Instrument Review: D4

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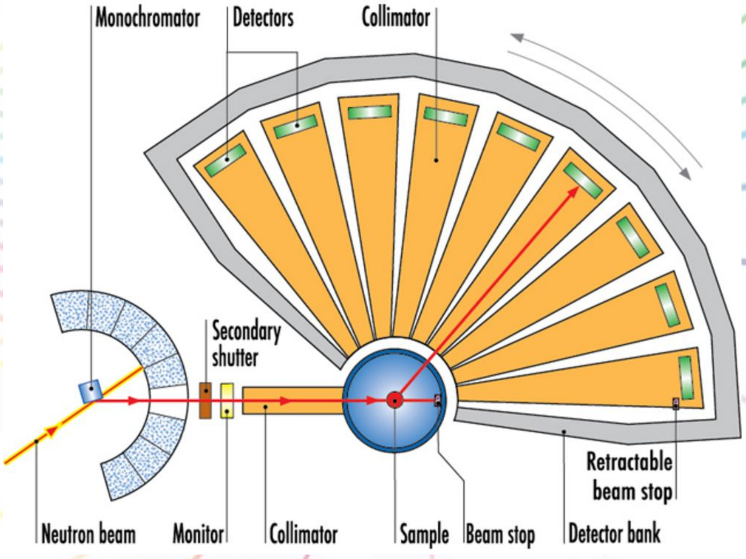
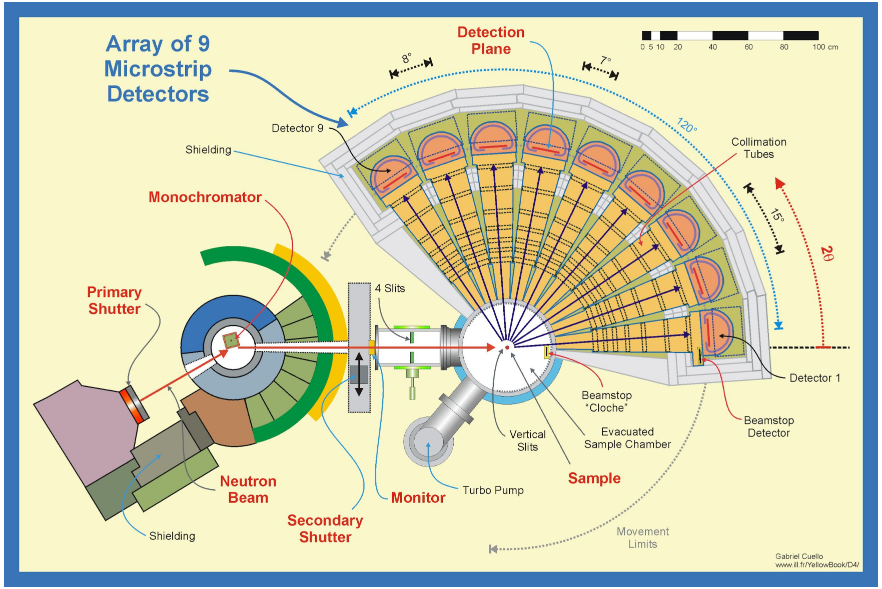
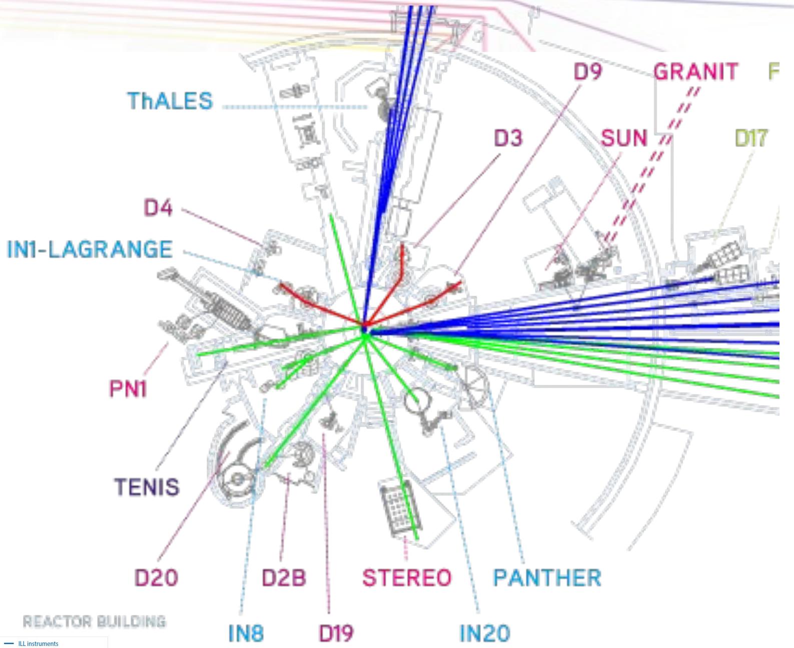
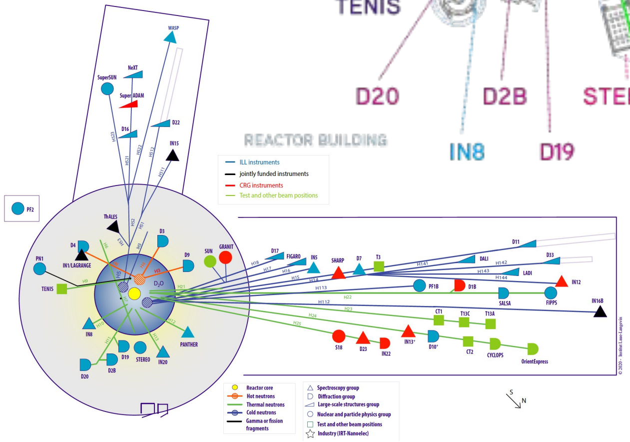


The instrument

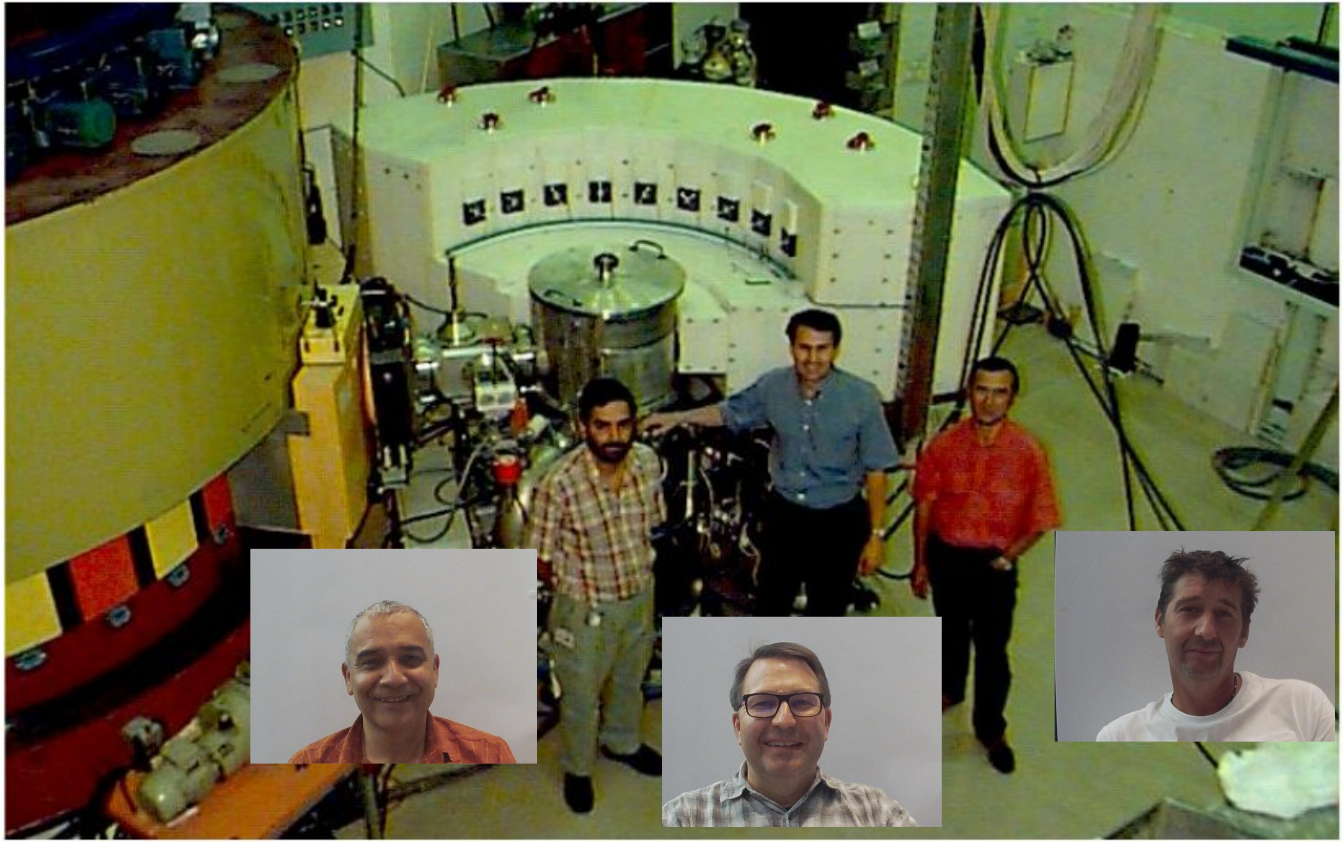
Level C / H8 - Hot neutrons
Time sharing with IN1-Lagrange







