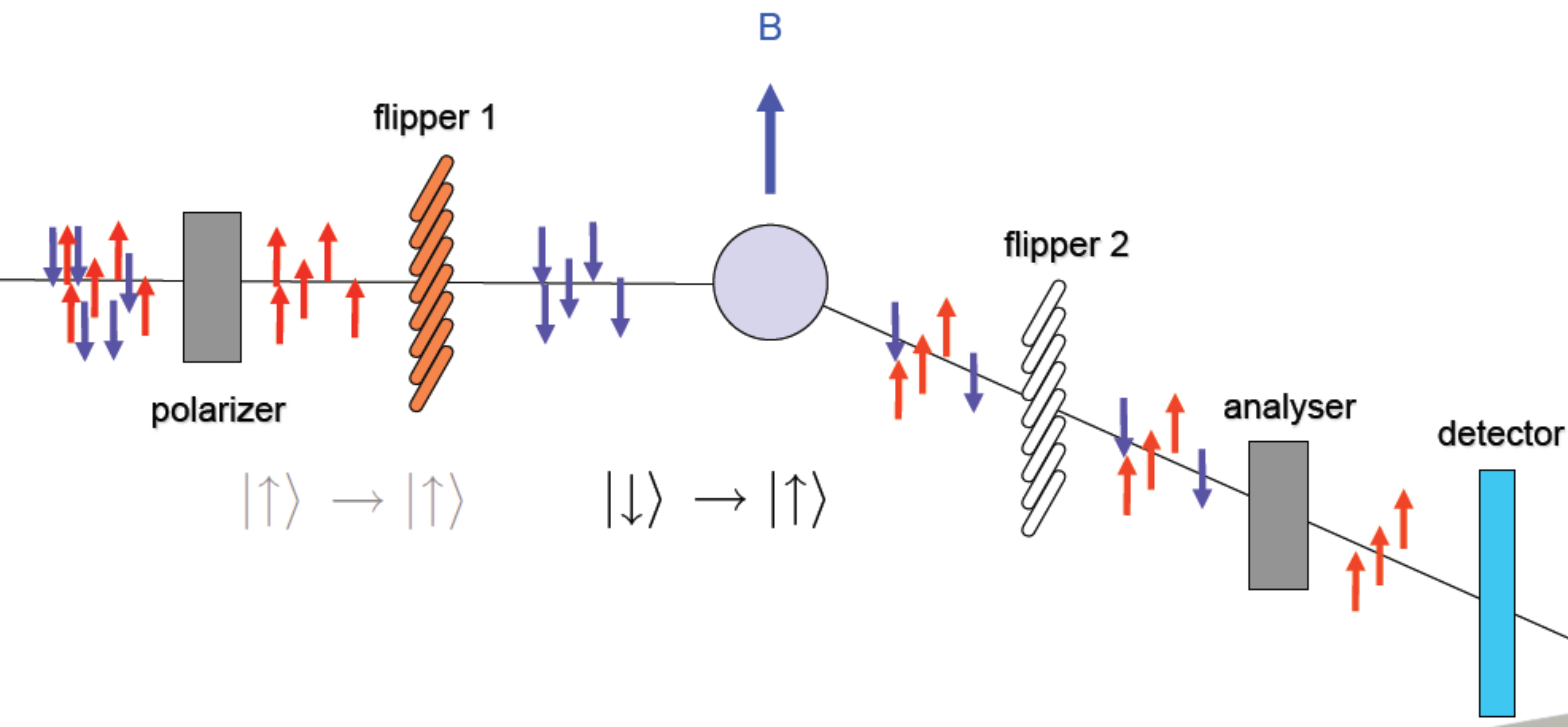


Vertical field:

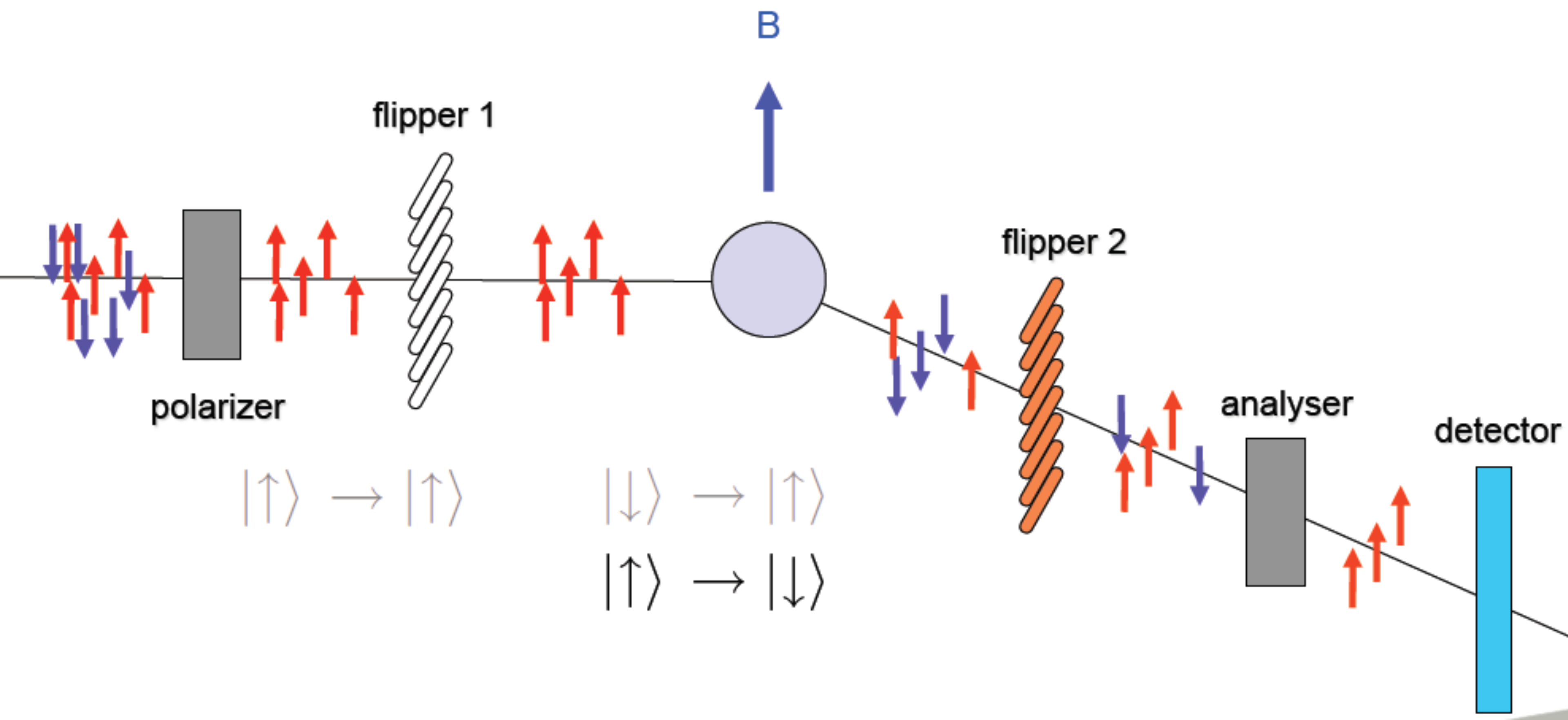
Uniaxial experiment



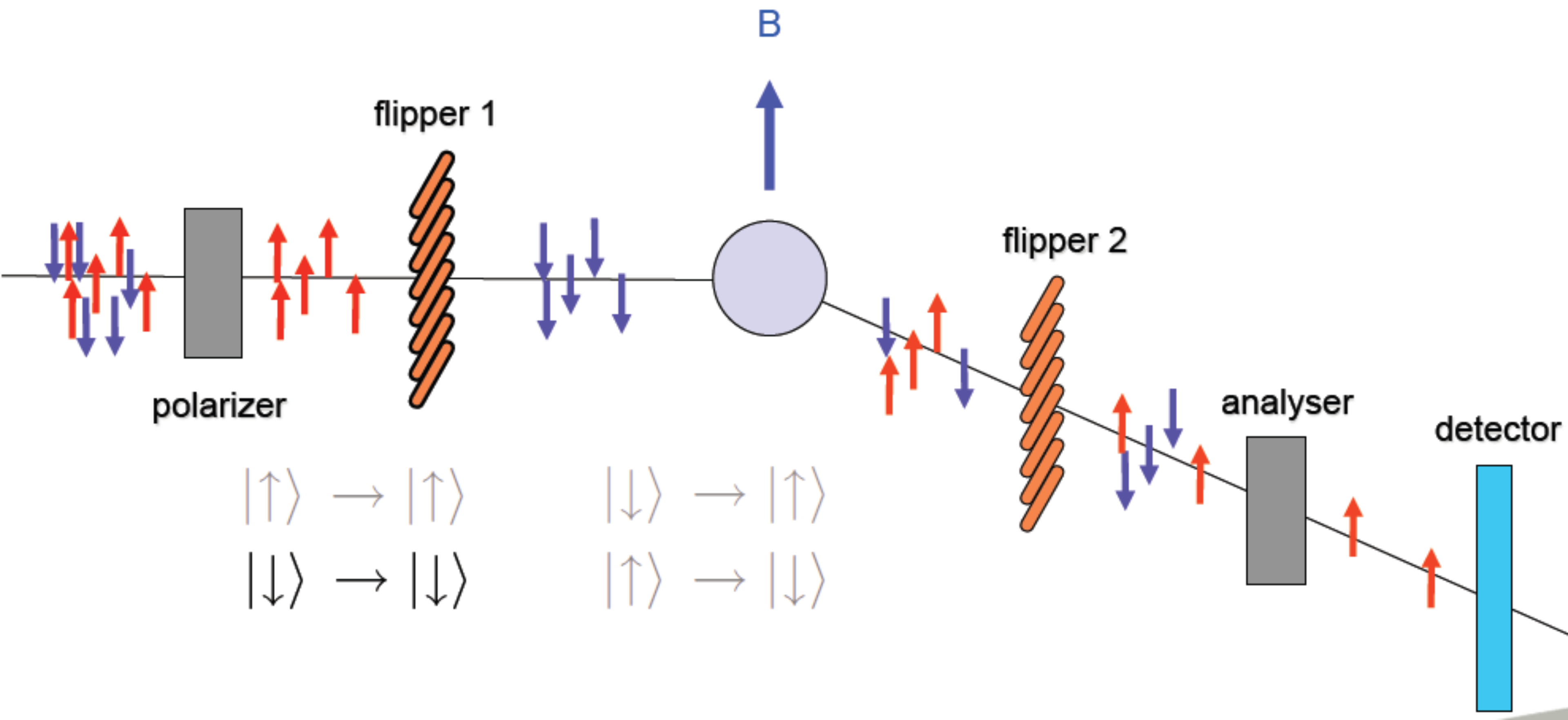
Uniaxial experiment



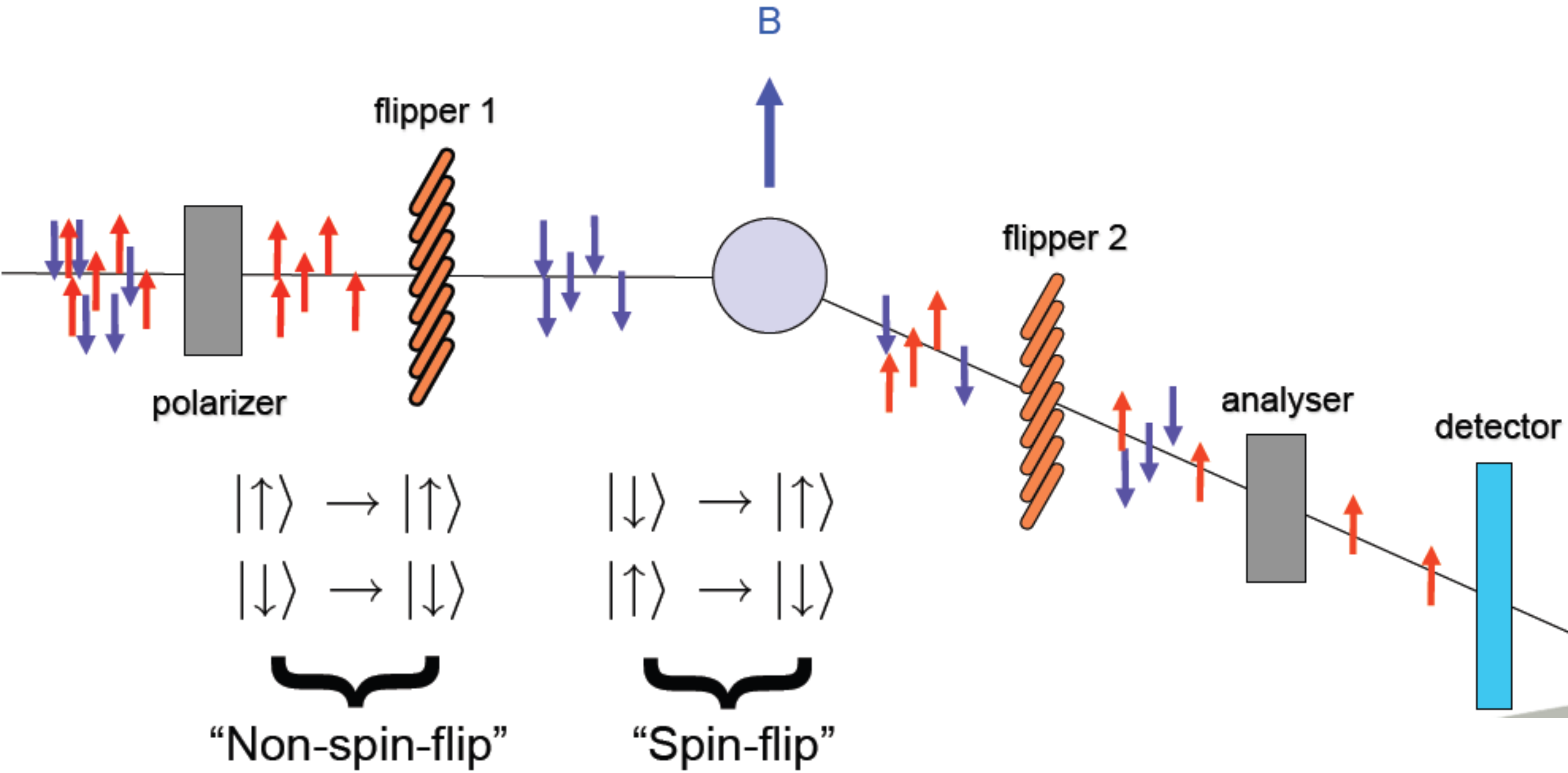
Uniaxial experiment



Uniaxial experiment



Uniaxial experiment

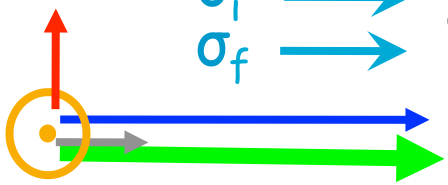


polarized ||x, analysis Q||x

H || x

Non-Spin Flip

$\sigma_i \rightarrow$ $\sigma_f \rightarrow$ or $\sigma_i \leftarrow$ $\sigma_f \leftarrow$

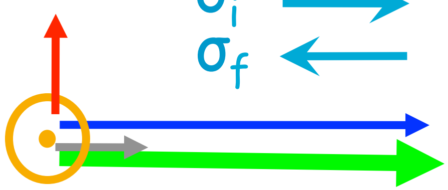


Q

$$\sigma^{xx} = \sigma^{\overline{xx}} = NN^* + \frac{1}{3}\mathrm{NSI} + \mathrm{BG}_{NSF}$$

Spin Flip

$\sigma_i \rightarrow$ $\sigma_f \leftarrow$

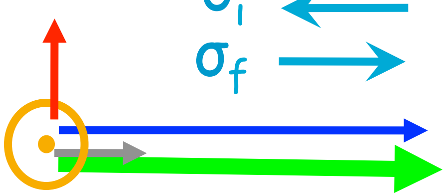


Q

$$\sigma^{x\overline{x}} = \boldsymbol{M}_\perp^* \cdot \boldsymbol{M}_\perp + i\,(\boldsymbol{M}_\perp^* \times \boldsymbol{M}_\perp) \cdot \hat{\boldsymbol{Q}} \; + \frac{2}{3}\mathrm{NSI} + \mathrm{BG}_{SF}$$

Spin Flip

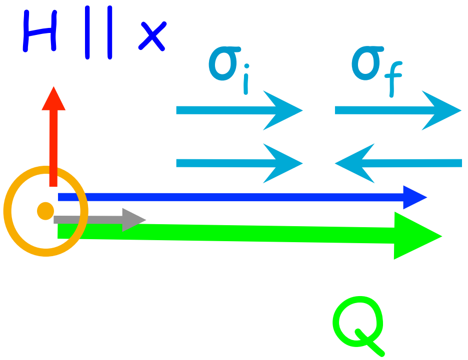
$\sigma_i \leftarrow$ $\sigma_f \rightarrow$



Q

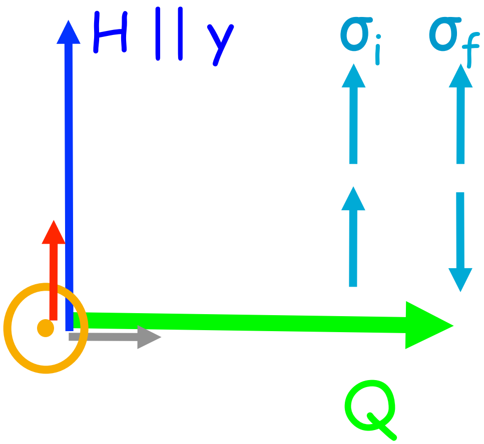
$$\sigma^{\overline{x}x} = \boldsymbol{M}_\perp^* \cdot \boldsymbol{M}_\perp - i\,(\boldsymbol{M}_\perp^* \times \boldsymbol{M}_\perp) \cdot \hat{\boldsymbol{Q}} \; + \frac{2}{3}\mathrm{NSI} + \mathrm{BG}_{SF}$$

NSF/SF along 3 "quantization" axes



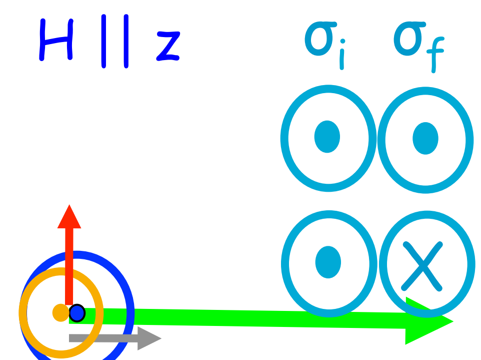
$$\sigma^{xx} = \sigma^{\overline{xx}} = NN^* + \tfrac{1}{3}\mathrm{NSI} + \mathrm{BG}_{NSF}$$

$$\tfrac{1}{2}\sigma^{x\bar{x}} + \tfrac{1}{2}\sigma^{\bar{z}z} = + \boxed{M_\perp^{y*}M_\perp^{y}} + \boxed{M_\perp^{z*}M_\perp^{z}} + \tfrac{2}{3}\mathrm{NSI} + \mathrm{BG}_{SF}$$



$$\tfrac{1}{2}\sigma^{yy} + \tfrac{1}{2}\sigma^{\overline{yy}} = NN^* + \boxed{M_\perp^{y*}M_\perp^{y}} + \tfrac{1}{3}\mathrm{NSI} + \mathrm{BG}_{NSF}$$

$$\sigma^{y\bar{y}} = \sigma^{\bar{y}y} = + \boxed{M_\perp^{z*}M_\perp^{z}} + \tfrac{2}{3}\mathrm{NSI} + \mathrm{BG}_{SF}$$



$$\tfrac{1}{2}\sigma^{zz} + \tfrac{1}{2}\sigma^{\overline{zz}} = NN^* + \boxed{M_\perp^{z*}M_\perp^{z}} + \tfrac{1}{3}\mathrm{NSI} + \mathrm{BG}_{NSF}$$

$$\sigma^{z\bar{z}} = \sigma^{\bar{z}z} = + \boxed{M_\perp^{y*}M_\perp^{y}} + \tfrac{2}{3}\mathrm{NSI} + \mathrm{BG}_{SF}$$

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DOI: 10.1051/sfn/201413010002

ThALES

Three Axis Low-Energy Spectrometer

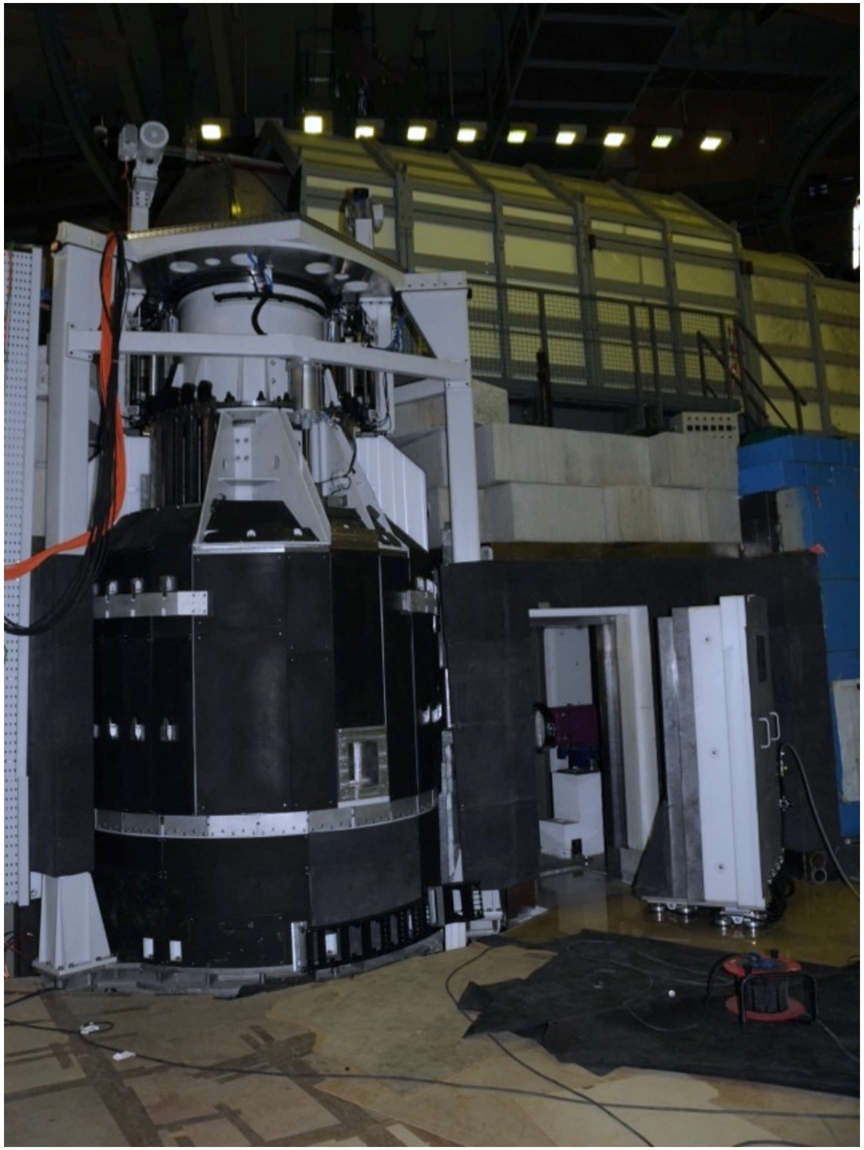
ThALES instrument construction

December 2013

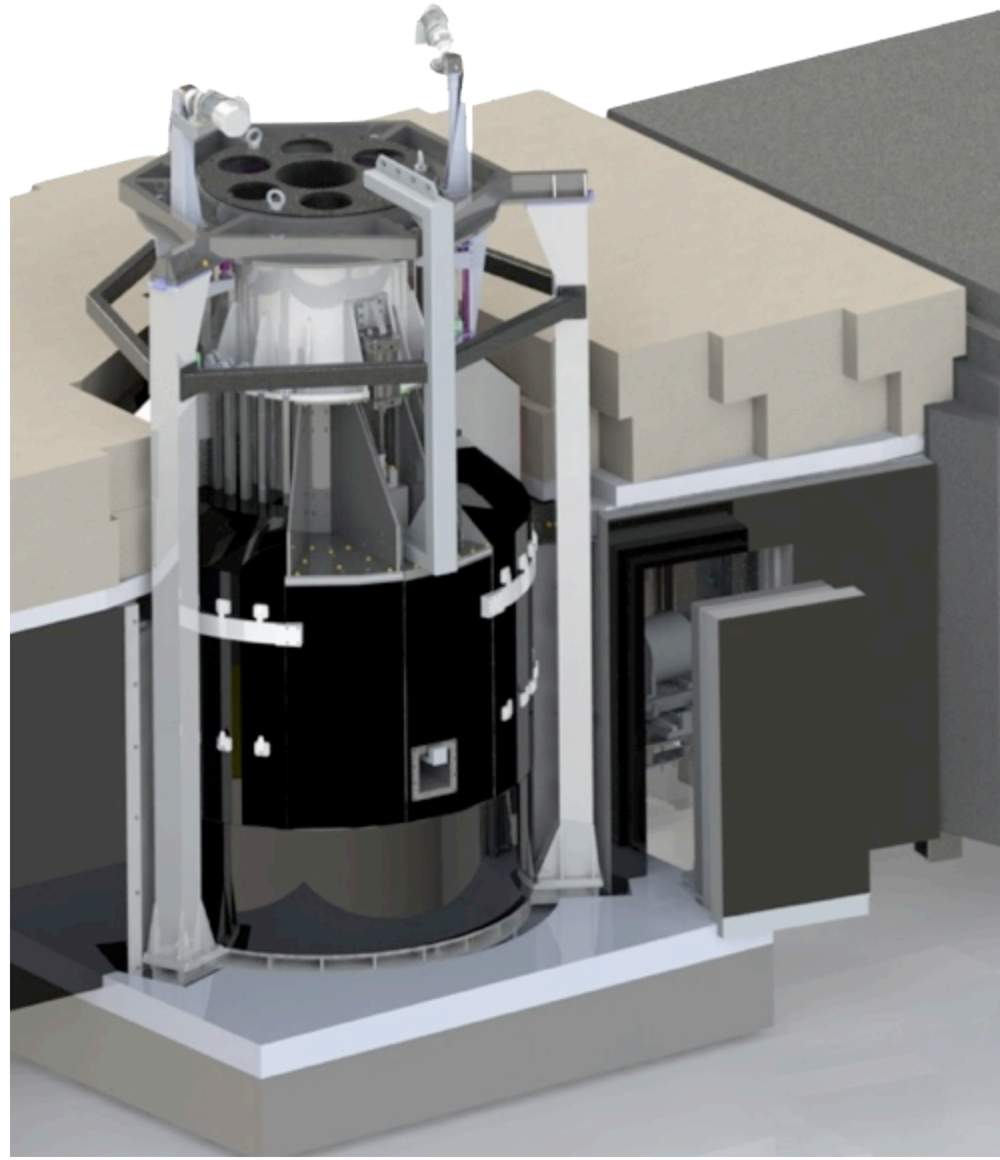
ThALES – instrument construction

March 2014

ThALES – instrument construction



4 July 2014



after detailed design study, 2011

ThALES instrument construction

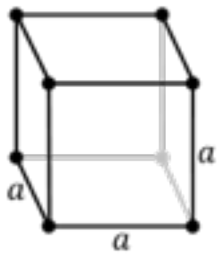
September 2014

ThALES

May 2015

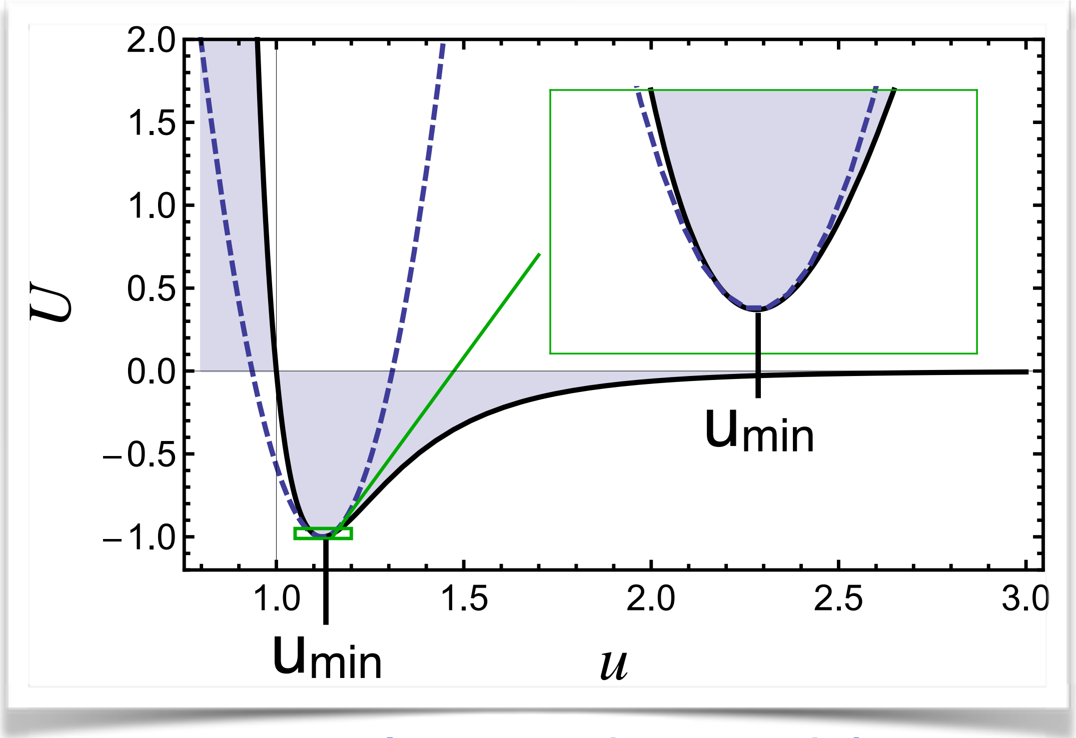
Phonon

CLASSICAL DESCRIPTION
Crystal = Periodic lattice of atoms
Phonon = Collective vibration of the atoms in the crystal



Crystal lattice

HYPOTHESIS
Harmonic approximation → small displacement u of the atoms
Interaction between nearest neighbors

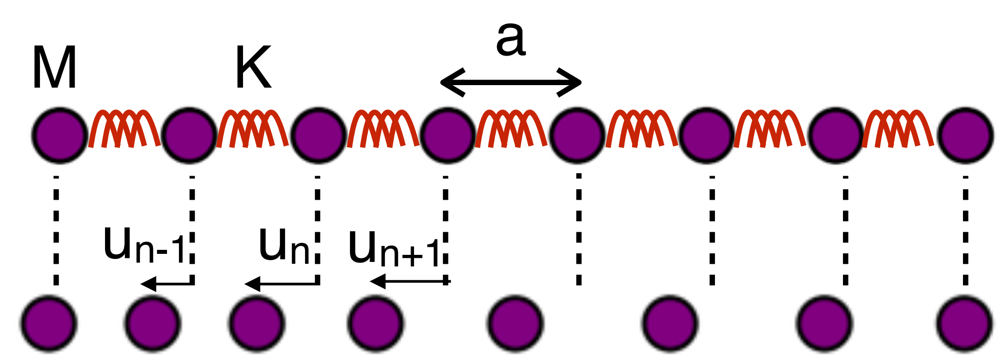


[Interatomic potential]

MODEL
Atoms connected by springs

Phonon

Monoatomic chain



Force exerted on the atom n : F=K(u(n+1)-u(n))+K(u(n-1)-u(n))

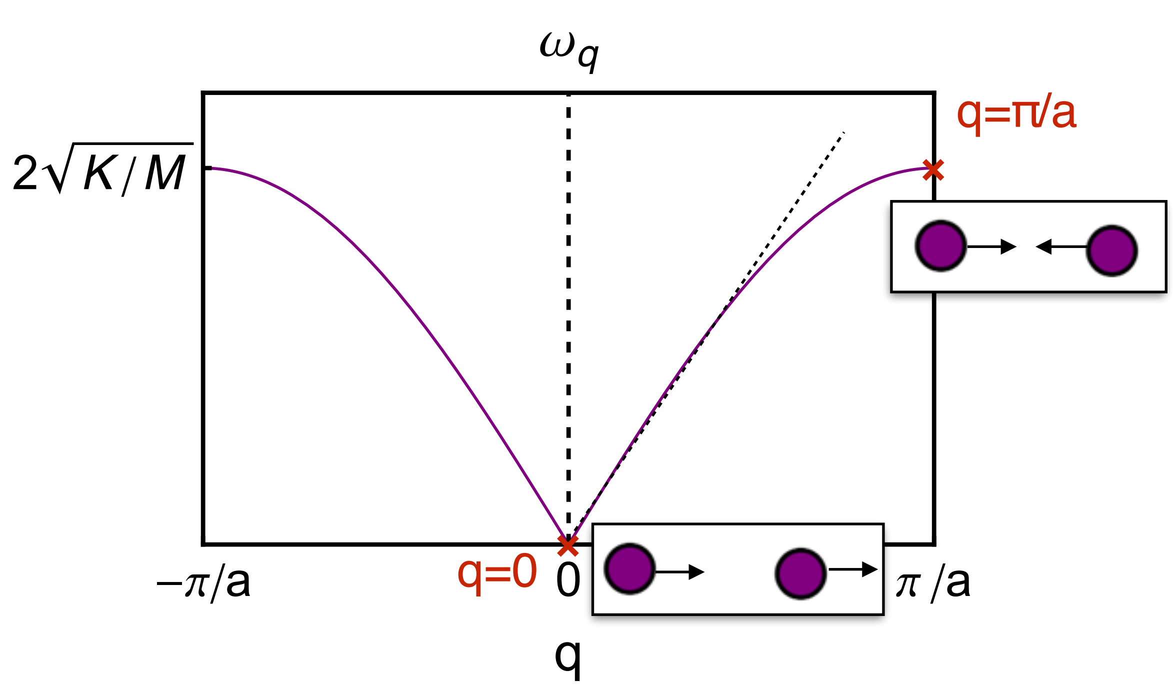
Fundamental principal of dynamics: $M\frac{\partial^2 u}{\partial^2 t} = -\frac{\partial U}{\partial u} = F$

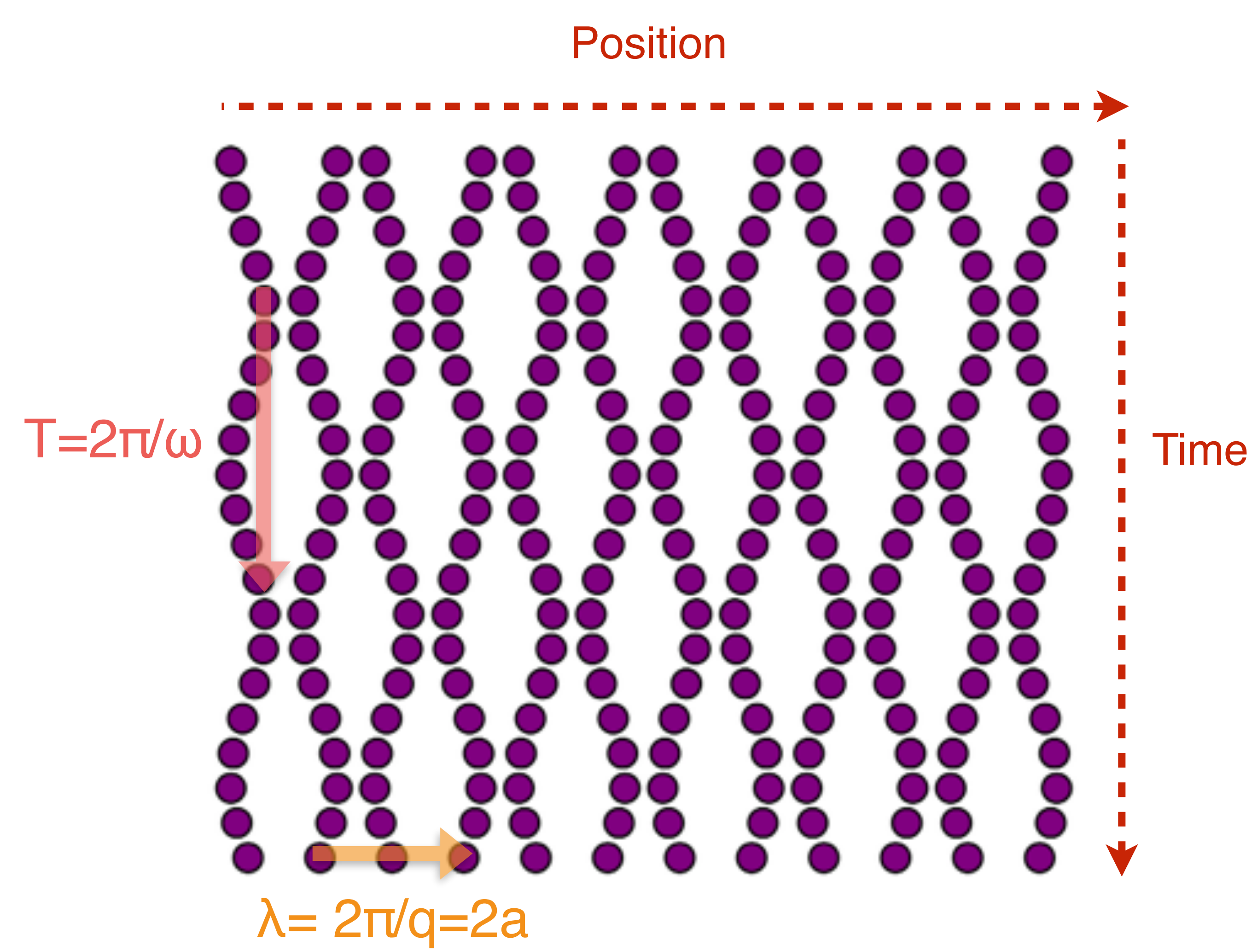
Solutions: collective waves $u(n)=u_0\exp(i(qna - \omega t))$