The team



Some highlight characteristics

D4 is an evolving instrument

D4 (≥ 1973) ➪ D4B (≥ 1983) ➪ D4C (≥ 2000) ➪ D4D (≥ 2030?)

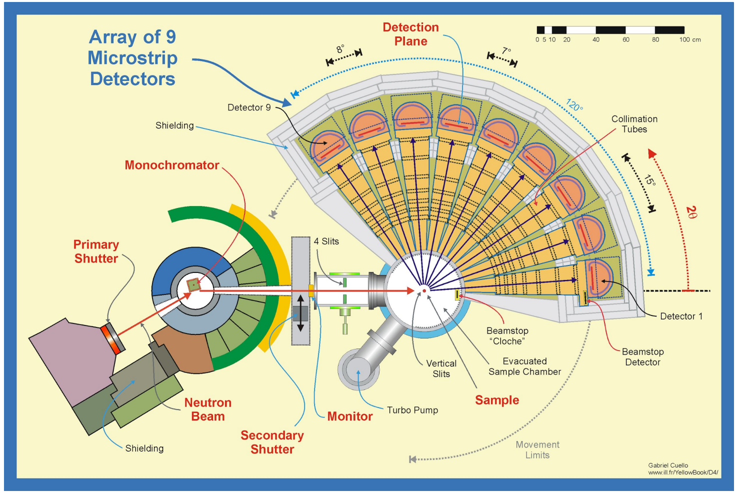
- Hot source : $0.3 < \boldsymbol{\lambda}/\text{Å} < 1.05$, standard $\boldsymbol{\lambda} = 0.5$ Å for $Q_{max} = 23.5$ Å^{-1}.
- Counting rate 10^6 (*i.e.* 0.1 % stats) per 0.125 in 3 hours.
- Overall counting-rate stability 10^{-4} over 3 days.
- Low background without parasitic peaks for all $\boldsymbol{\lambda}$ and detector positions.
  $\Rightarrow$ Champion of experiments involving low-contrast ($\Delta b < 0.5$ fm) isotope substitution and/or very small samples.

- Q-resolution: $\Delta Q/Q \lesssim 0.025$ for $2\theta > 15°$ $\Rightarrow$ $R_{FWHM} = 60$ Å.
- R-resolution: for $\boldsymbol{\lambda} = 0.5$ Å, $\Delta R = 3.79/(Q_{max} = 23.5$ Å$^{-1}) = 0.16$ Å.
  $\Rightarrow$ Confirmed performance in PDF-analysis.

The monochromator

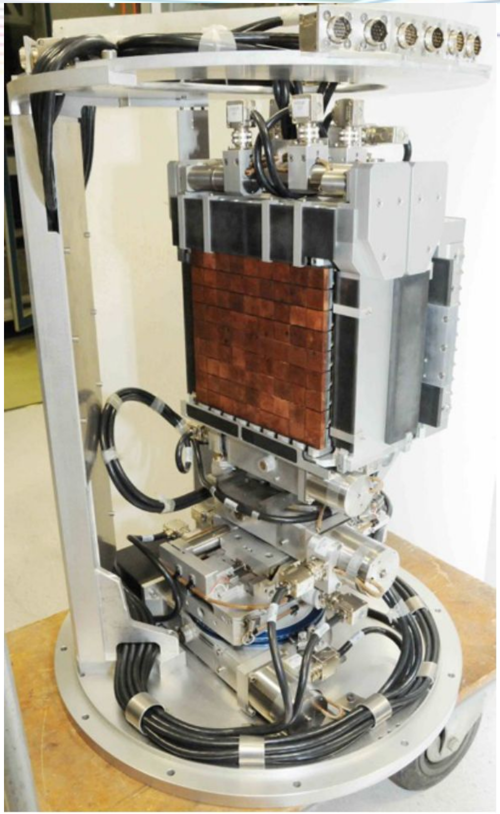
Large double-focussing multi-face crystal monochromator supplying monochromatic beam in the intermediate and high neutron energy ranges (Cu) as well as in the lower energy range (Si), in operation since 2009.

D4 uses only the Cu faces for the typical wavelengths: 0.7 Å, 0.5 Å and 0.35 Å.



Collimator α_1 → Vacuum tube → Shutter → Filter → Incident beam monitor → Diaphragm tube with horizontal+vertical slits.

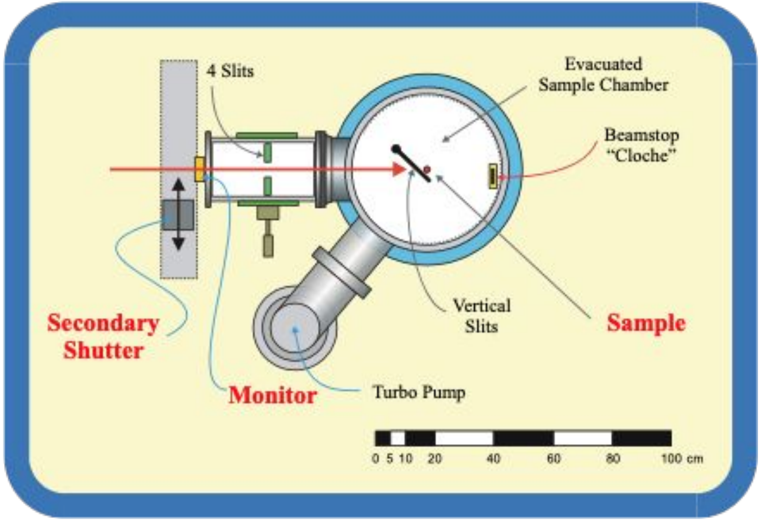
Elastically bent Si111 and Si311 reflections

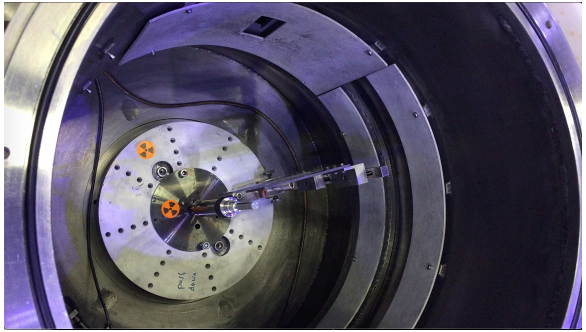


Mosaic Cu220 and Cu331 crystal faces

The sample chamber

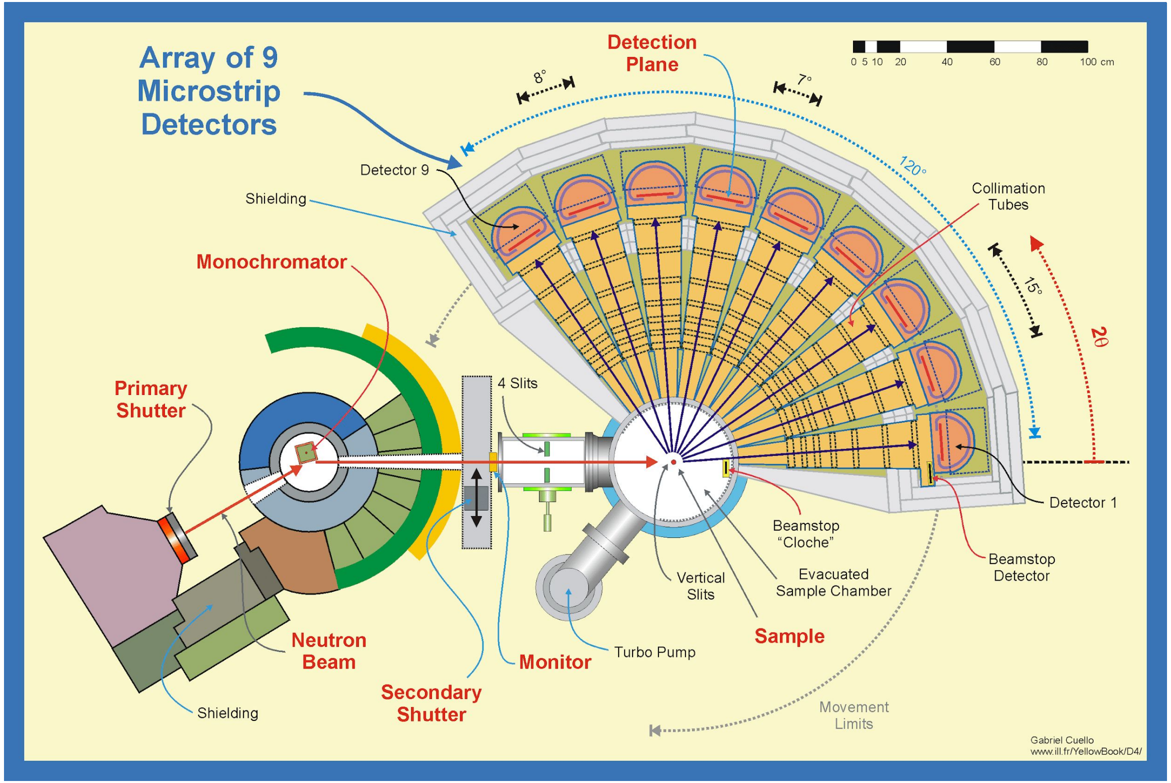
- Cylindrical evacuated chamber of 50 cm diameter and about 50 cm height.
- It houses all sample environments.
- Rotating omega plate.
- Adjustable vertical slits of high-density $^{10}B_4C$.
- The vacuum is about 10^{-6} mbar.
- Beamstop inside the bell jar





The detector

In operation since 2000.

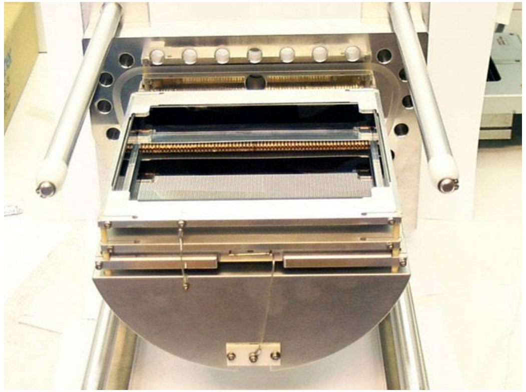


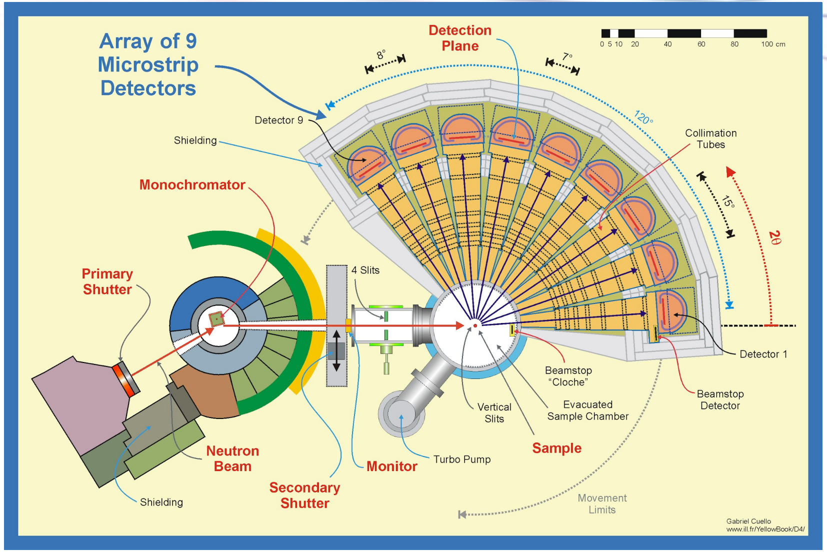
Between the bell jar and each of the detectors there is an evacuated collimation tube.

These collimation tubes eliminate air-scattering and also block the Al reflections coming from the bell jar exit.

The first detector has a mobile dedicated beamstop.

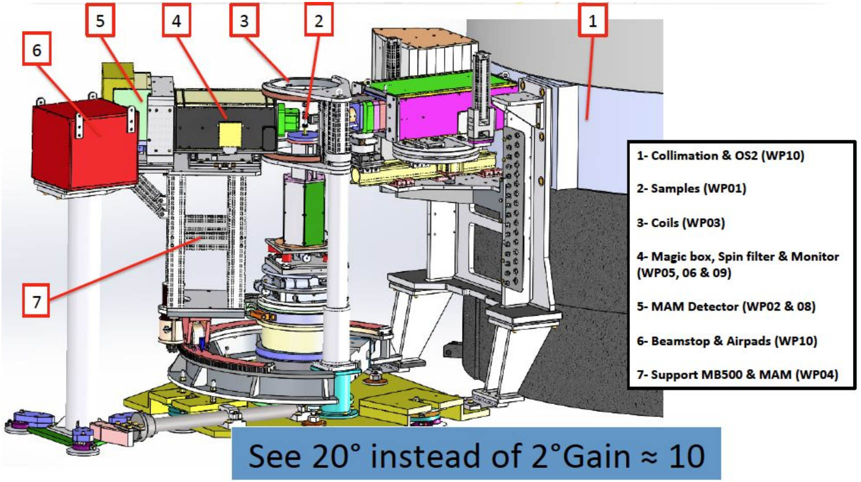
The detector



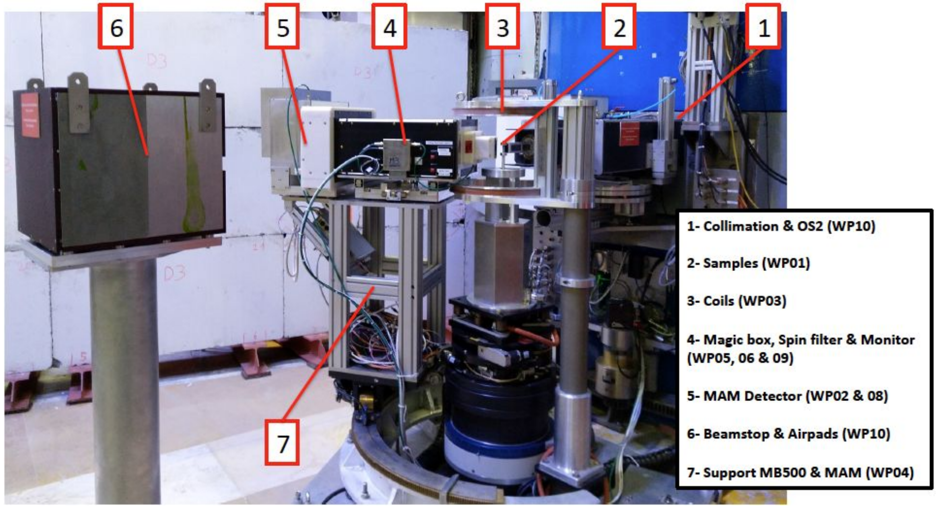


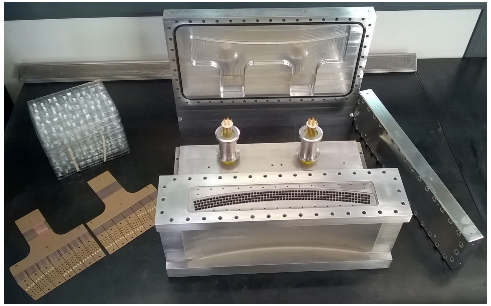
- 9 microstrip detectors in a fan-like array.
- Detection plane at 1.146 m from the sample position.
- Each detector has 64 microstrip detector cells.
- Detector cell 100 mm high and 2.5 mm wide, thus subtending 0.125° in 2θ per cell.
- Angular range: 8° per detector, with an angular gap of 7° between detectors.
- Useful angular range: 1.5° to 139° in 2θ.
- Detection efficiency of about 81% at 0.5 Å.
- Detection depth of 30 mm for the ^3He detection gas at 15 bar.