Dispersion relation: $\omega_q = 2\sqrt{\frac{K}{M}}\left|sin\left(\frac{qa}{2}\right)\right|$

For small qa: $\omega_q = \sqrt{\frac{K}{M}}qa$, $v = \frac{\partial \omega_q}{\partial q} = \sqrt{\frac{K}{M}}a$

One acoustic mode of vibration

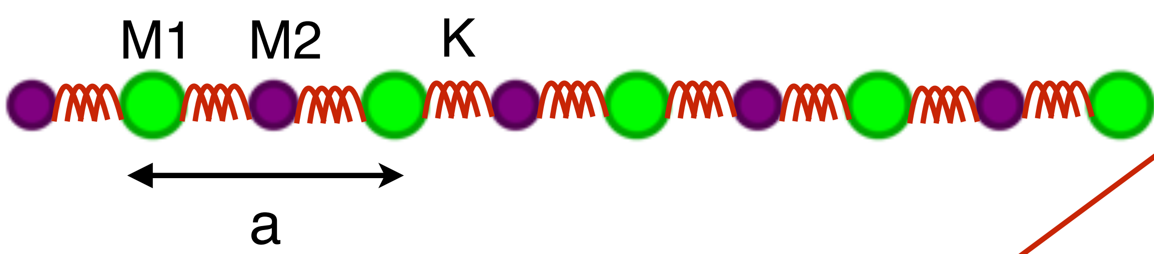




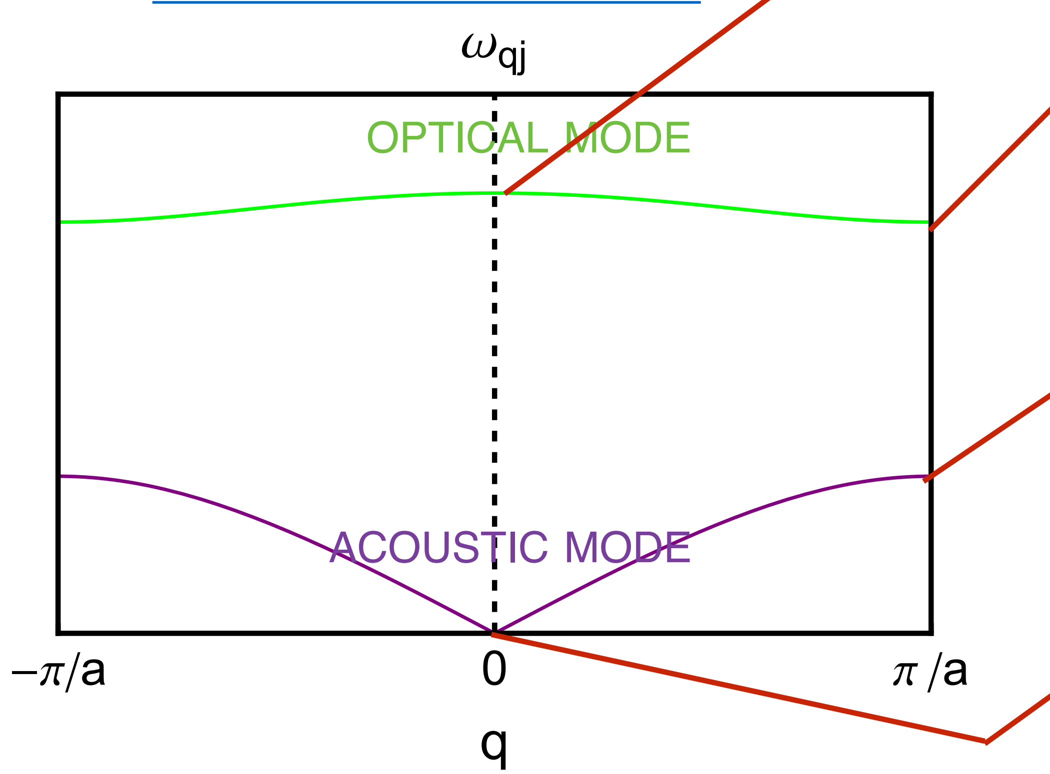
Phonon

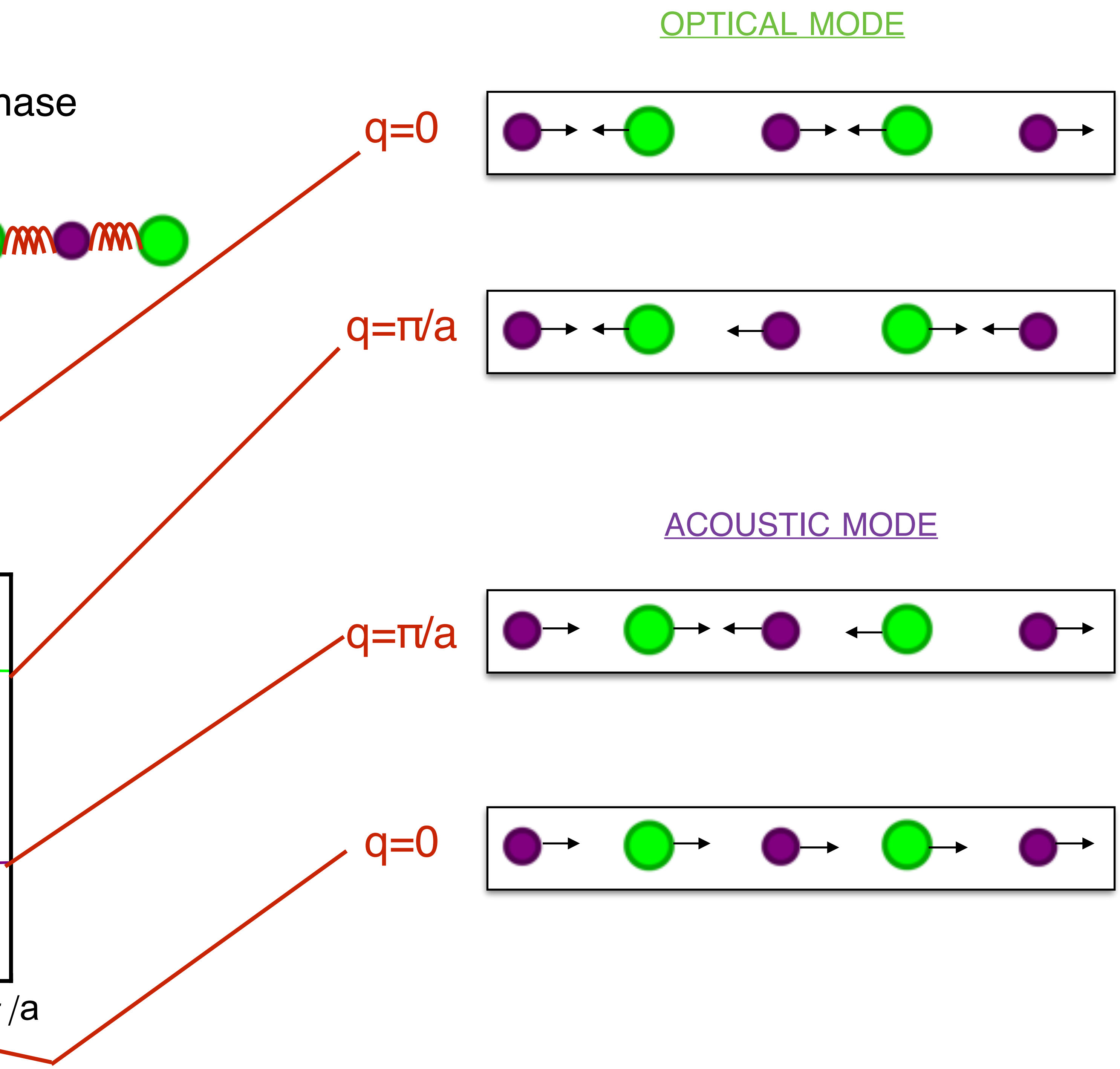
Diatomic chain

Acoustic phonon: atoms vibrate in phase
Optical phonon: atoms vibrate in opposite phase



Two modes of vibration

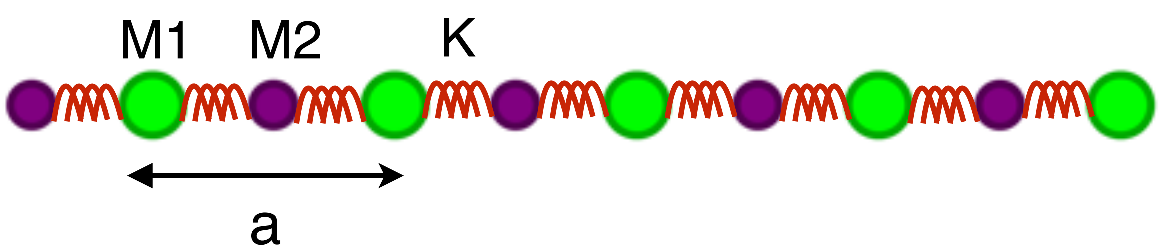


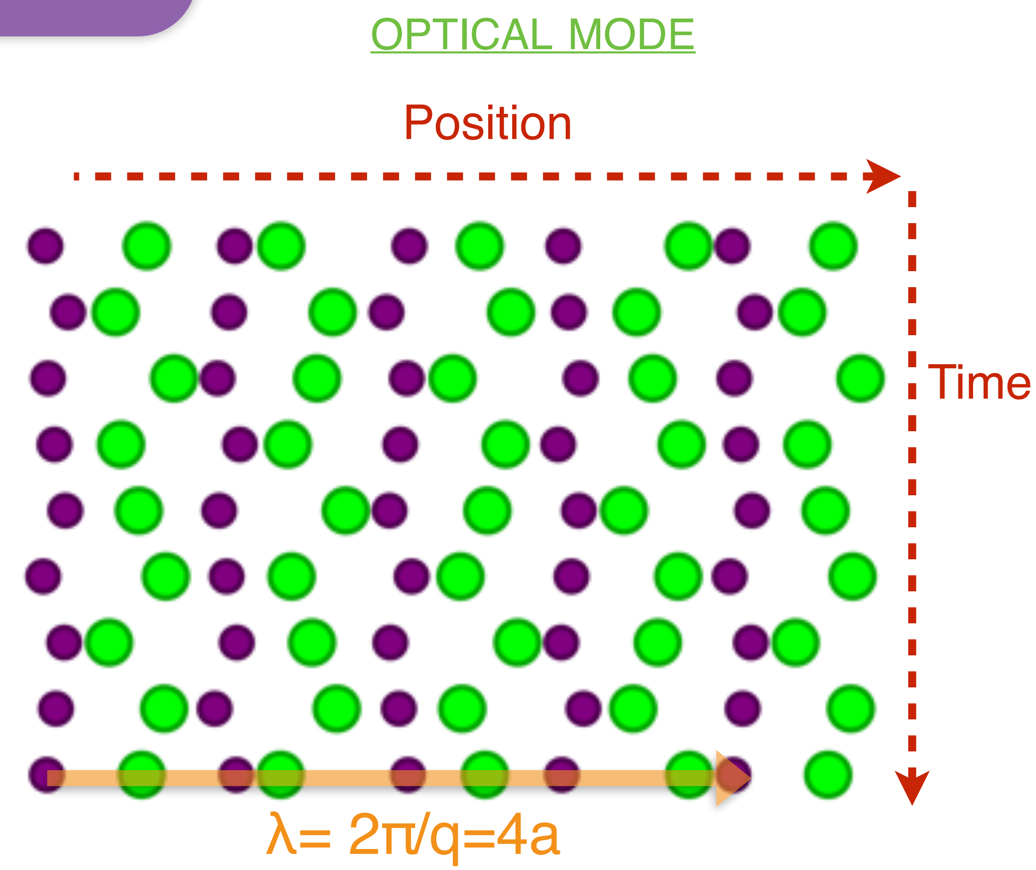


Phonon

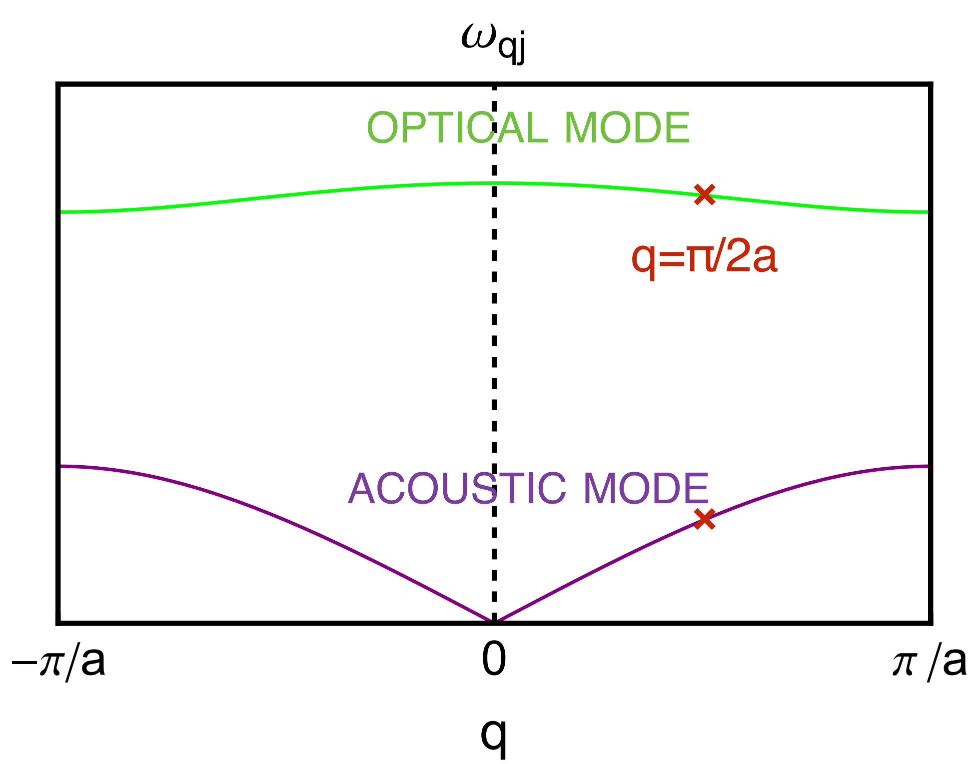
DIATOMIC CHAIN

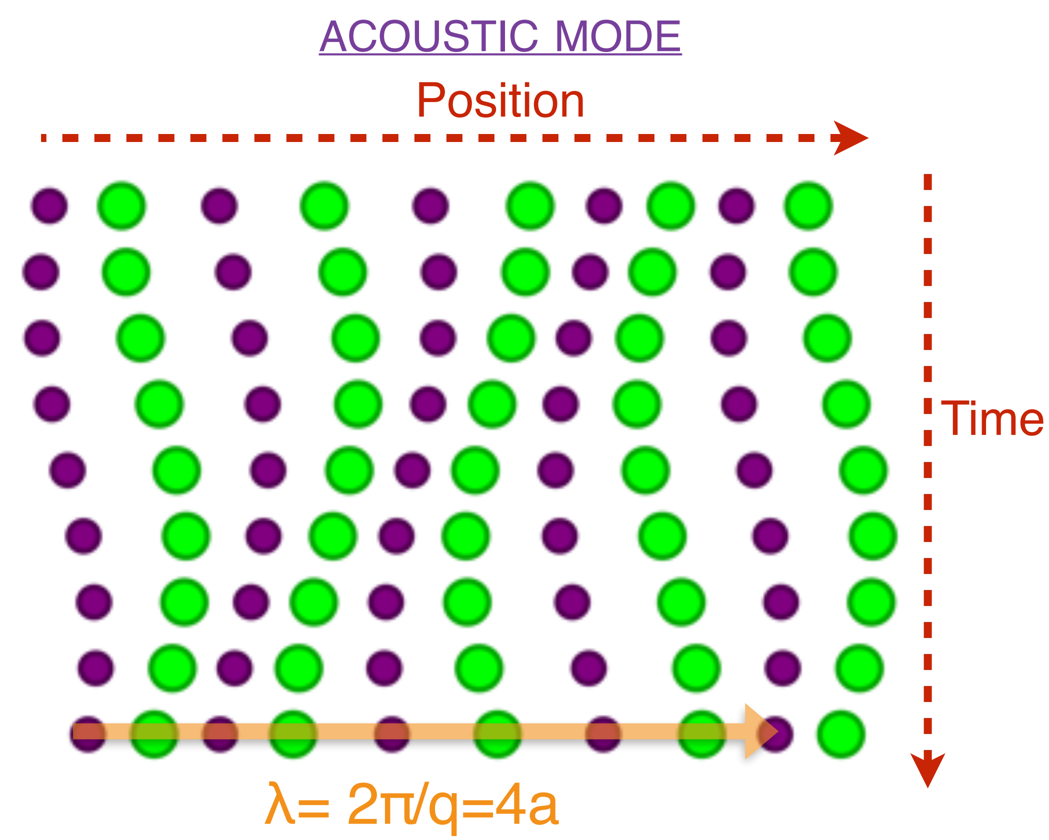
Acoustic phonon: atoms vibrate in phase
Optical phonon: atoms vibrate in phase opposition