Complementarity with D3

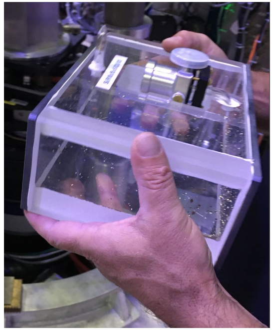


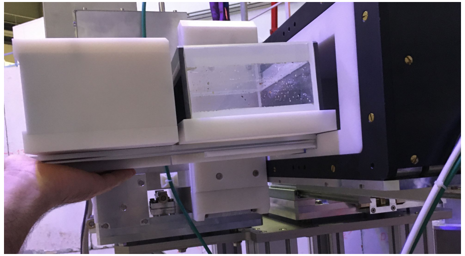
See 20° instead of 2°Gain ≈ 10





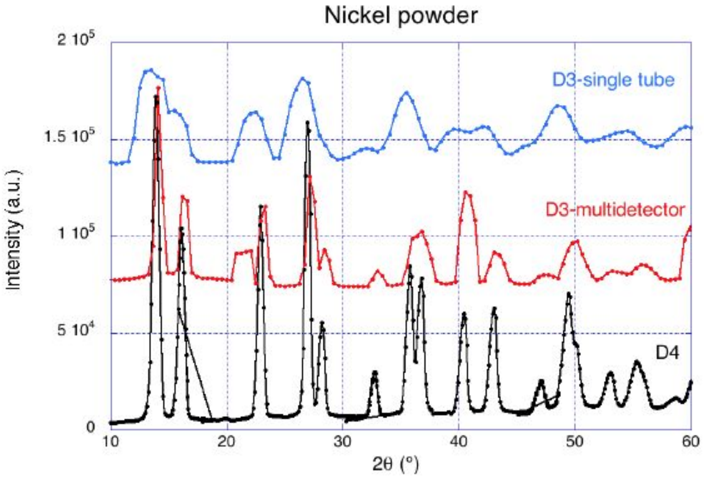
- 4 rows of 56 detectors (square section tubes)
- 15 atm of ^{3}He





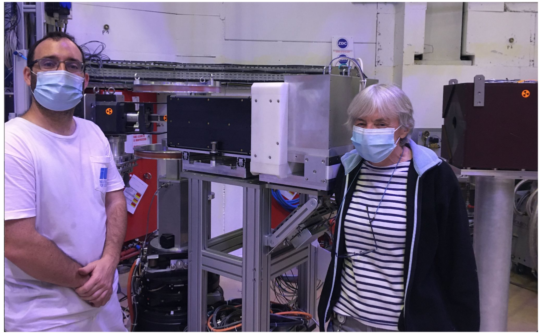
Complementarity with D3

- New 1-D detector covering 20° in 2Θ (only 2° before).
- About 6 times faster than the previous configuration.
- Polarisation analysis.
- Allows a direct measurement of the spin incoherent scattering.
- Useful for hydrogenated materials.
- Could help with about 10% of the proposals done at D4.



$$
\begin{aligned}
I_{spin-incoherent}(Q) &: 1/3\; non-spin-flip \\
&\quad\; 2/3\; spin-flip \\
I_{coherent}(Q) &: non-spin-flip \\
I_{isotope-incoherent}(Q) &: non-spin-flip
\end{aligned}
$$

- Not yet commissioned.
- Worse *Q*-resolution than D4.
- Longer counting times than D4.
- Data analysis more tricky.



Available sample environments

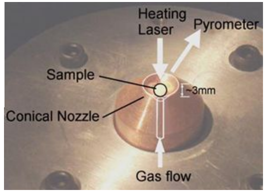
Ambient: In belljar under vacuum. If needed, possibility of having Ar or He atmosphere.

Furnace (F1): Vanadium heating element, T_{max} ~ 1300 C when using Ar leak at ~ 10^{-4} mbar.

Cryofurnace: 2 K < T < 550 K, T-control and heating above and below sample in calorimeter.

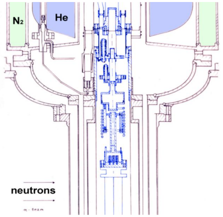
ILL-standard pressure cells (clamp or gas loaded): Two TiZr cell

Paris-Edinburgh press: (see later slides).



Aerodynamic levitation with laser heating: (see later slides).

Dilution inserts: Never used but compatible with our cryofurnace.



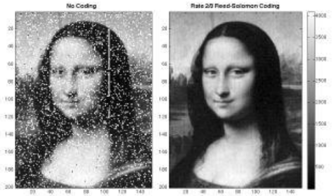
Custom/user-supplied sample environments:
Inserted into bell jar (about 500 mm diameter) or cryofurnace (50 mm diameter).

Software

Data reduction

- **`d4creg`**: regroups several runs (numors) in a single diffractogram
- **`d4opr`**: performs simple operations between regrouped files
- **`d4eff`**: produces a relative efficiency file
- **`d4nifit`**: fits a Ni diffractogram varying the wavelength and the zero angle
- **`d4cget`**: extracts parameters registered in numor files

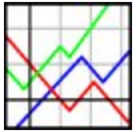
Data correction/analysis



- **`correct`**: performs corrections of attenuation, multiple scattering and normalisation
- **`polyfit`**: performs inelasticity corrections fitting a polynomial
- **`lorgaun`**: performs inelasticity corrections fitting a Lorentzian-Gaussian function
- **`d4fou`**: performs a sine Fourier transform of the structure factor
- **`d4soq`**: produce a variety of useful *Q*-space functions

Plotting

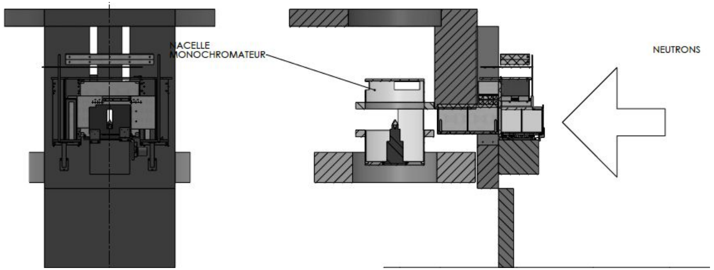
- **`gnuplot`**: a portable command-line driven graphing utility



MANTID (for the near future)

Recent upgrades

Only the past 5 years



- New vanadium tail for the D4-specific cryofurnace (2016).
- α_1 incident beam collimation in place and tested successfully (2016).
- Isostatic mechanical support for the air-pad system under the detector block (2017).
- Rewiring of the instrument: detector and motor cables principally (2017).
- New FPGA detector signal processing, installed under the detectors (2017).
- Successful first ASI/DSI texts and certification of the instrument (spring 2018).
- Updates+improvements to the D4-version of the data-analysis program **`correct`**, making it much more user-friendly and efficient, remaining of course command-line/script driven (2019).
- Start including D4 data treatment into MANTID (2020).

Risks

Low manpower

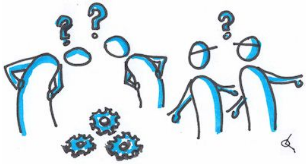
Quite small team dedicated to the study of the structure of disordered materials

ILL-internal risks

- Brainstorming ideas (see discussion later)
- Re-siting of D4: As in the case of IN1-Lagrange, D4 could make use of more beam time. But the low and stable background that we have at the present position would not be available elsewhere on Level C. The best option for having a 100% instrument is to remain at the H8 beam tube, but that seriously disadvantages the partner instrument. All things considered, for D4 it is much preferable to remain a 50% instrument at H8 rather than being a 100% instrument elsewhere.

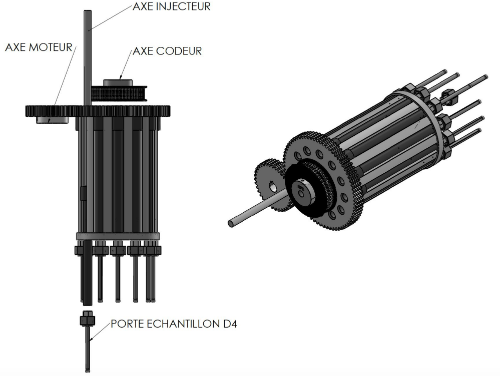
Technical risks

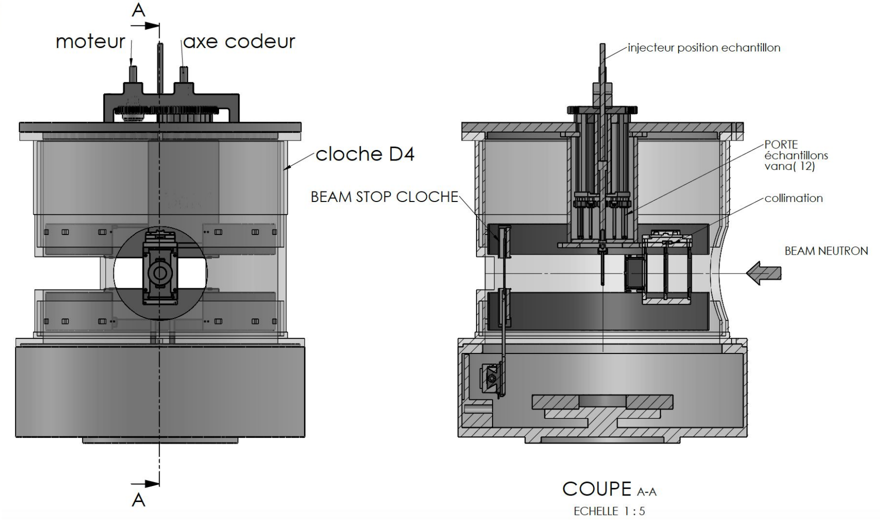
- Microstrip detectors (9 individual detectors + 1 spare)
- Incident beam monitor
- Linear motor of beamstop
- Special materials (high-density $^{10}B_4C$, for example)



Future developments

- Incident beam collimation
- Sample changers

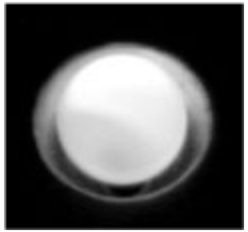




- Migration of D4-software to MANTID

- Levitation environment

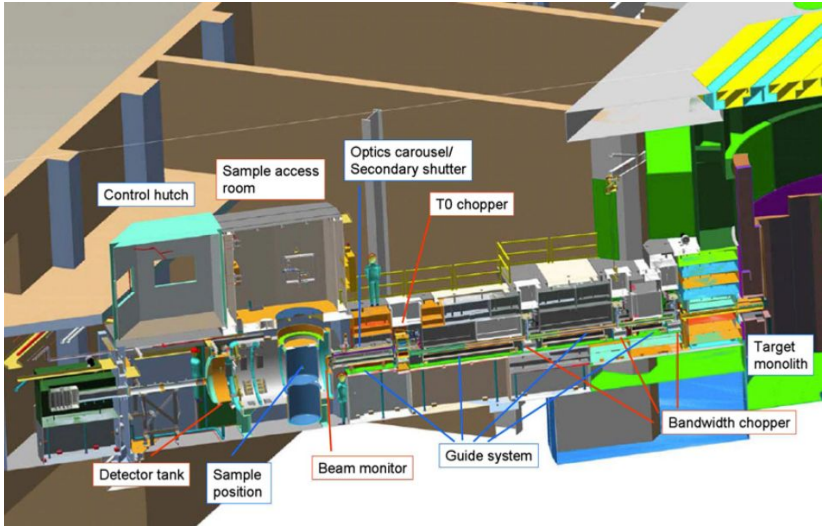
  The SANE group has part or even all the equipment to develop the aerodynamic levitation with laser-heating. With a relative small effort, this group could make again levitation experiments possible, for D4 and other instruments.

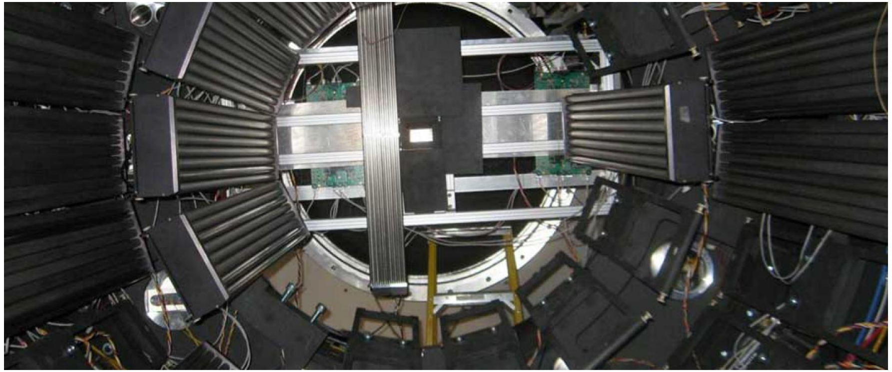


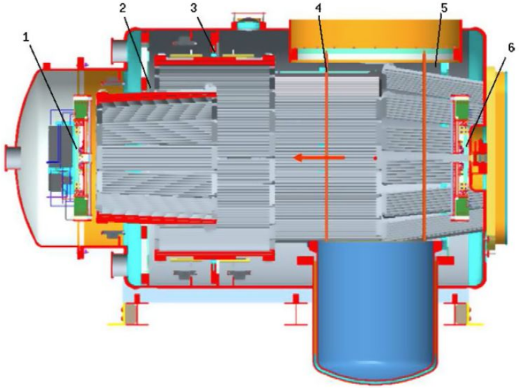
We pass the floor to Henry ...