Two modes of vibration





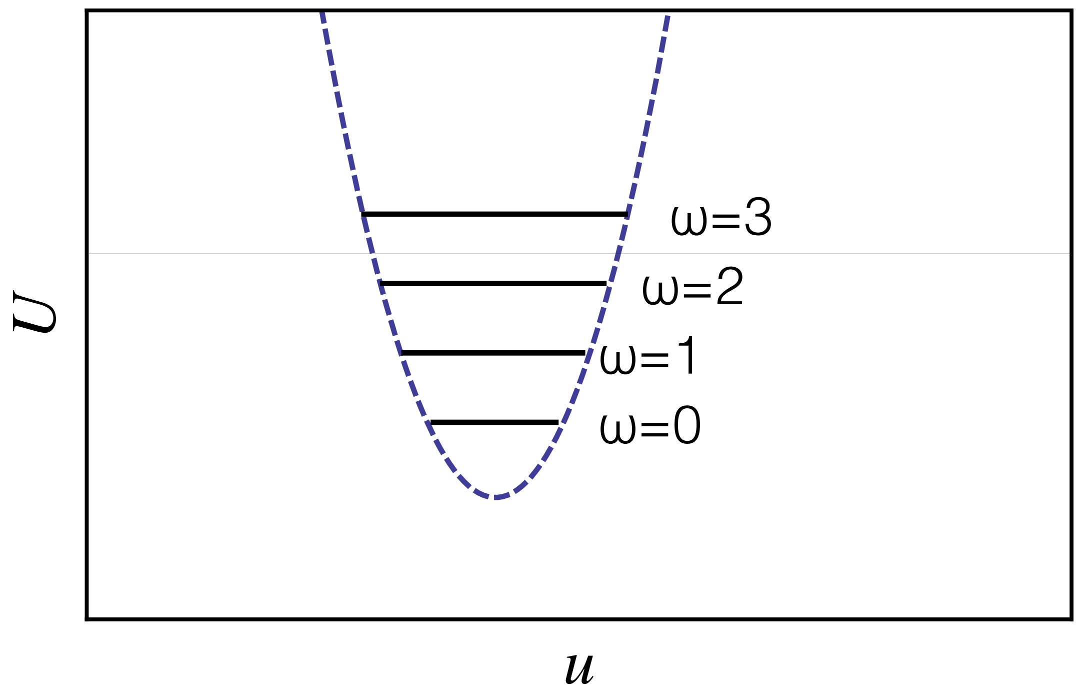
Phonon

Quantum description

Each vibrational mode is associated to quasi particles called phonons. Their energy is quantized (analogous to the concept of photons, the quantum of electromagnetic radiation)

$$E_{qj} = \left(\langle n_{qj} \rangle + \frac{1}{2}\right) \hbar\omega_{qj} \quad \text{with} \quad \langle n_{qj} \rangle = \frac{1}{e^{\frac{\hbar\omega_{qj}}{k_B T}} - 1}$$

Phonon = quasi particle with an energy (frequency ω) and a momentum (wave-vector q)



Interatomic potential

Phonon

3D CRYSTAL

r atoms in unit cell

Each atom has 3 degrees of freedom (3 directions of polarisation $\vec{e}$

for 1 direction of propagation $\vec{q}$ belonging to the first Brillouin zone) ⇒ 3r degrees of freedom

⇒ 3r eigenvectors associated to 3r eigenvalues ω_j

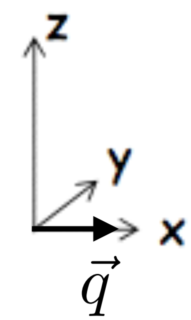
3 acoustic branches (one longitudinal and two transverse) + (3r-3) optical branches

$\vec{e} \, // \, \vec{q}$: longitudinal mode (L mode)

$\vec{e} \perp \vec{q}$: transverse mode (T mode)

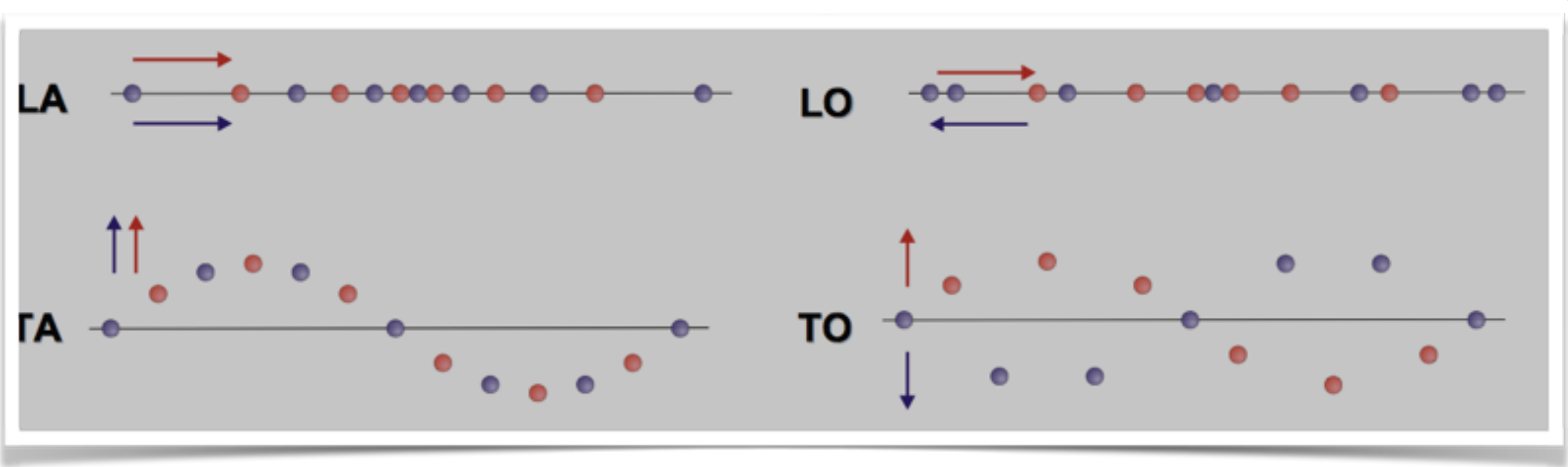
<u>2 atoms in unit cell</u>

The eigenvectors are:



$$e^x_\pm = \begin{pmatrix} \begin{pmatrix} +1 \\ 0 \\ 0 \end{pmatrix} \\ \\ \begin{pmatrix} -1 \\ 0 \\ 0 \end{pmatrix} \end{pmatrix}$$

$$e^y_\pm = \begin{pmatrix} \begin{pmatrix} 0 \\ +1 \\ 0 \end{pmatrix} \\ \\ \begin{pmatrix} 0 \\ -1 \\ 0 \end{pmatrix} \end{pmatrix} \qquad e^z_\pm = \begin{pmatrix} \begin{pmatrix} 0 \\ 0 \\ +1 \end{pmatrix} \\ \\ \begin{pmatrix} 0 \\ 0 \\ -1 \end{pmatrix} \end{pmatrix}$$



Phonon

Coherent 1 phonon differential scattering cross section

$$\left(\frac{d^2\sigma}{d\Omega dE_f}\right)_{\pm ph} = \frac{k_f}{k_i}\frac{(2\pi)^3}{V}\sum_{\vec{\tau}}\sum_{s}\frac{1}{\omega_s}\left|F_s(\vec{Q})\right|^2\langle n_\pm\rangle_T\,\delta(\omega\mp\omega_s)\,\delta(\vec{Q}\mp\vec{q}-\vec{\tau})$$

Intensity   Position

V: Volume of the unit cell

s: Double index (q,j) representing a mode s (s=1 to 3r)

$\vec{q}$: Wave-vector of the mode s

j: Polarisation index of the mode s

r: Number of atoms per unit cell

ω_s: Angular frequency of mode s

$\vec{\tau}$: Reciprocal lattice vector

$$F_s(\vec{Q}) = \sum_{d=1}^{r}\frac{\bar{b}_d}{\sqrt{M_d}}e^{-W_d}(\vec{Q}\cdot\vec{e}_{ds})e^{i\vec{Q}\cdot\vec{d}}$$

$F_s(\vec{Q})$: structure factor of the phonon of the mode s

d: Equilibrium position of the d^{th} atom in the unit cell

$\vec{e}_{ds}$: Polarisation vector for the atom at position d for the mode s / Eigenvector of the phonon

M_d: Mass of the atom at the position d

e^{-Wd}: Debye-Waller factor

$e^{i\vec{Q}\cdot\vec{d}}$: phase factor

$\vec{Q}\cdot\vec{e}_{ds}$: polarisation factor

$\langle n_\pm\rangle_T$: Occupation number, Bose factor, average population of the phonons

$\bar{b}_d$: Mean value of the scattering length at the position d

Phonon

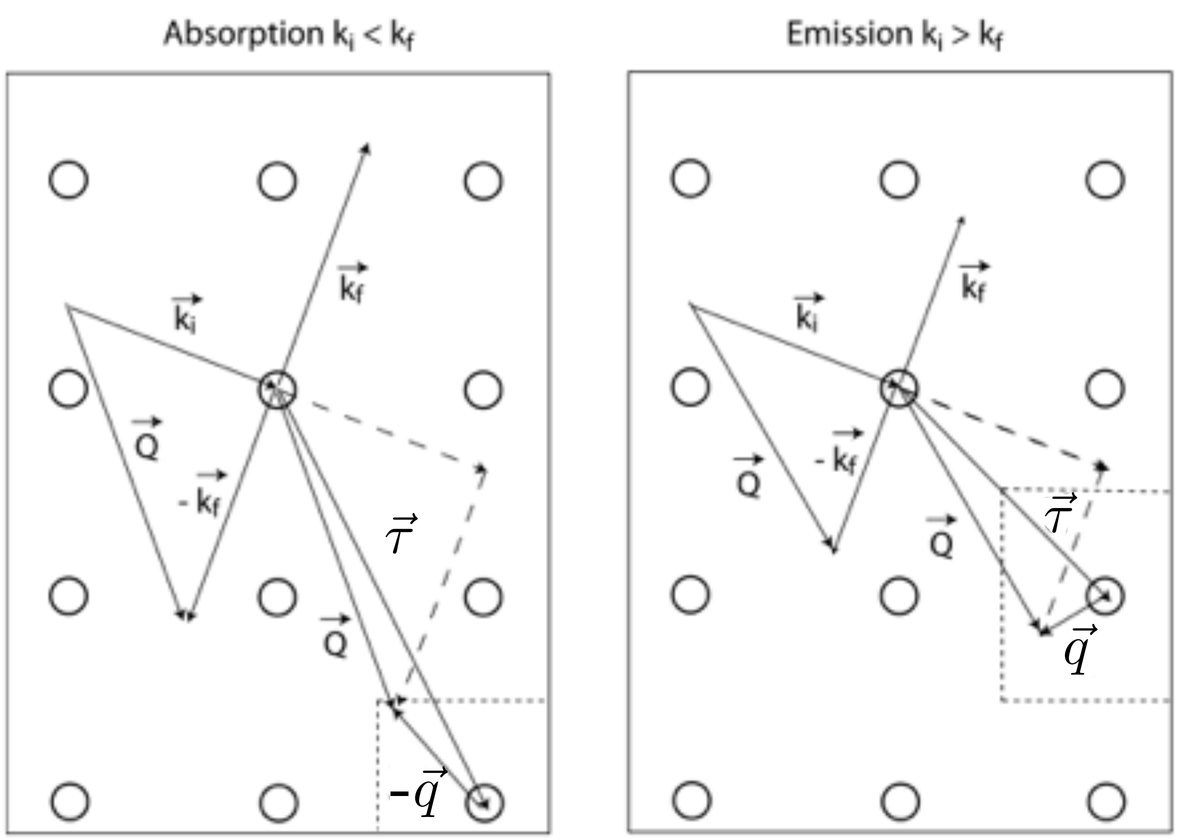
POSITION

$$\boxed{\delta(\omega \mp \omega_s)\delta(\vec{Q} \mp \vec{q} - \vec{\tau})}$$

Momentum and energy conservation

$\vec{Q} = \vec{q} + \vec{\tau}$ ω=$\omega_{s>0}$ Phonon creation (emission)

$\vec{Q} = -\vec{q} + \vec{\tau}$ ω=-$\omega_{s<0}$ Phonon annihilation (absorption)



INTENSITY

Thermal occupancy factor
= Mean number of phonons

Phonon creation

$$\boxed{\langle n+ \rangle_T} = \langle n_s + 1 \rangle_T = \frac{1}{1 - e^{\left(\frac{-\hbar\omega_s}{k_B T}\right)}} \xrightarrow[T=0]{} 1 \quad \xrightarrow[T=\infty]{} \infty$$

Phonon annihilation

$$\boxed{\langle n- \rangle_T} = \langle n_s \rangle_T = \frac{1}{e^{\left(\frac{\hbar\omega_s}{k_B T}\right)} - 1} \xrightarrow[T=0]{} 0 \quad \xrightarrow[T=\infty]{} \infty$$

Phonon

INTENSITY

$$\boxed{F_s(\vec{Q}) = \sum_{d=1}^{r} \frac{\bar{b}_d}{\sqrt{M_d}} e^{-W_d} (\vec{Q}\cdot\vec{e}_{ds}) e^{i\vec{Q}\cdot\vec{d}}}$$

Polarisation factor $\vec{Q}\cdot\vec{e}_{ds}$

Transverse phonons of the first Brillouin zone ($\vec{Q} = \vec{q}$ because $\vec{\tau} = 0$)

$\vec{e}_{ds}\cdot\vec{q} = 0 \;\forall d$

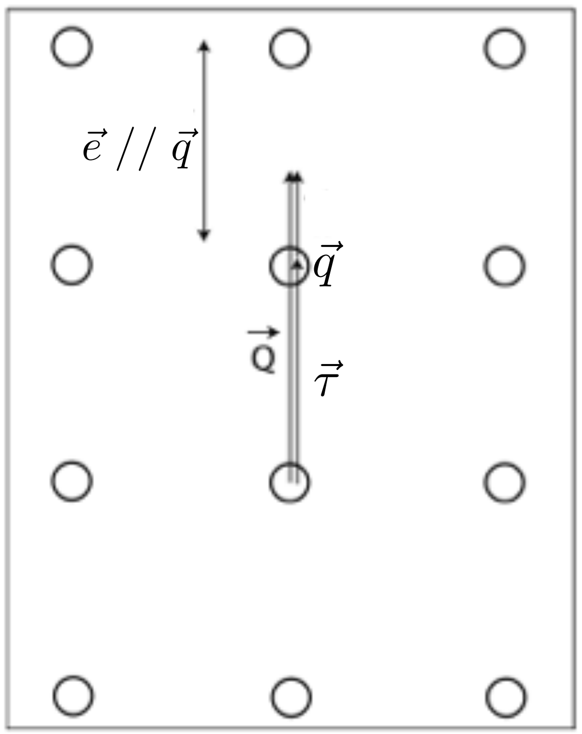
$\vec{Q}\cdot\vec{e}_{ds} = 0$, $F_s(\vec{Q}) = 0$

⇒ The transverse modes do not give any signal in the first Brillouin zone

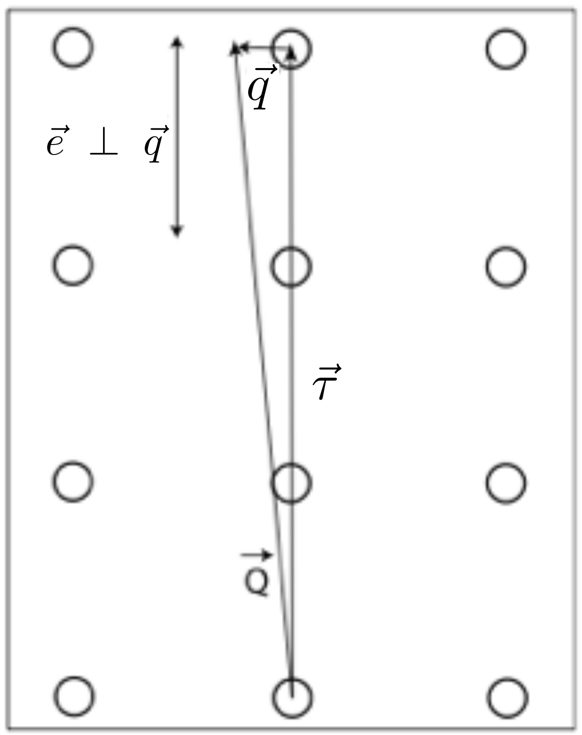
- Intensity $|F_s(\vec{Q})|^2$ min for $\vec{Q} \perp \vec{e}_{ds}$
- Intensity $|F_s(\vec{Q})|^2$ max for $\vec{Q} // \vec{e}_{ds}$
- Intensity increases with Q^2

⇒ For a wave vector $\vec{q}$ given, choose $|\vec{Q}| = |\vec{\tau} \pm \vec{q}|$ the largest possible

Longitudinal configuration
$\vec{e} // \vec{q}$



Transversal configuration
$\vec{q}$ is more or less $\perp$ to $\vec{e}$



Debye-Waller factor e^{-W_d}

$$e^{-2W_d} = e^{-\langle(\vec{Q}\cdot\vec{u}_d)^2\rangle_T}$$

Scattering length $\bar{b}_d$

Phonon

PHONONS IN CsCl

- cc single crystal
2 atoms in unit cell ⇒ 3*2 = 6 branches

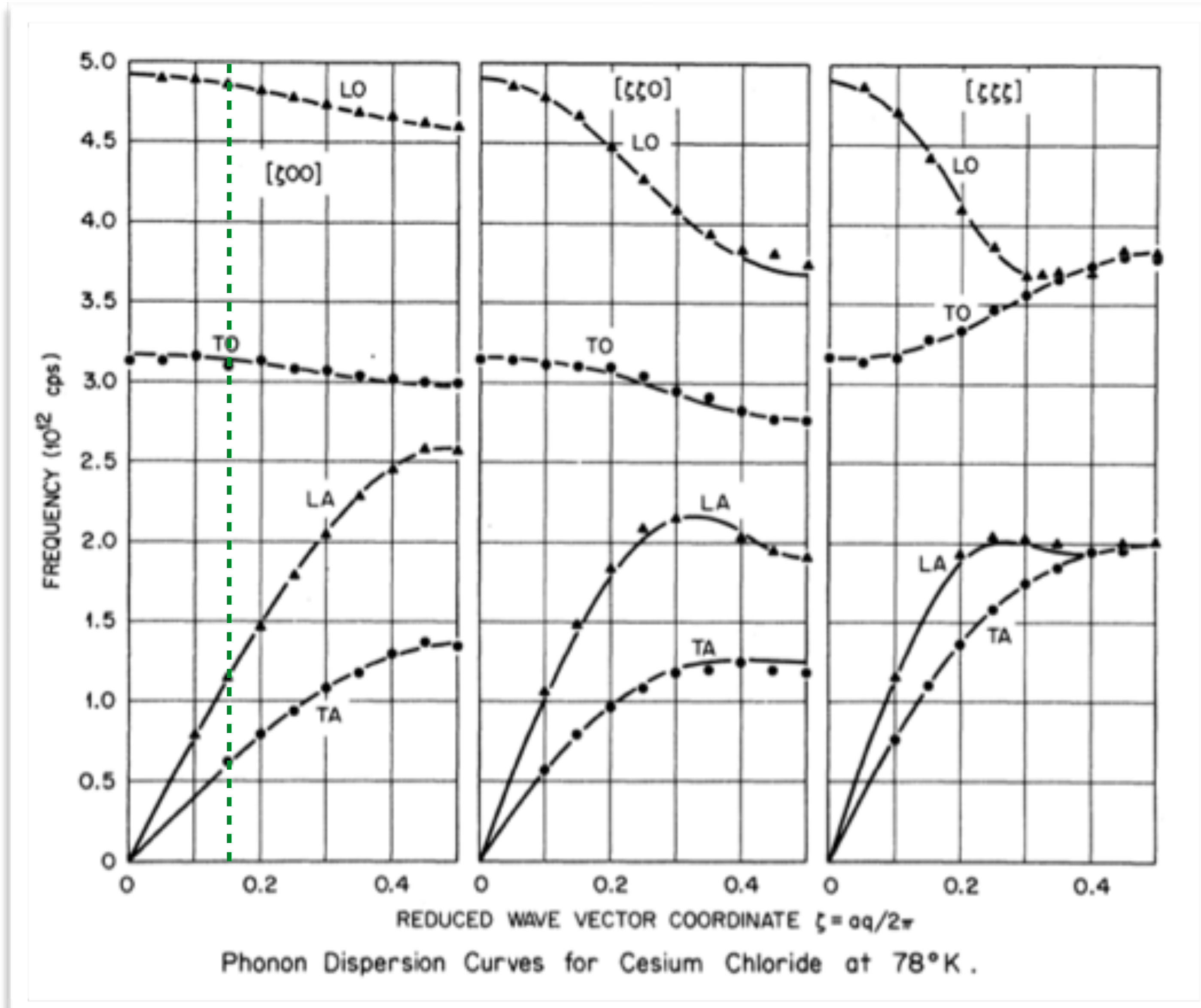
- Measurements done with a TAS at Oak Ridge