Comparison with other instruments - SNS

NOMAD: Nanoscale-Ordered Materials Diffractometer



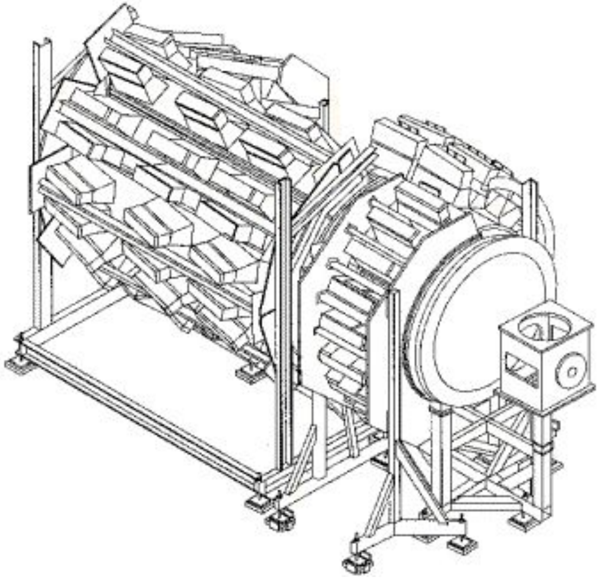


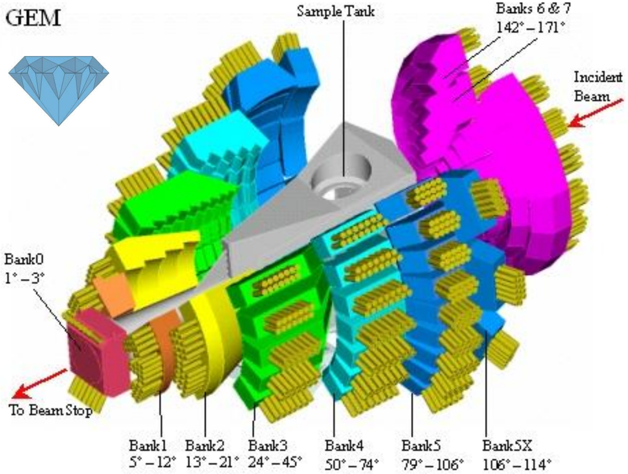


Comparison with other instruments - ISIS

ISIS Neutron and Muon Source

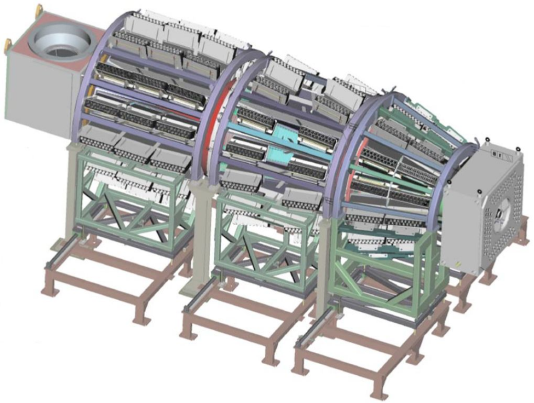
SANDALS





GEM

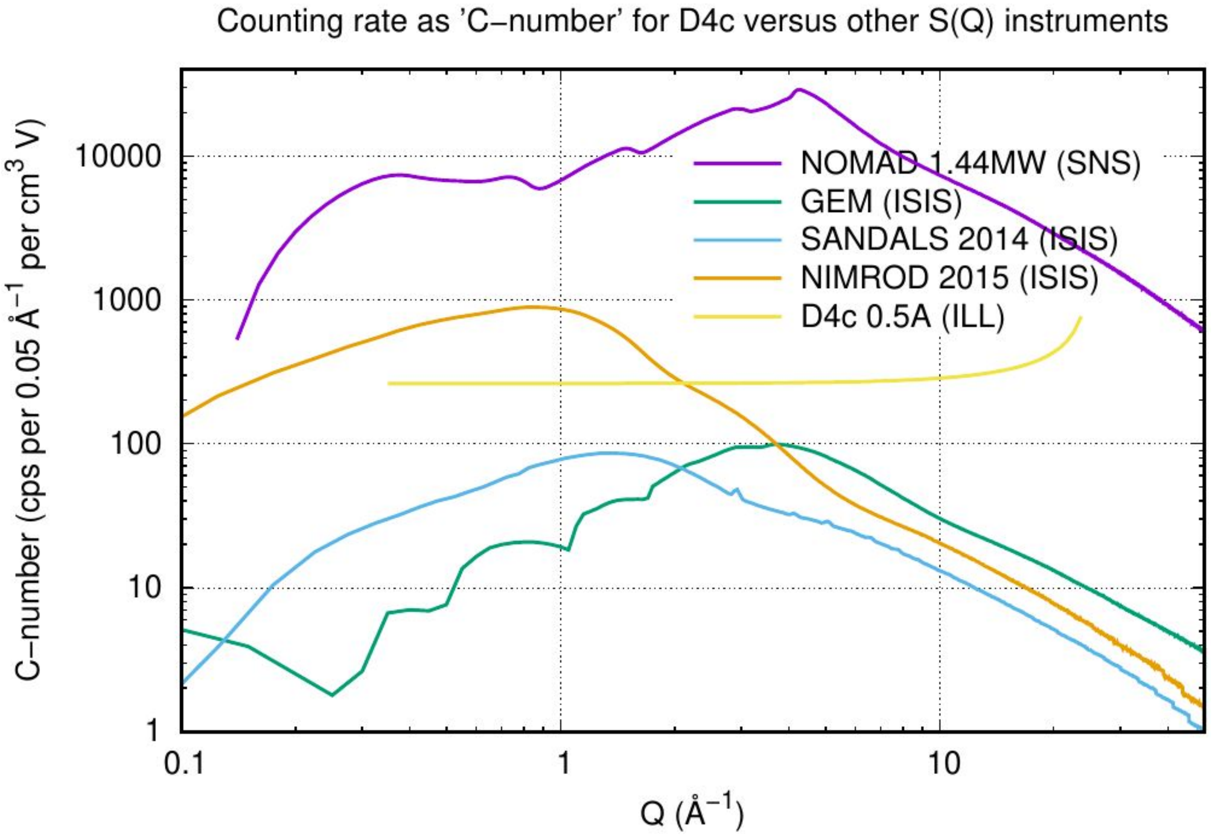
NIMROD



Comparison with other S(Q) instruments: C-number

The high-Q upturn in D4's counting rate is due to "Q compression" of Q = 4π/λ * sin(ϑ) as sin(ϑ) -> 1 at 90 deg.

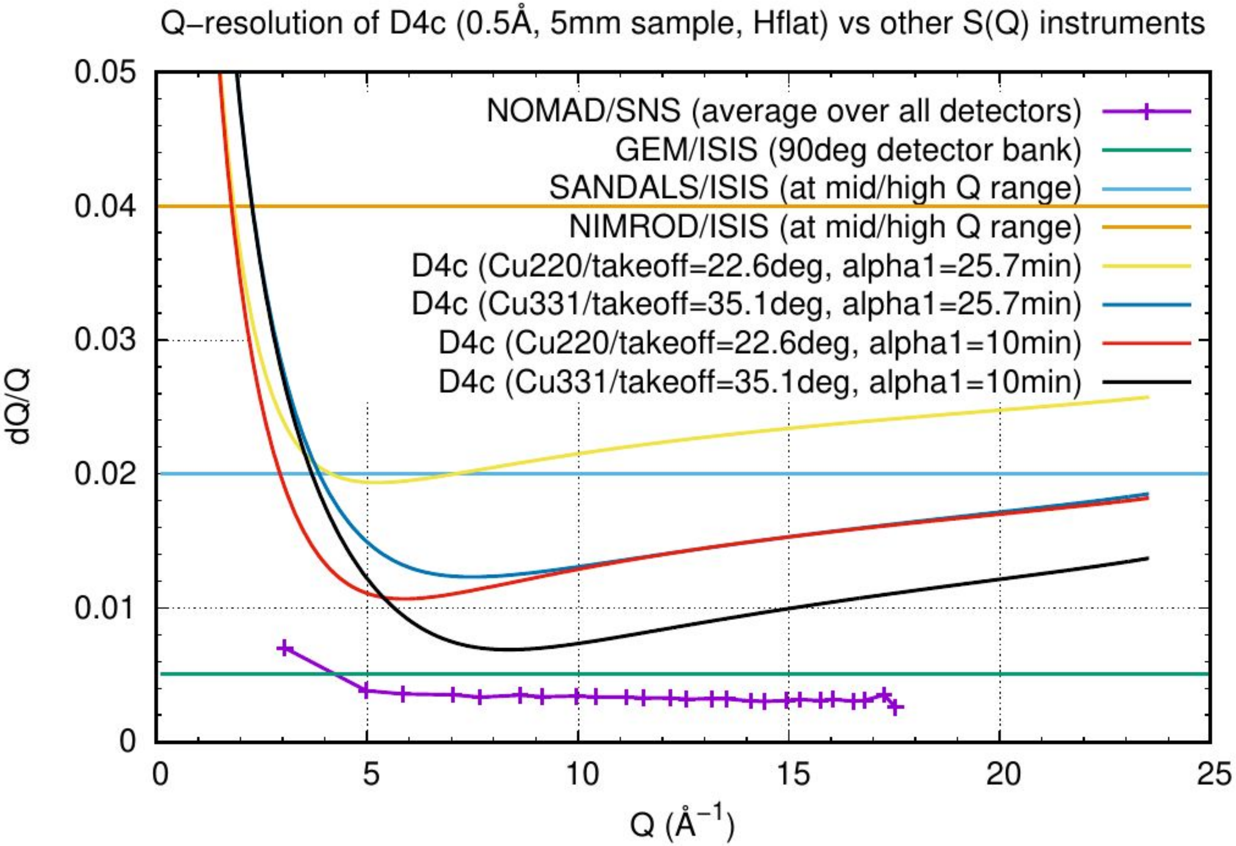
The shape of the curves for spallation source instruments is a function of the incident spectrum and detector coverage.



Comparison with other instruments: Q-resolution

D4's Q-resolution at 0.5 A is improved with higher takeoff angle and with the α_1 collimation, but naturally at the cost of reduced flux.

NB: D4's upturn at low Q does not here include the "umbrella effect".



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NEUTRONS FOR SOCIETY

Comparison with other $S(Q)$ instruments: Detector counting rate stability σ and instrument background counts

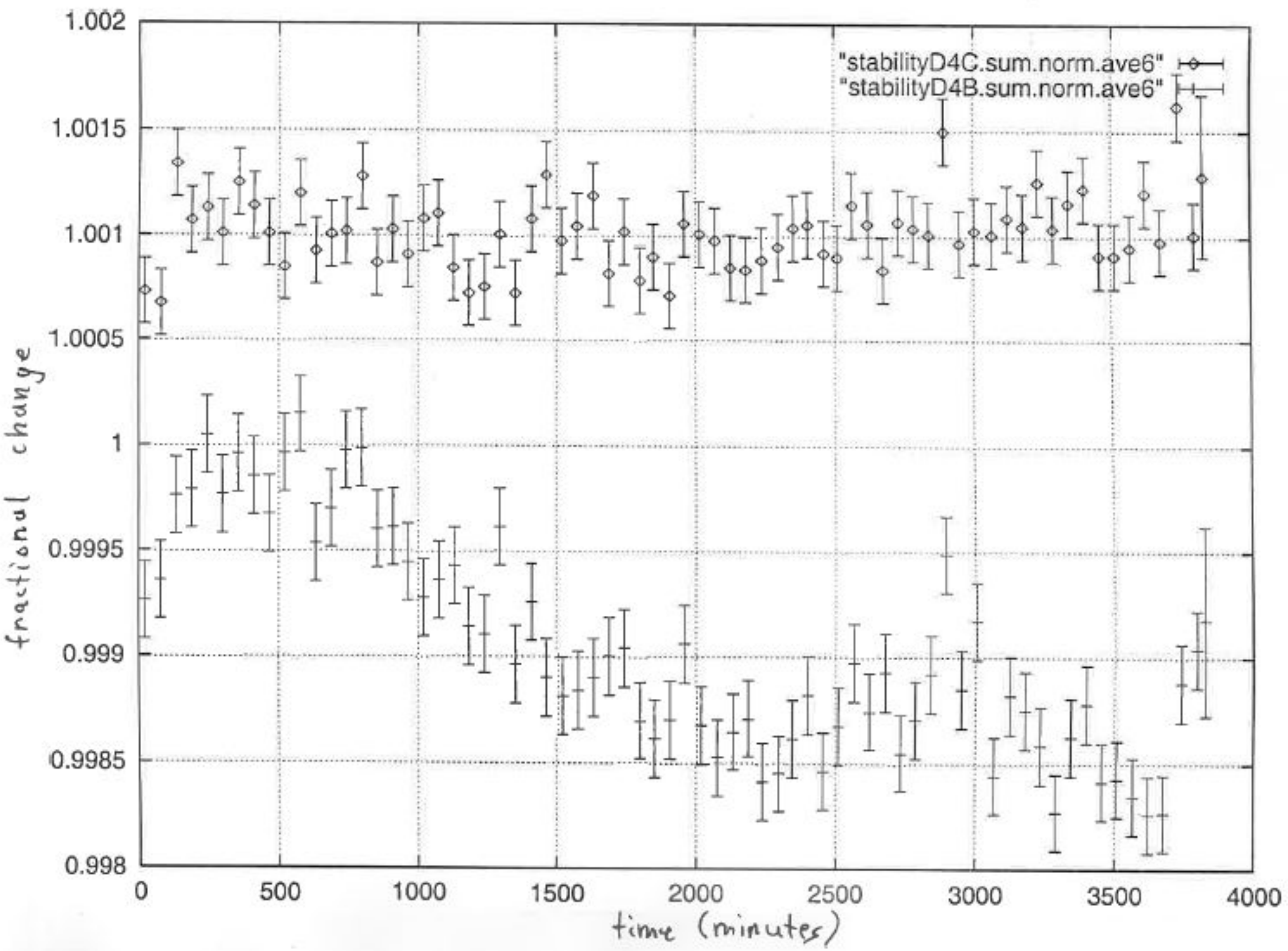
Recall that Fourier techniques in diffraction require very clean data, normalized to barns/str/atom, corrected for attenuation and multiple scattering, and as free from systematic errors as possible. NDIS in particular requires high reproducibility of diffraction patterns, and therefore high stability in detector (and monitor) counting rate, as well as low and stable background counts. The latter however is difficult to define/measure, but is rather learned from experience with the instrument.

D4c: measured $\sigma = 1 \times 10^{-4}$ over a few days (microstrip detectors).
NOMAD: measured $\sigma = 1 \times 10^{-4}$ over at least 4 hours.
NIMROD: nominally $\sigma < 5 \times 10^{-3}$ over several hours.
GEM: σ probably comparable to NIMROD.
SANDALS: σ estimated to be no better than 1% over a few hours (still so?).

As for background counts, NOMAD@SNS had some bad luck: the neighboring instrument has a monochromator (on a spallation source!) that is only a couple meters from the NOMAD sample. In general it is more difficult on a spallation source to have background counts that are low (e.g. from epithermal neutrons getting past the T0 chopper) and stable (ring current, pulse reproducibilities).

Stability over 2.6 days of the total counts of the D4c prototype detector and detector 1 of D4b, counting simultaneously on the same V sample, and here plotted after correction for monitor deadtime and averaging to 1-hour time intervals.

(from the ILL Report on the D4c prototype, No. ILL98FI15T, Nov 1998)



Science served by the instrument (highlights)

Traditional: Structure of liquids and glasses.

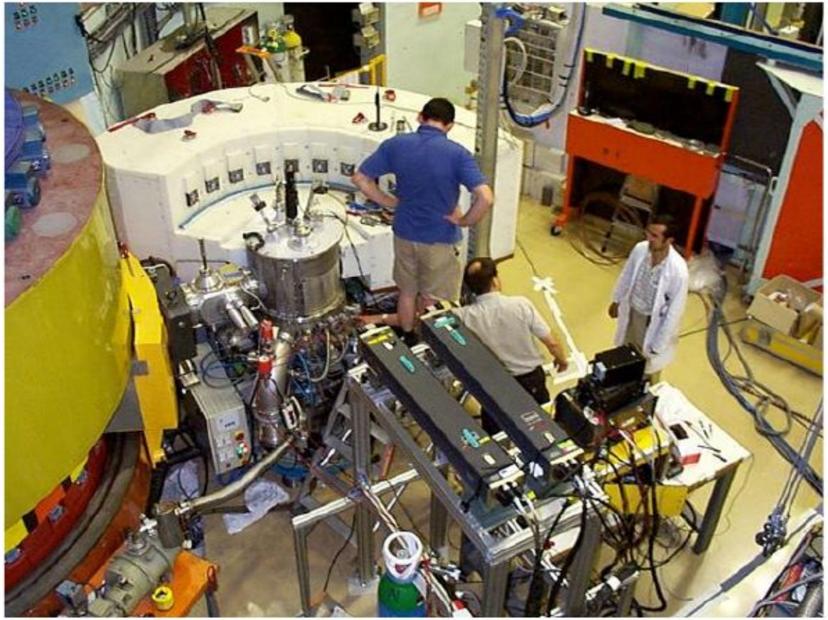
Opportunistic: Structure of neutron-absorbing materials (Cd, B, Gd, Sm, Eu, other REs), including magnetic systems.

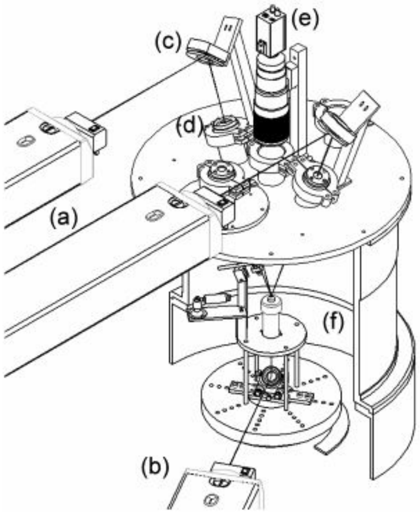
Recent: Structure of disordered crystals or nano-structured materials, sometimes still referred to as “PDF-analysis”.