- Point by point measurement in (Q,ω) space
ω-scan at constant Q

- Only 4 branches are distinguishable
⇒ Transverse modes doubly degenerated

- Calculated frequencies based on the nature of the forces between atoms

1THz = (1/2π)10^{12} cps (counts s^{-1}) = 4.13 meV = 33.3 cm^{-1}



A. A. Z. Ahmad, H. G. Smith, N. Wakabayashi, and M. K. Wilkinson. *Phys. Rev. B* **6**, 3956 (1972)

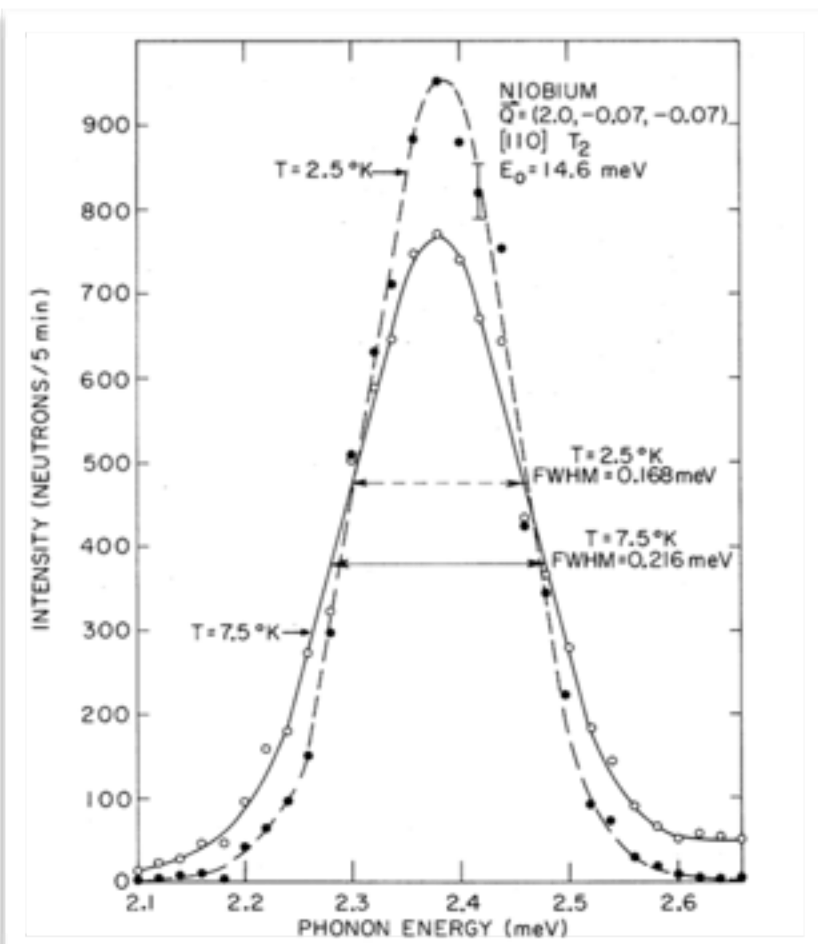
Phonon

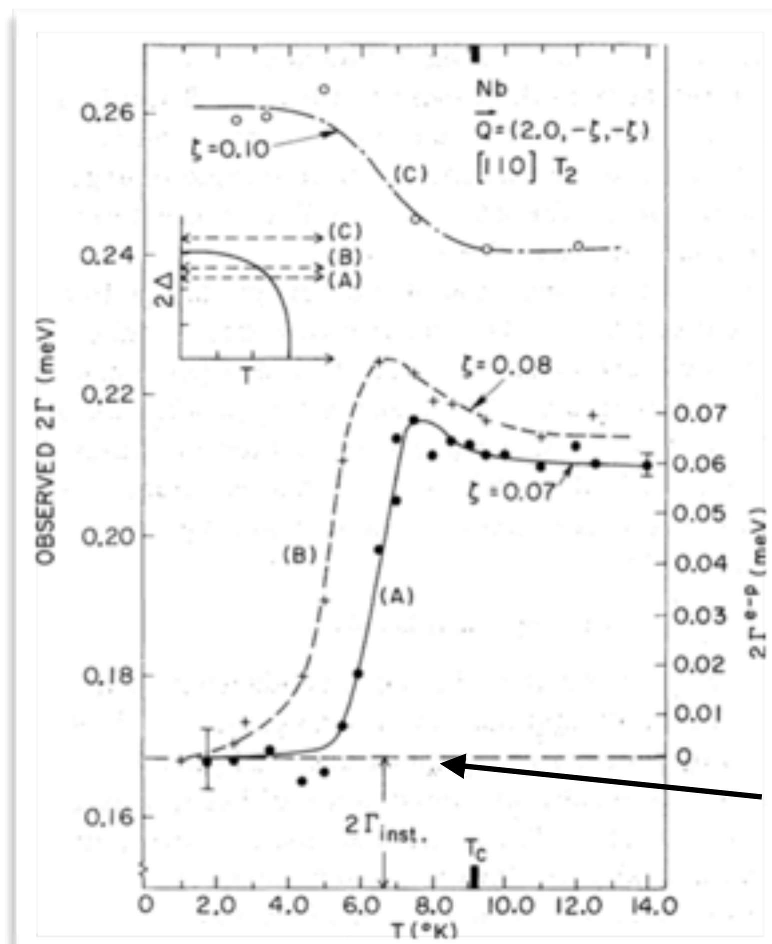
PHONON IN Nb: A CONVENTIONAL SUPERCONDUCTOR (T_C=9.2 K)

- cc single crystal
- In real systems, phonon–phonon and electron–phonon interactions tend to give single phonons a finite lifetime

⇒ Damped harmonic oscillator model

- Triple axis spectrometer at Brookhaven (ω-scan at constant Q)
- FWHM=2Γ ($\tau \propto \frac{1}{2\Gamma}$): the line width decreases below Tc (the lifetime increases)
- Resonant condition: $\hbar\omega_p = 2\Delta(T)$. (A) and (B): $\hbar\omega_p < 2\Delta(0)$, (C): $\hbar\omega_p > 2\Delta(0)$





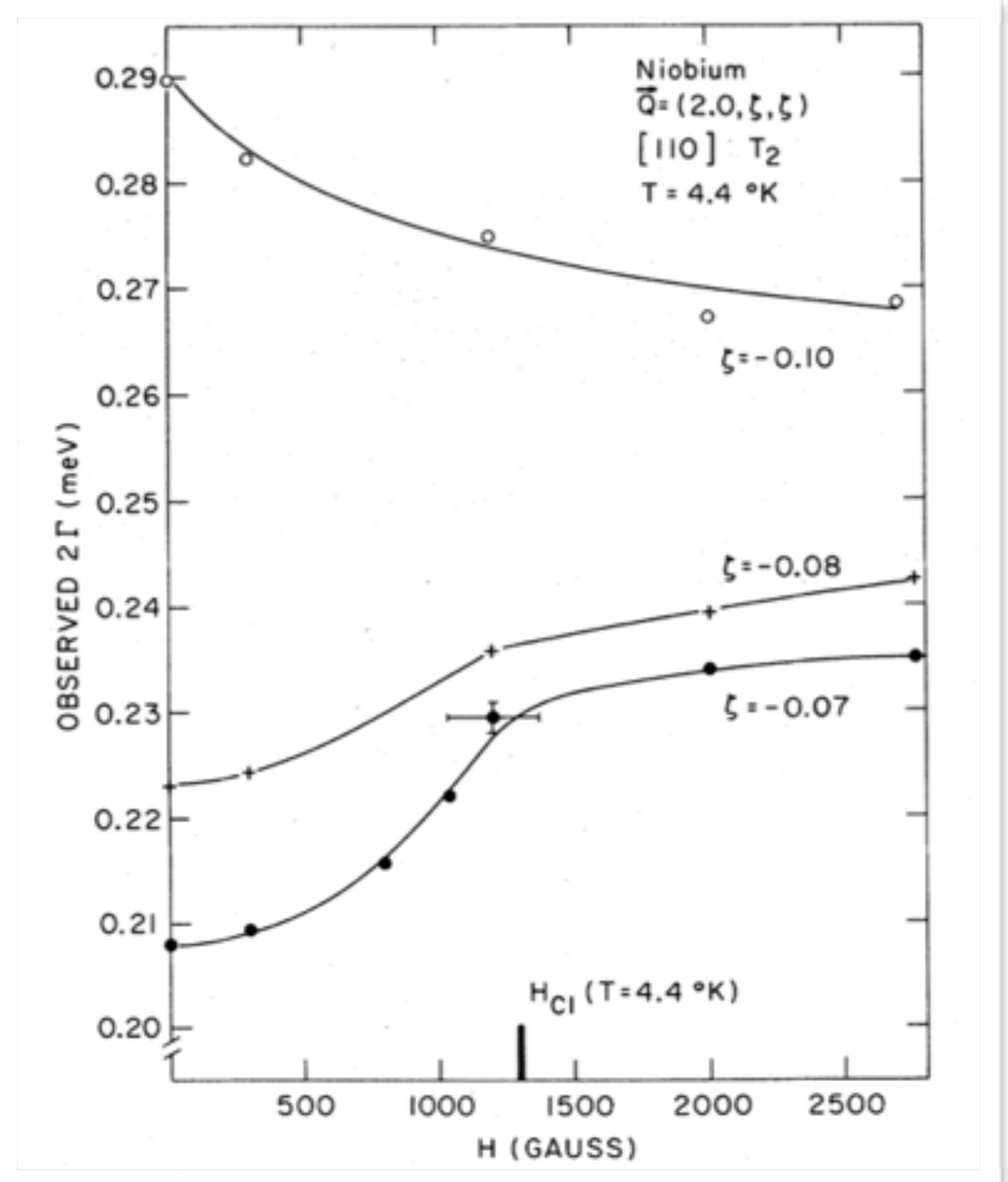
Line width of the instrumental resolution

S. M. Shapiro, G. Shirane, and J. D. Axe. *Phys. Rev. B* **12**, 4899 (1975)

Phonon

PHONON IN Nb: A CONVENTIONAL SUPERCONDUCTOR (T_C=9.2 K)

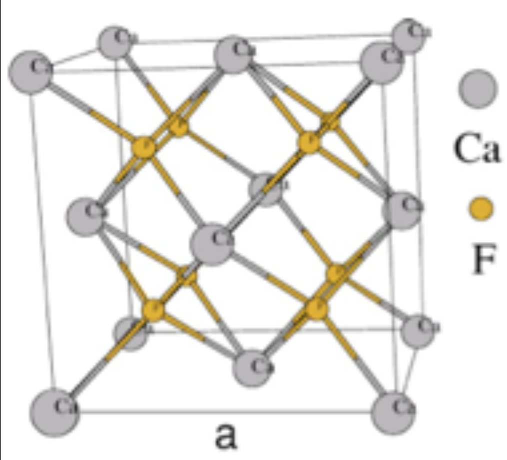
- H_{C1} = Critical field



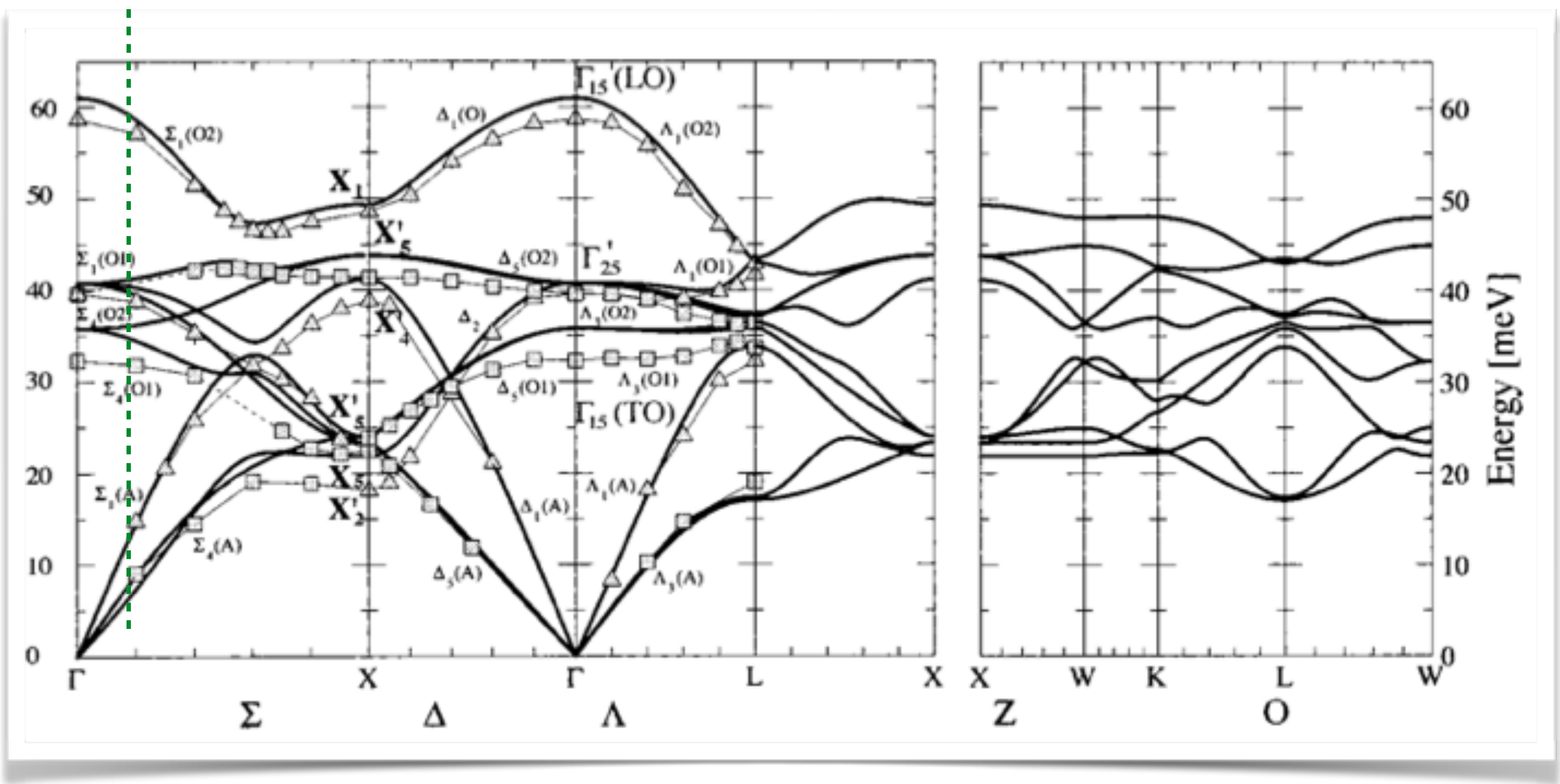
S. M. Shapiro, G. Shirane, and J. D. Axe. *Phys. Rev. B* **12**, 4899 (1975)

Phonon

PHONON IN CaF_2: A SUPER-IONIC CONDUCTOR



- fcc single crystal with 3 atoms in the unit cell ⇒ 3*3 = 9 branches
- IN3 (preliminary) and Lagrange-IN1 (mostly) TAS at ILL (ω-scan at constant Q at room temperature)
  Symmetry directions [001] (Δ), [111] (Λ), and [110] (Σ)
- As the process of ionic conduction involves hopping over potential barriers, ionic motion is an anharmonic process
- Along Σ, 3 branches are not seen because $\vec{Q} \perp \vec{e}_{ds}$
- Calculation of the dispersion of phonon frequencies by density-functional perturbation theory

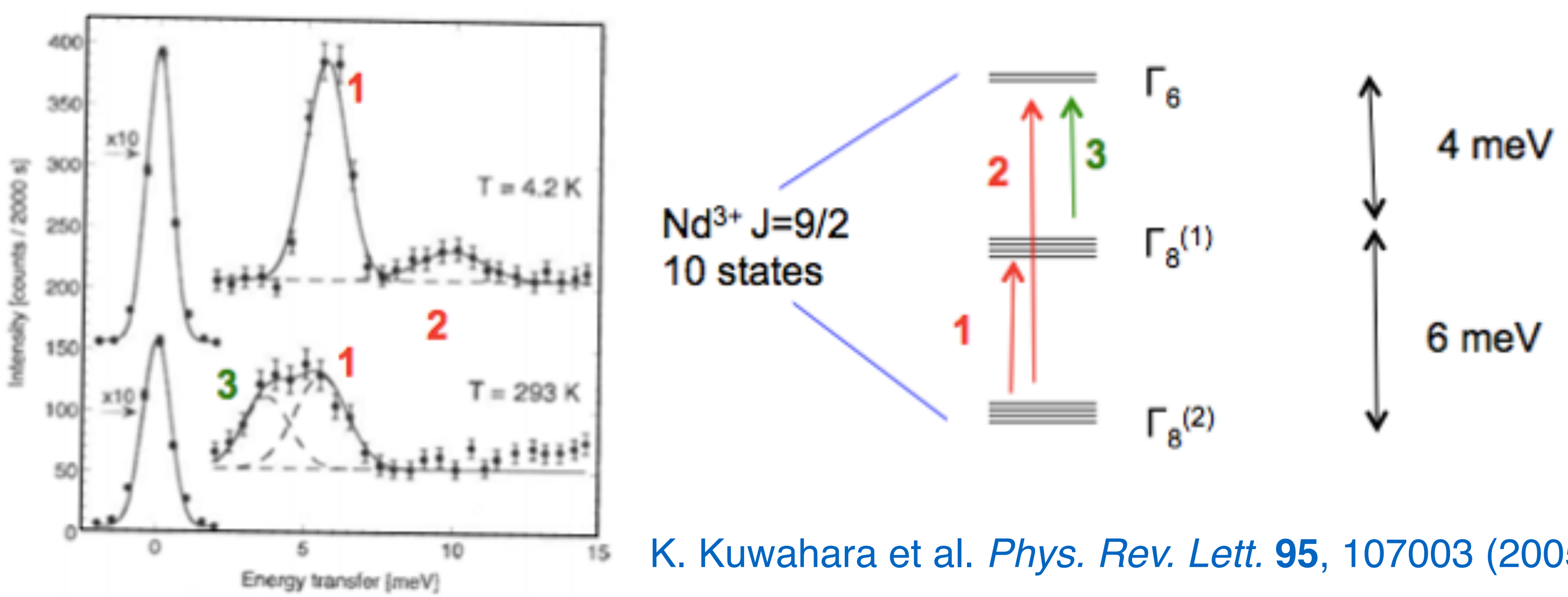


K. Schmalzl, D. Strauch, and H. Schober. *Phys. Rev. B* **68**, 144301 (2003)

Magnetic excitations

LOCAL EXCITATION

Crystal field transition

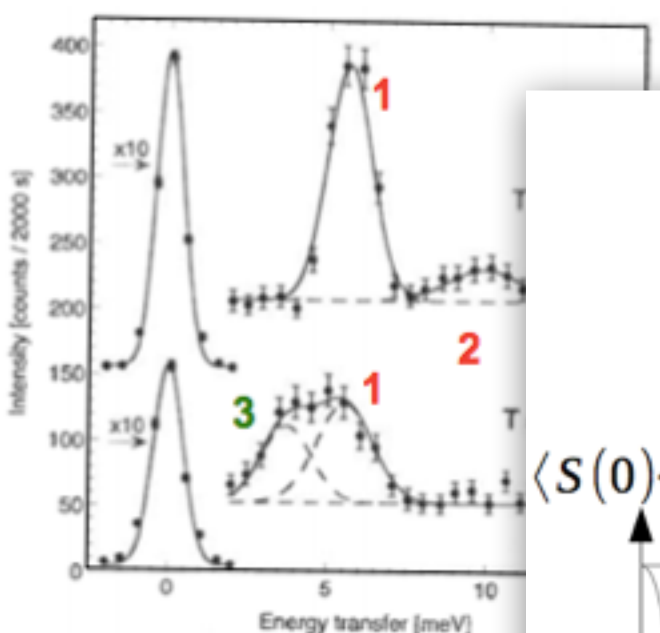


K. Kuwahara et al. *Phys. Rev. Lett.* **95**, 107003 (2005)

Magnetic excitations

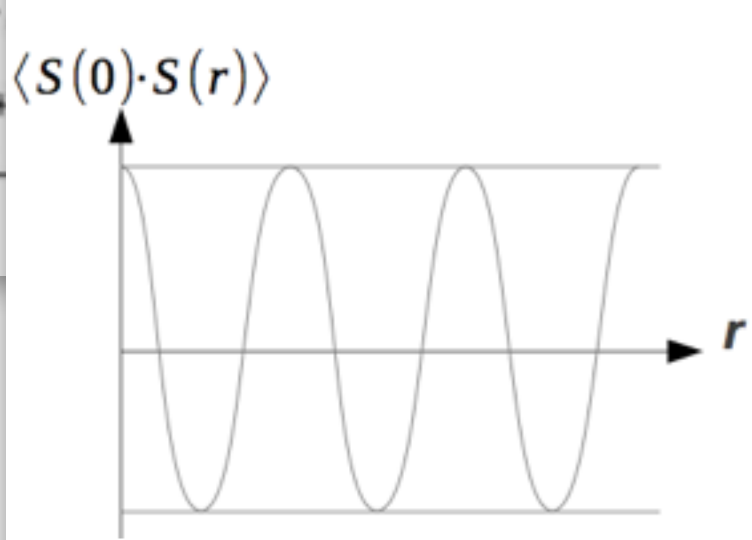
LOCAL EXCITATION

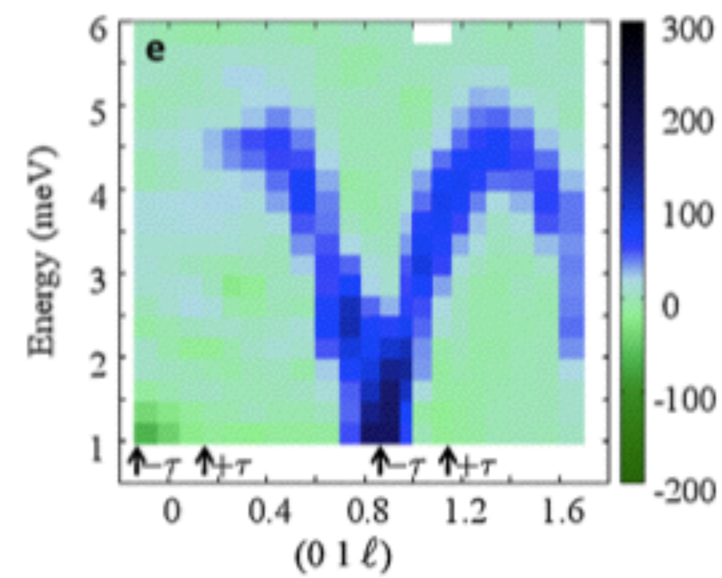
Crystal field transition



DISPERSIVE EXCITATION

Spin wave



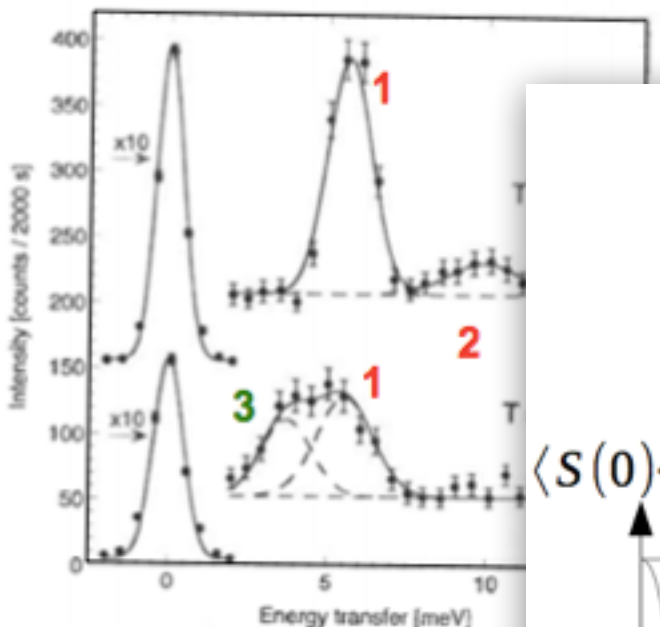


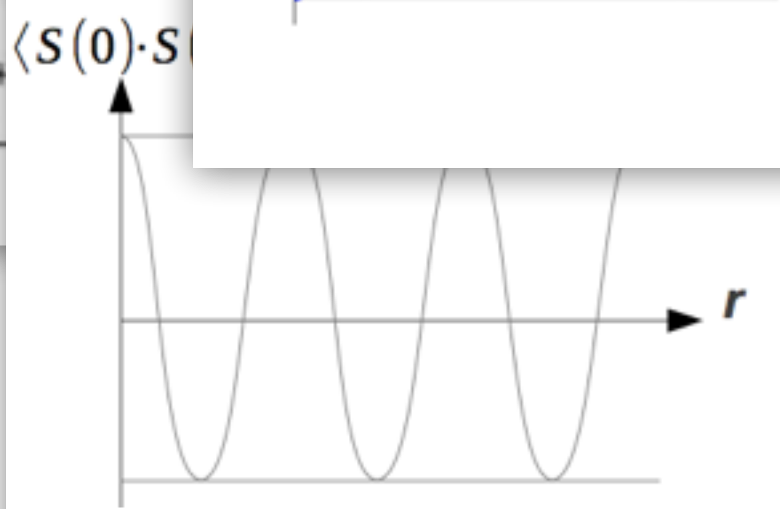
M. Loire et al. *PRL* **106**, 207201 (2011)

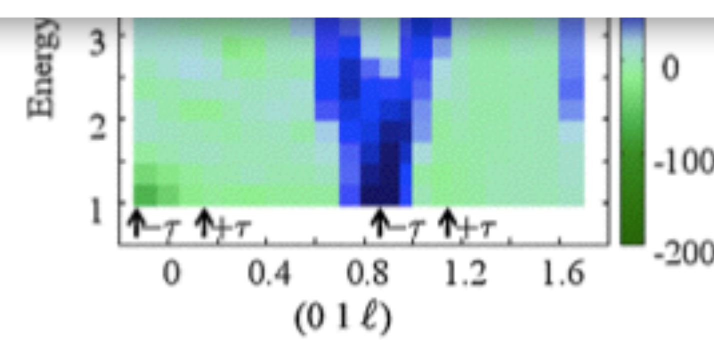
Magnetic excitations

LOCAL EXCITATION

Crystal field transition

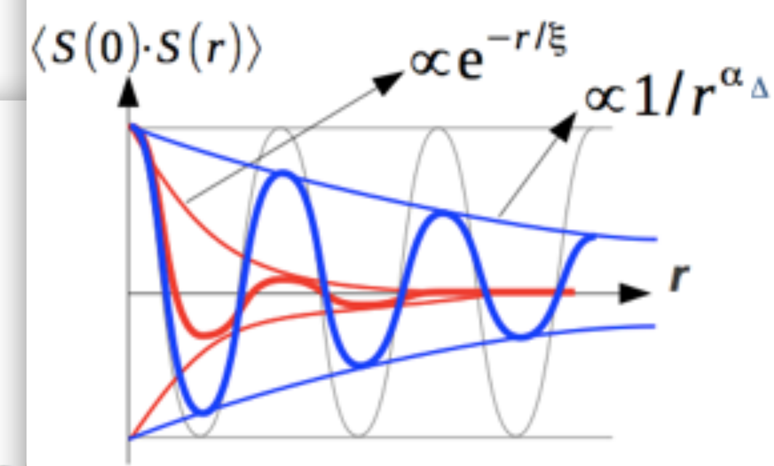




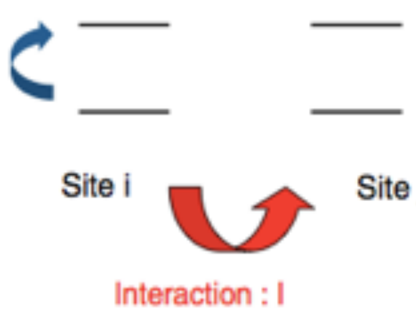


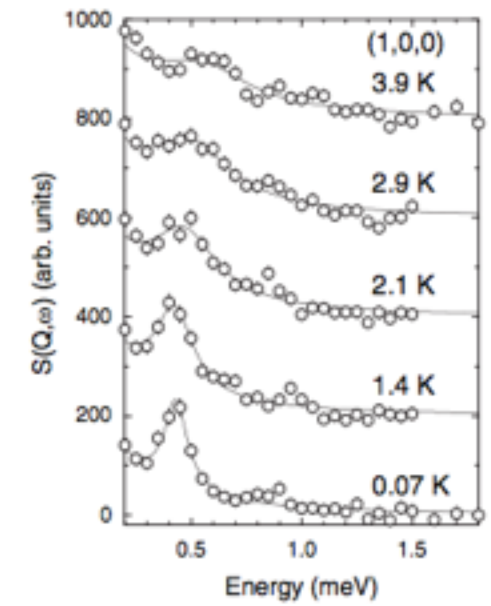
M. Loire et al. *PRL* **106**, 207201 (2011)

DIFFUSE SCATTERING



Crystal field exciton



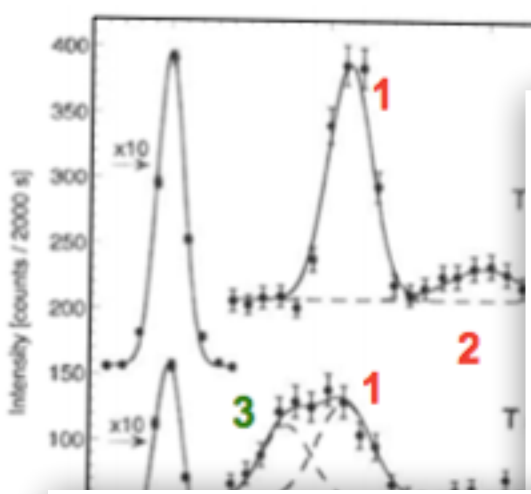


K. Kuwahara et al. *PRL* **95, 107003** (2005)

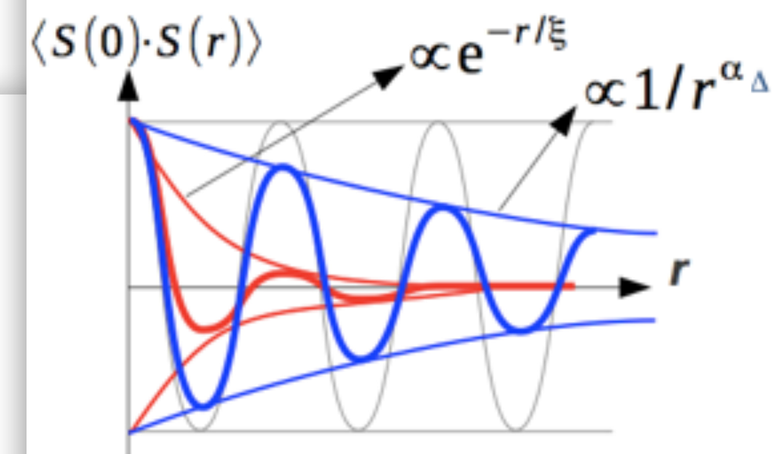
Magnetic excitations

LOCAL EXCITATION

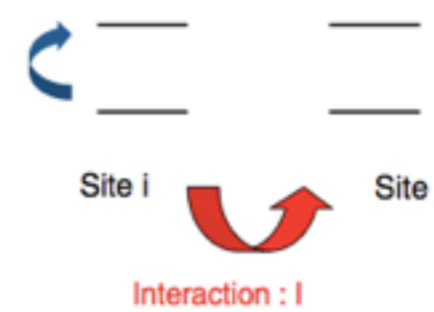
Crystal field transition

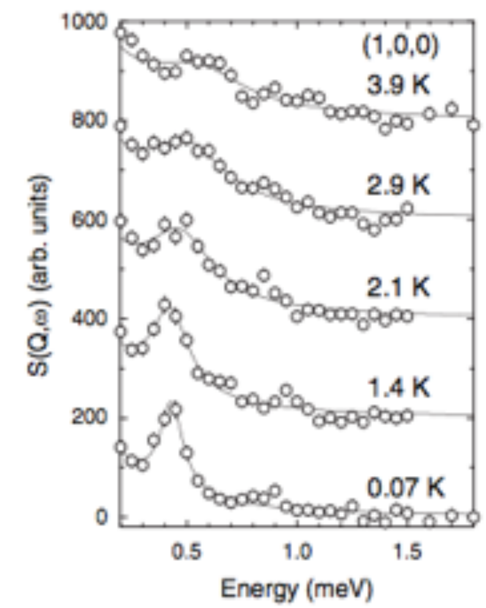


DIFFUSE SCATTERING

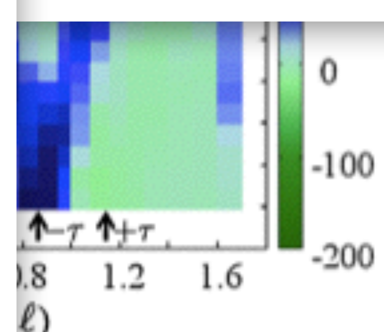


Crystal field exciton



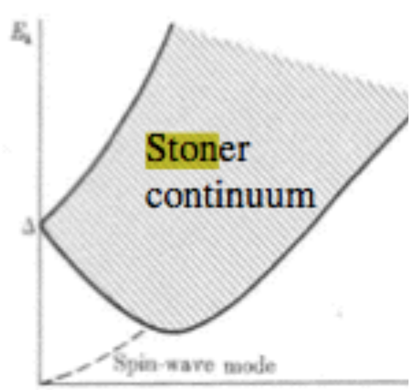


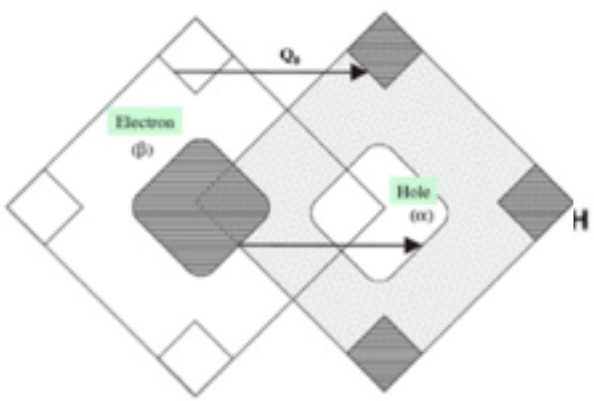
K. Kuwahara et al. *PRL* **95, 107003** (2005)

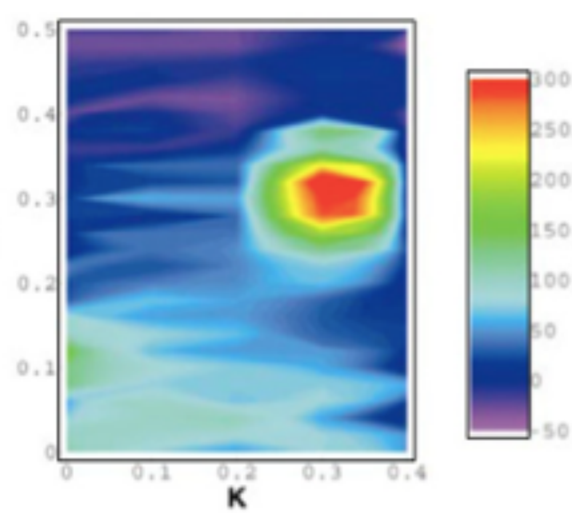


6, 207201 (2011)

EXCITATIONS IN ITINERANT ELECTRON SYSTEMS

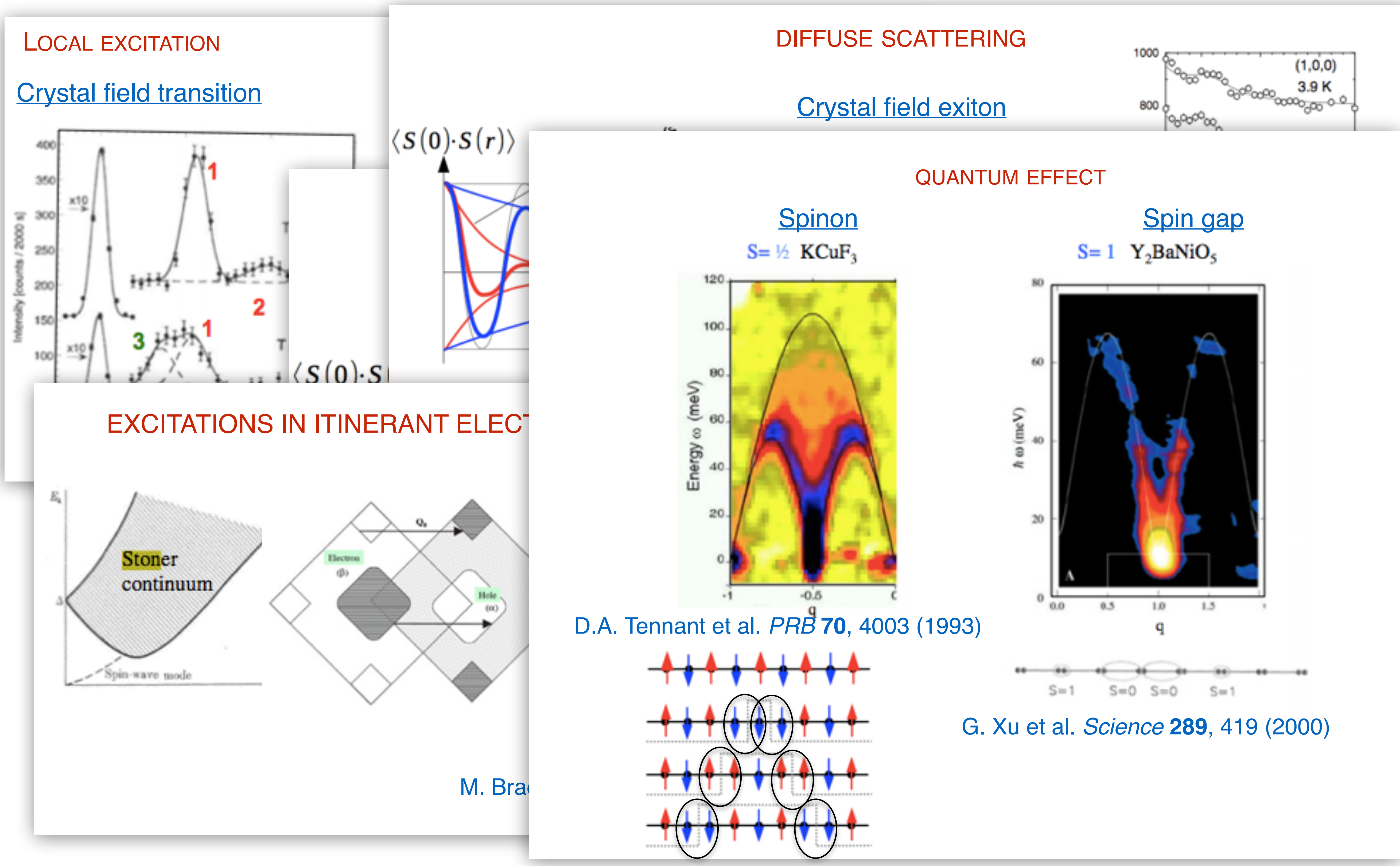






M. Braden et al. *PRB* **66**, 064522 (2002)

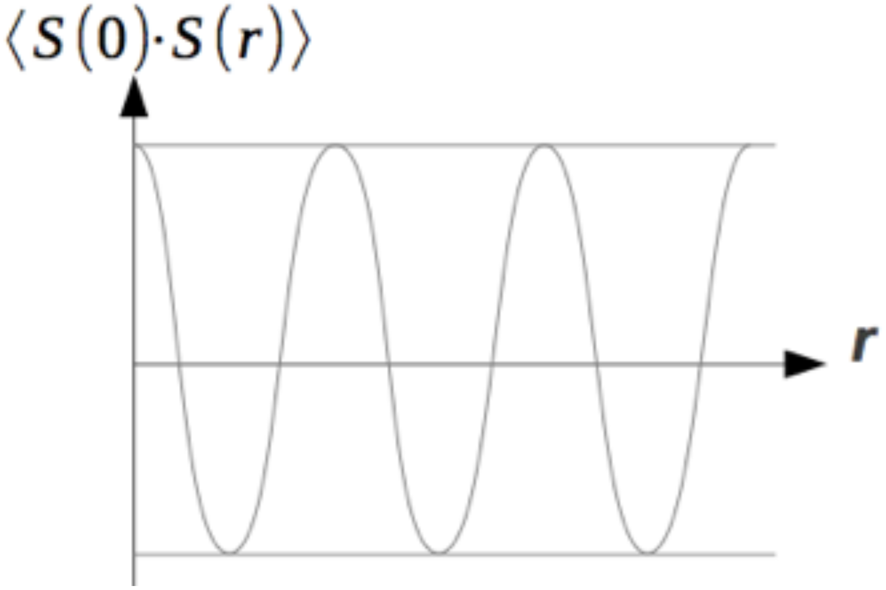
Magnetic excitations

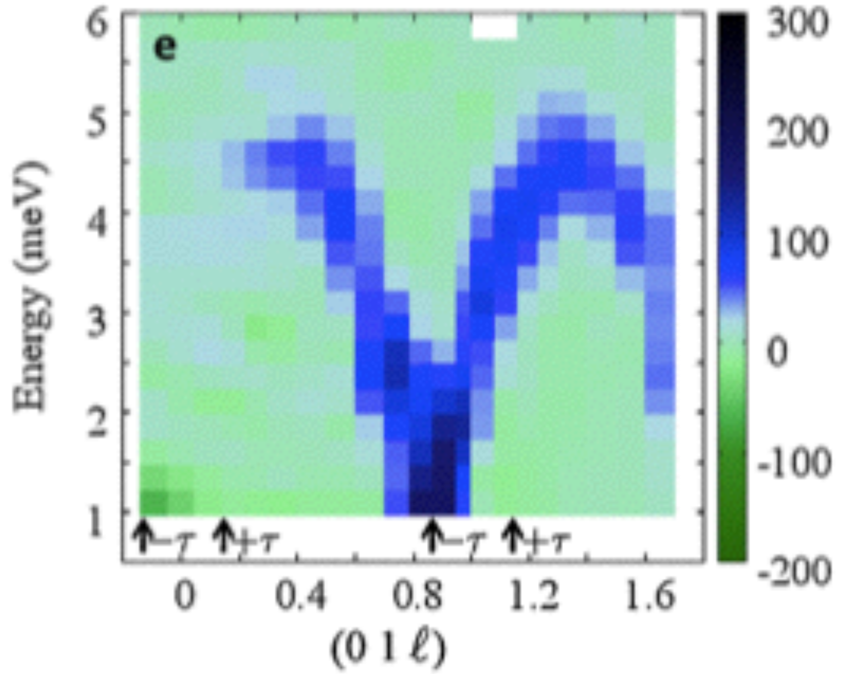


Magnon

DISPERSIVE EXCITATION

Spin wave

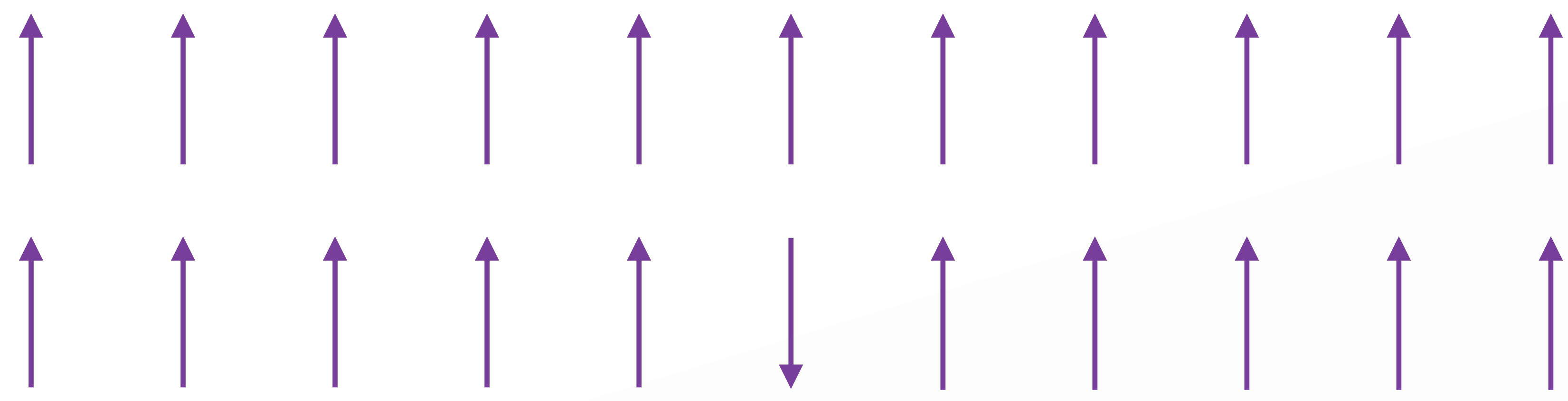




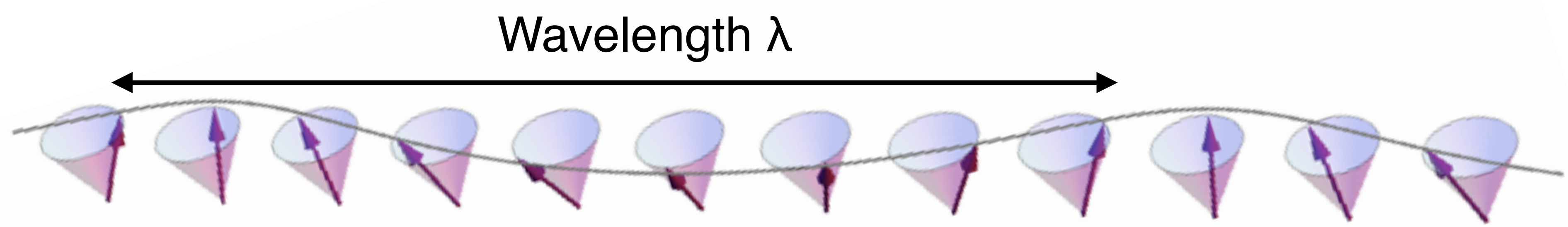
M. Loire et al. *PRL* **106**, 207201 (2011)

Magnon

Ferromagnetic Ground state



First stable excited state



Spin waves = Periodic precessions of the spins with a characteristic wavelength

Quantum description

Like sound waves, spin waves can only take energies in discrete quanta of energy.
This energy is carried by quasi particles called magnons

Magnon = quasi particle with an energy (frequency ω) and a momentum (wave-vector q)

$$E_q = \left(\langle n_q \rangle + \frac{1}{2}\right)\hbar\omega_q$$

Magnon

CLASSICAL DESCRIPTION

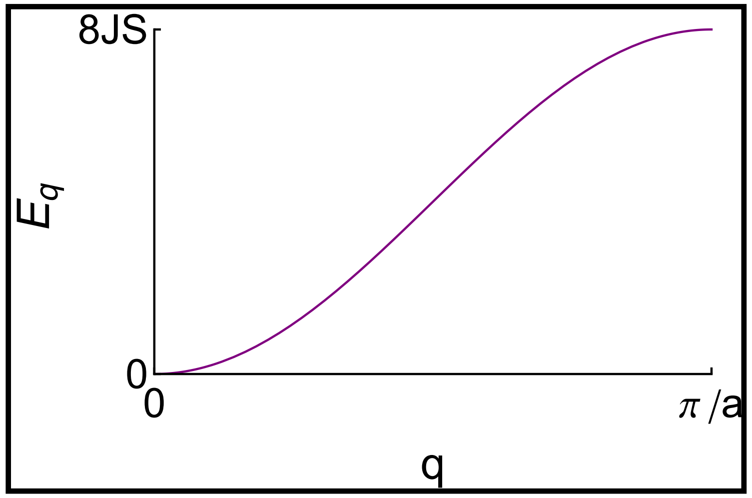
HYPOTHESIS
Small oscillations
Interaction between nearest neighbors

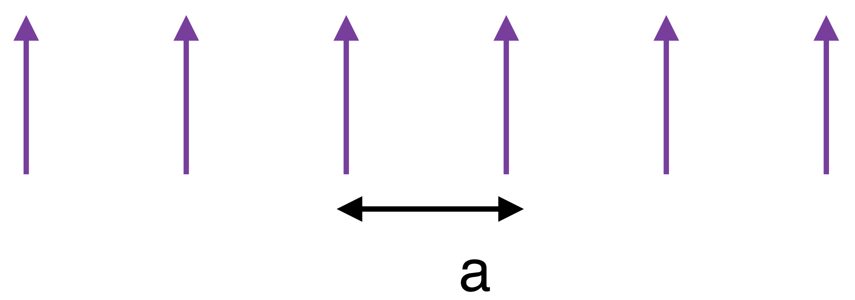
Heisenberg Hamiltonian

$$H = -2J\vec{S}_j(\vec{S}_{j-1} + \vec{S}_{j+1})$$

ONE DIMENSIONAL LINEAR FERROMAGNETIC CHAIN

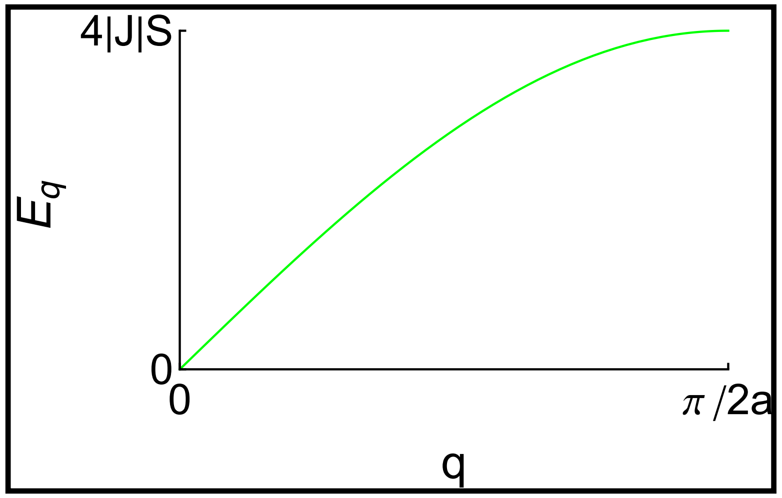
- J>0
- $E_q = \hbar\omega_q = 4JS(1 - cos(qa))$
- For qa <<1, $E_q=2JSq^2a^2=Dq^2$

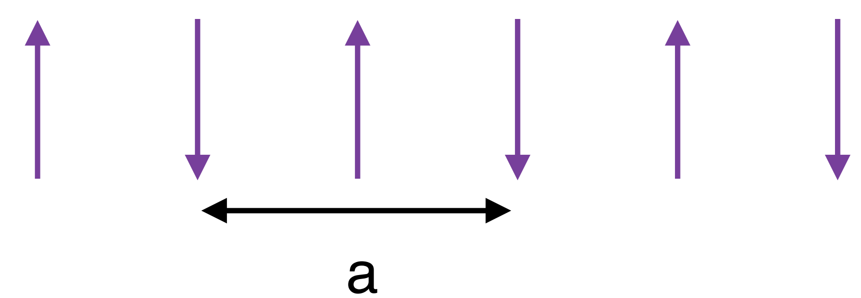




ONE DIMENSIONAL LINEAR ANTIFERROMAGNETIC CHAIN

- J<0
- $E_q = \hbar\omega_q = -4JS|sin(qa)|$
- For qa <<1, $E_q=-4JSaq=Dq$





Rq: Flip of one spin costs an energy $4JS^2$

Magnon

COHERENT MAGNETIC DIFFERENTIAL SCATTERING CROSS SECTION

$$\left(\frac{d^2\sigma}{d\Omega dE_f}\right) = (2p)^2 \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} dt\, exp(-i\omega t)\langle \vec{D}_\perp(-\vec{Q},0)\cdot\vec{D}_\perp(\vec{Q},t)\rangle_T$$

S($\vec{Q}$,ω)

1 MAGNON DIFFERENTIAL SCATTERING CROSS SECTION

1 magnetic atom in magnetic unit cell, all magnetic moments aligned along the z-axis

$$\left(\frac{d^2\sigma}{d\Omega dE_f}\right)_{\pm mag} = (2p)^2 \frac{k_f}{k_i}\frac{(2\pi)^3}{V}\frac{1}{2}S(1+\hat{Q}_z^2)\left(\frac{1}{2}gf(\vec{Q})\right)^2 e^{-2W}\sum_{\vec{\tau},\vec{q}} < n_\pm >_T \delta(\omega \mp \omega_s)\delta(\vec{Q}\mp\vec{q}-\vec{\tau})$$

$p = 0.2695 \times 10^{-12}\ cm$

S: Spin amplitude of the atom

Intensity amplitude: $(2p)^2S$

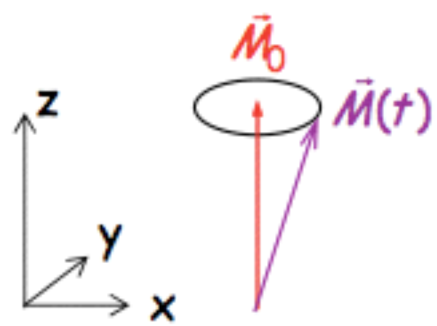
$1+\hat{Q}_z^2\left(\hat{Q}_z = \frac{\vec{Q}\cdot\vec{z}}{|\vec{Q}|}\right)$: polarisation factor. Only the fluctuations perpendicular to $\vec{Q}$ are observed

Moments // z: fluctuations // to x and y

$\vec{Q}$ // z: $1+\hat{Q}_z^2$ = 2 → we see the fluctuations // to x and y

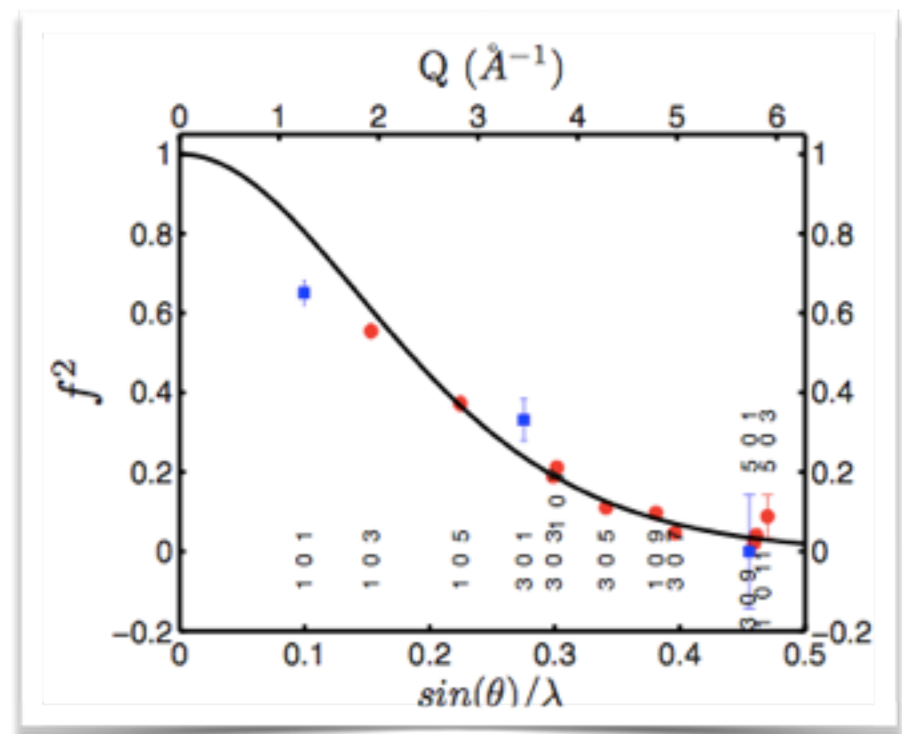
$\vec{Q}$ // x: $1+\hat{Q}_z^2$ = 1 → we see the fluctuations // to y

$\vec{Q}$ // y: $1+\hat{Q}_z^2$ = 1 → we see the fluctuations // to x



f($\vec{Q}$): Magnetic form factor |f($\vec{Q}$)|↘ when Q↗

g: Lande factor



f(Q)² obtained by diffraction on $SrFe_2As_2$ (Iron based system)

Comparison Phonon / Magnon

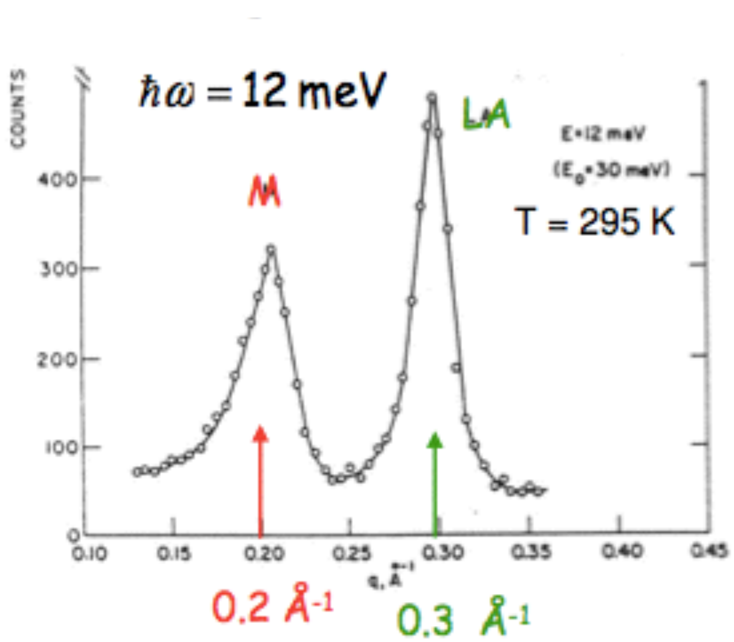
How distinguish the nuclear and magnetic intensities in the inelastic neutron scattering spectrum with unpolarized neutrons ?

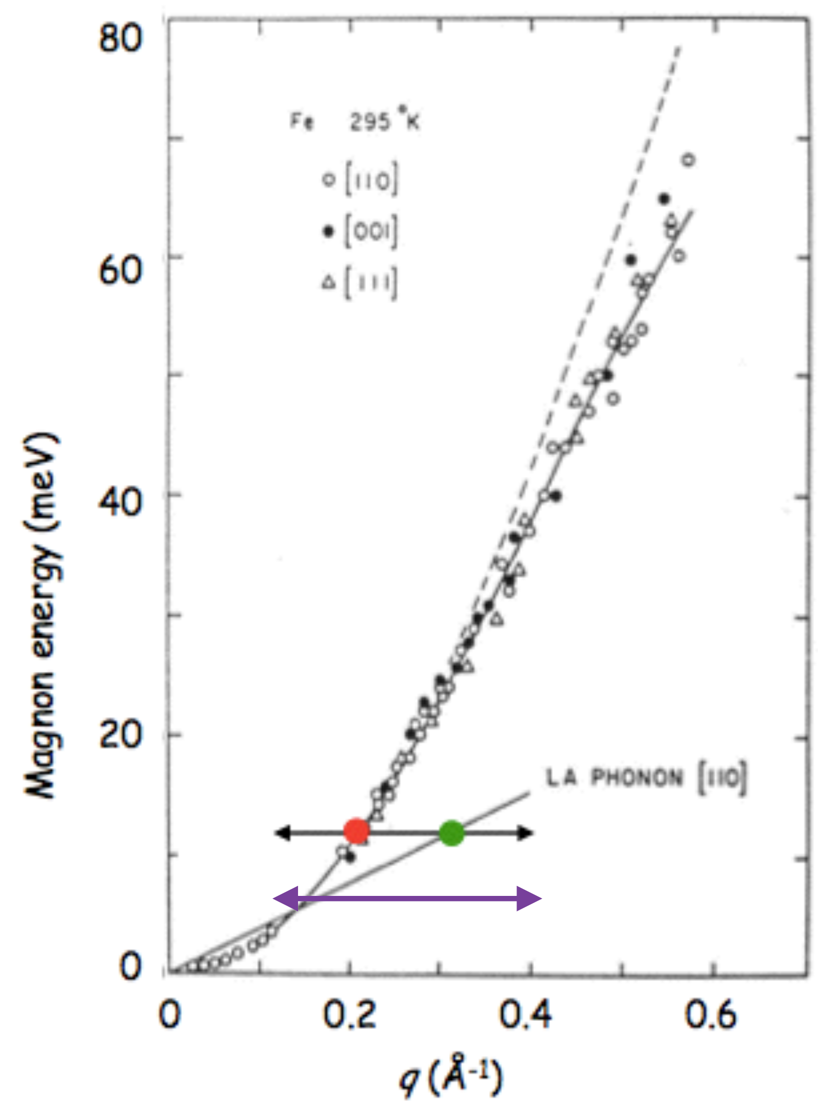
	Phonons	Magnons
1. Q dependence	$I^{ph} \propto Q^2$	$I^{magnon} \propto f(Q)^2 \searrow$ when $Q \nearrow$
2. Polarisation factor	$I^{ph} \propto (\vec{Q} \cdot \vec{e}_{ds})^2$ We see lattice vibrations // $\vec{Q}$	$I^{magnon} \propto \left(1 + \frac{Q_z^2}{\|\vec{Q}\|^2}\right)$ We see spin fluctuations $\perp \vec{Q}$
3. Temperature dependence	$\forall$ T, I^{ph} increases with T	$I^{magnon} = 0$ for $T>T_c$
4. Intensity Amplitude	$I^{ph} \propto b^2$	$I^{magnon} \propto (2p)^2S$
5. Number of branches	n atoms in unit cell → 3n phonon branches	n magnetic atoms in magnetic unit cell → n magnons branches in simple cases

Magnon

MAGNONS IN IRON: A 3 DIMENSIONAL METAL

- cc single crystal
- Triple axis spectrometer at the Brookhaven High Flux Beam Reactor - T = 295 K (Tc ≃ 500 K)
- Q-scan at constant ω
- Ferromagnetic: For qa <<1, $E_q = 2JSa^2q^2 = Dq^2 \rightarrow D = 281$ meV Å^2



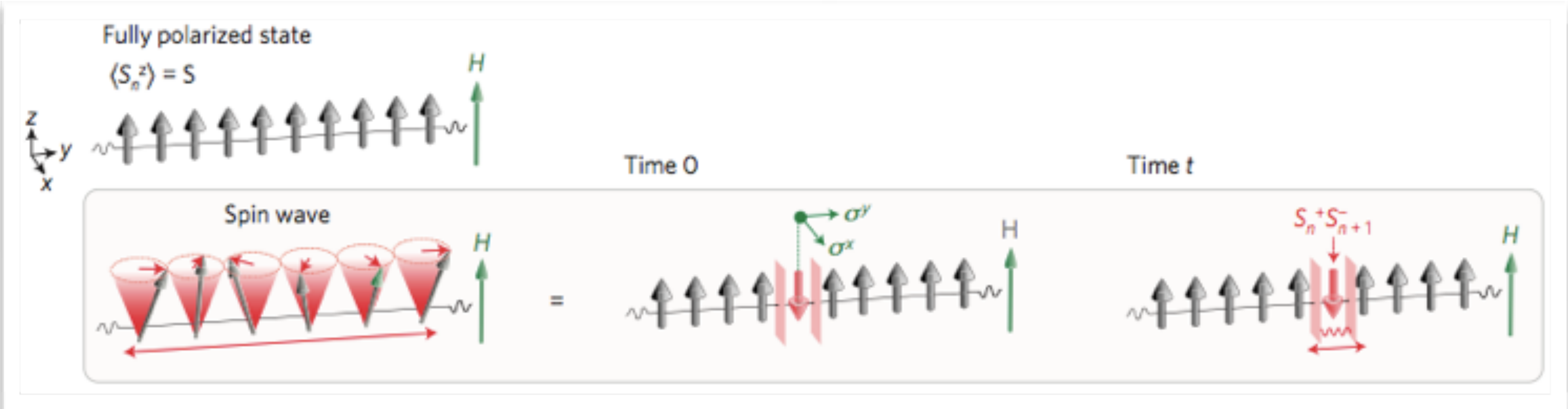


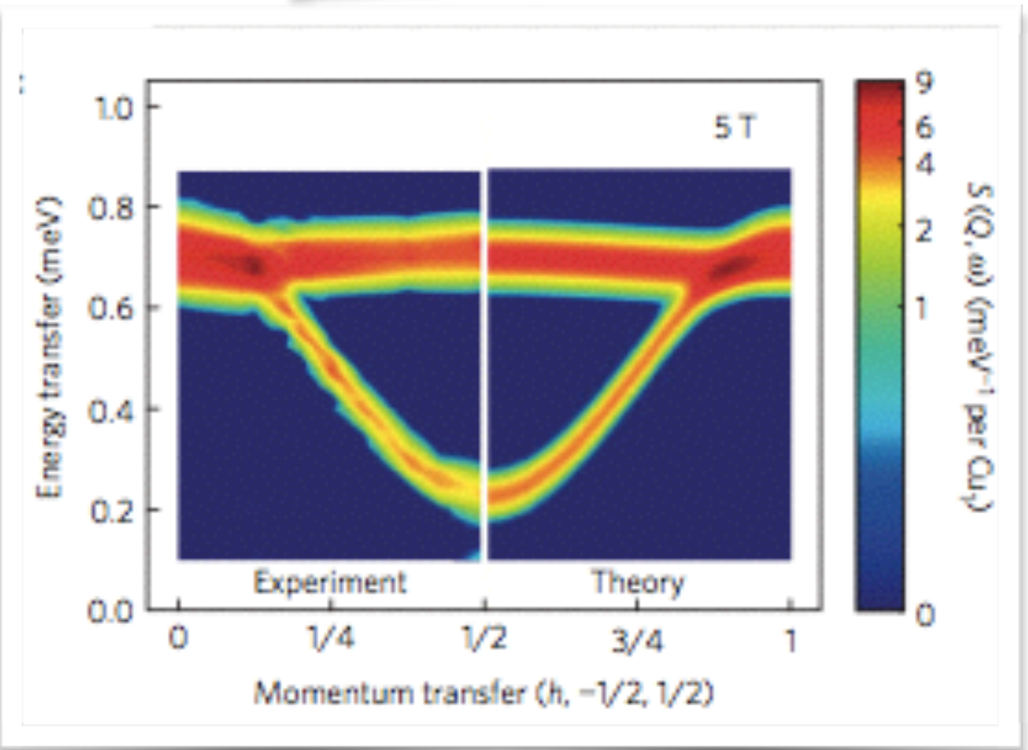
G. Shirane et al. Spin Waves in 3d Metals. *J. Appl. Phys.* **39**, 383 (1968)

Magnon

MAGNONS IN $CuSO_4.5D_2O$: A SPIN-1/2 HEISENBERG ANTIFERROMAGNETIC CHAIN COMPOUND

- Deuterated single crystal
- IN14 cold TAS at ILL (T = 100mK) T just above the Néel transition temperature to three-dimensional antiferromagnetic ordering
- High-field fully polarized state ($\mu_0 H = 5T > \mu_0 H_{sat}$)





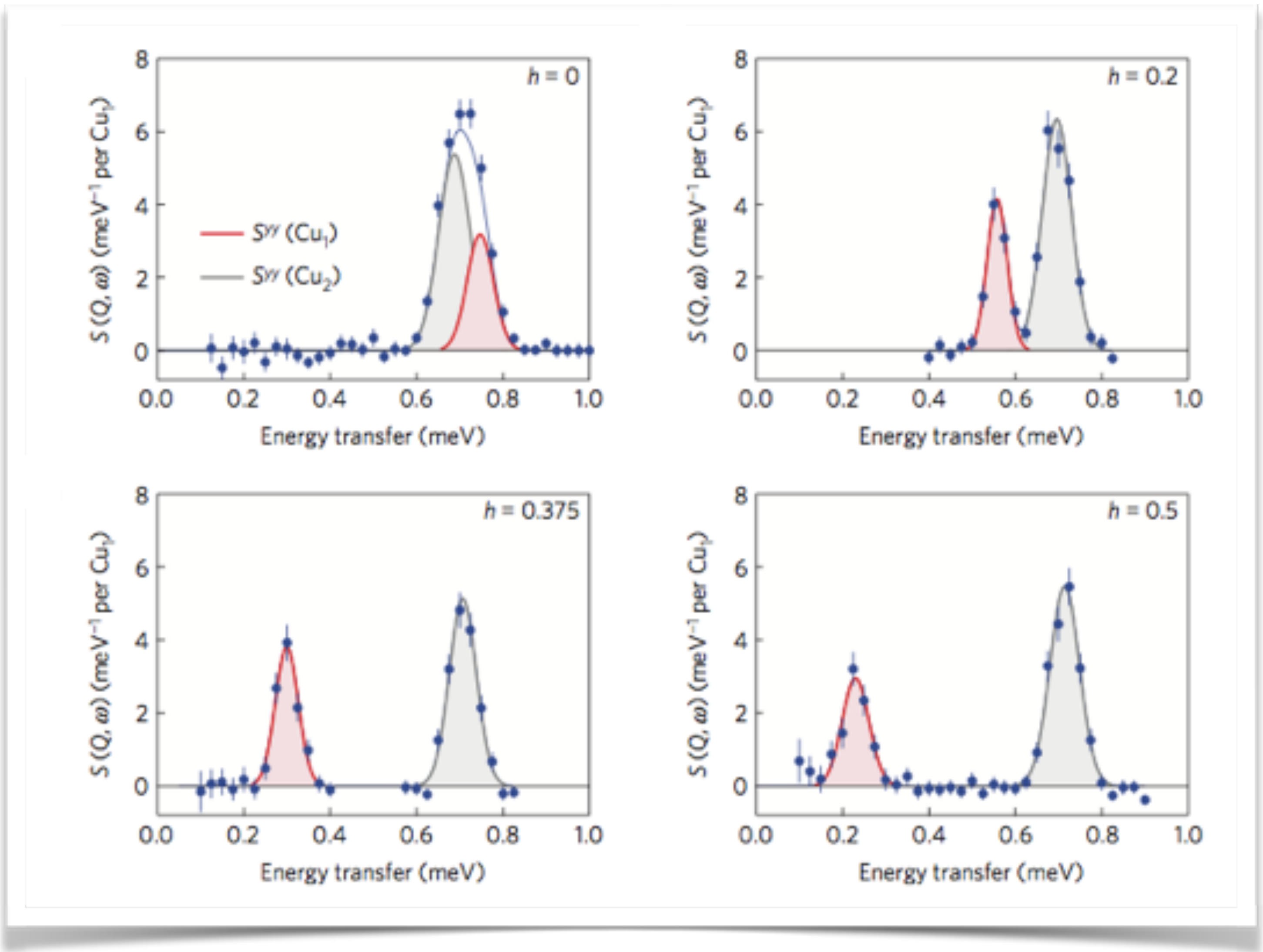
- Intensity color maps:
  - The cosine-shaped dispersion corresponds to Cu_1 chains
  - The flat branch corresponds to the decoupled Cu_2 sites

M. Mourigal et al. *Nature Physics* **9**, 435–441 (2013)

Magnon

MAGNONS IN A $CuSO_4.5D_2O$, A SPIN-1/2 HEISENBERG ANTIFERROMAGNETIC CHAIN COMPOUND

- Selected energy scans at constant wave vector (h,-1/2,-1/2)
- Spin-wave fit - theoretical magnon intensity: $S^{xx}(\mathbf{Q},\omega) = S^{yy}(\mathbf{Q},\omega) = (S/2)\delta(\omega-\omega(h))$ per Cu_1



M. Mourigal et al. *Nature Physics* **9**, 435–441 (2013)

Parasitic peaks

- Bragg peaks from sample holder, cryostat
- Incoherent scattering from monochromator, analyzer
  higher-order reflections
- Beam on detector
- Phonons from monochromator, analyzer

  → temperature evolution
  → sample angle scans

Advantages x disadvantages of TAS

- intensity focused on specific $(\vec{Q}, \hbar\omega)$ point
- measurements along any trajectory in scattering plane
- constant-***Q or –E scans,*** *depending on type of excitation*
- focusing and other 'tricks' to improve the signal/noise
- polarization analysis to separate electronic (magnetic) and phonon signals can be used – spin flip x no spin flip

- in principle less efficient to cover large region of $(\vec{Q}, \hbar\omega)$ space; solution – multiplex detectors
- possible contamination of parasitic reflections

Measurement on sample - preconditions

- single crystal – sufficient size, one grain crystal

  → Laue neutron diffraction, e.g. OrientExpress

- crystal structure

- basic physical properties

- magnetic structure – at least propagation vector

  → Laue diffraction (CYCLOPS), powder diffraction

- energy scale – roughly estimated

  → ToF, susceptibility

- aligned single crystal

Conclusion

- Inelastic neutron scattering: ideal probe for phonon and magnon investigation.
- 2 approaches - 2 types of instruments: TAS and TOF spectrometer
- Be careful: before an experiment you have to think if you need a TAS or a ToF spectrometer

Advantages / Disadvantages	TOF	TAS
Sample	Powder (possibly sufficiently large single crystal) Liquid, Amorphous	Crystal (sometimes powder)
Dynamic Structure factor	S(Q,ω)	$S(\vec{Q},\omega)$
Detector	Multidetector	Monodetector
Resolution	ΔE/E ≃ 1 %	ΔE/E ≃ 5 %
Typical physics	Dynamic in a liquid, amorphous Localized excitations Individual excitations 1D magnon	Collective excitations: -phonons -magnons
Particular use	To make a map to know where to go in the (Q,ω) space with a TAS	To see the dependence of an excitation in temperature, magnetic field…

Main references

- Collection SFN
  - Les excitations dans la matière condensée: vibrations et phonons. H. Schober and S. Rols
  - Diffusion des neutrons par la matières cristalline ou amorphe non magnétique. H. Schober
  - Magnetic excitations. S. Raymond
- Books
  - Neutron Scattering with a Triple-Axis Spectrometer. G. Shirane, S. M. Shapiro and J. M. Tranquada
  - Introduction to the theory of thermal neutron scattering. G. L. Squires

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