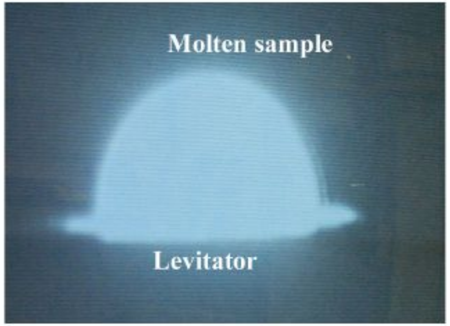
New: Magnetic PDF-analysis, a probe of static and dynamic spin-spin correlations through Fourier transform of (diffuse) magnetic scattering.

Aerodynamic levitation with laser-heating

Containerless technique, top+bottom IR lasers heat small (ϕ3 mm) samples (*e.g.* molten alloys, oxides) to 3000 °C w/o contamination, significant undercooling also possible.

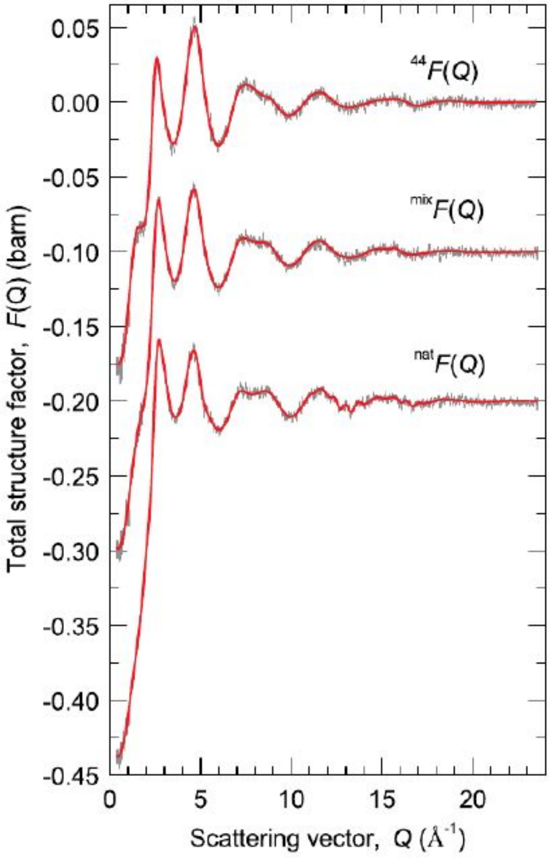




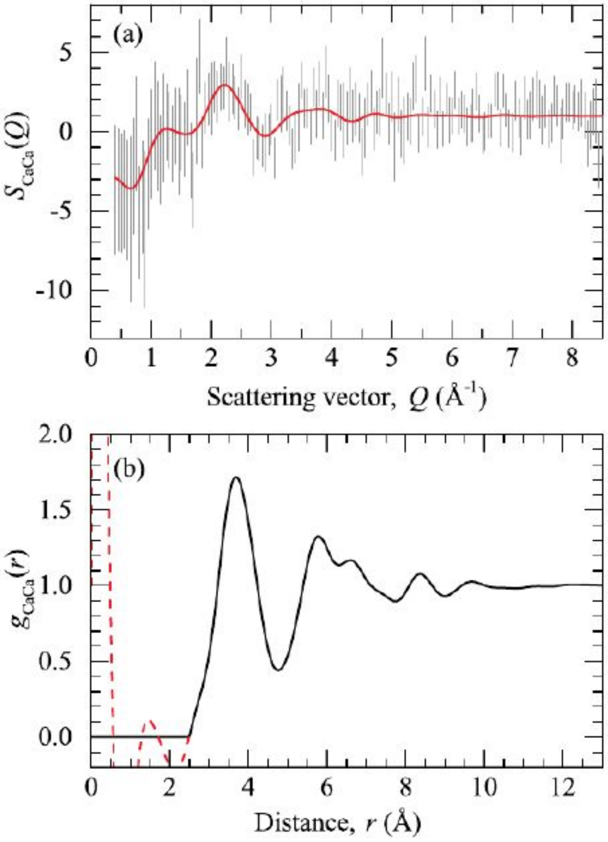


(developed by L. Hennet, et al, Orléans)

Total structure factors:



(44-mix) - (mix-nat):



A *double-difference* NDIS (Ca-isotope substitution) experiment on $CaAl_2O_4$ at 2000 C using aerodynamic levitation with laser heating.

Sample diameter ~ 2.5 mm.
Scattering length contrast:
b(natCa) - *b*(44Ca) =
4.70 fm - 1.42 fm = 3.28 fm.

=> Low+stable background counts are essential!

J.W.E. Drewitt, et al, Phys. Rev. Lett. **109** *(2012) 235501 (***22** *citations).*

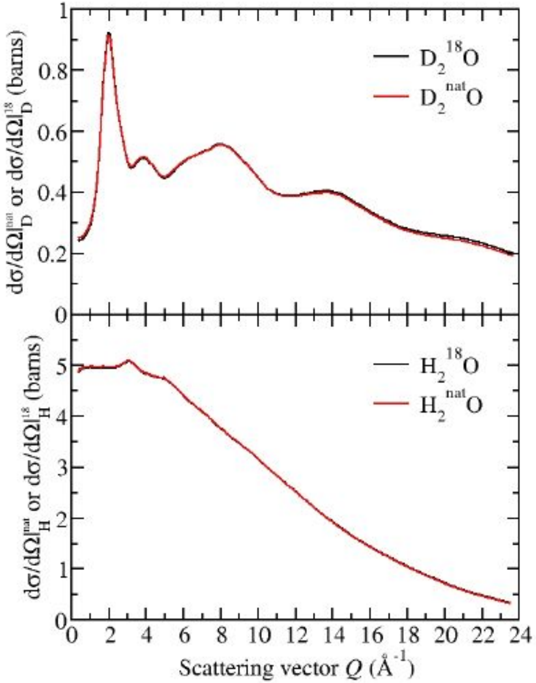
Ambient NDIS:

Very low contrast NDIS expt on D_2O and H_2O (ambient).

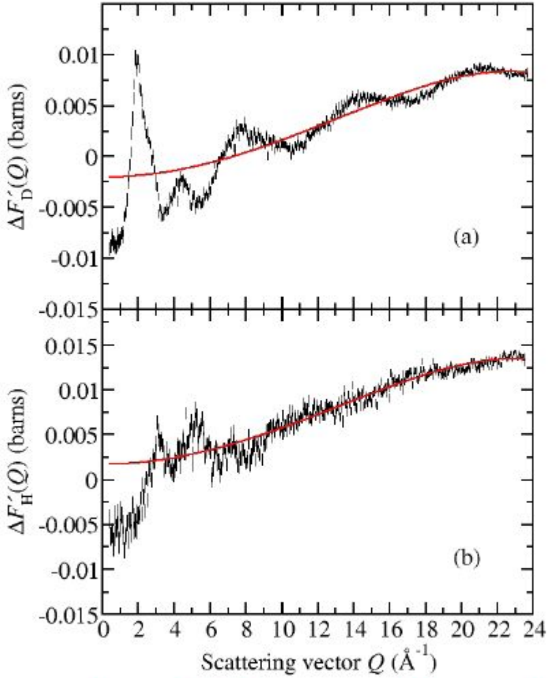
$b(^{18}O)$ - $b(^{nat}O)$ ~ 0.2 fm, as measured at S18 using neutron interferometry.

=> Low+stable background counts are again essential, and also ulta-stable monitor and detector counting rates (dependent on the ambient temperature of Niveau C).

Total structure factors:



^{18}O—^{nat}O first-differences:



Counting times of about 1 day per D sample and 3 days per H sample.

The measured signal for the ^{18}O—^{nat}O first-difference in the case of H_2O is only 0.2 % of the incoherent “background” intensity!

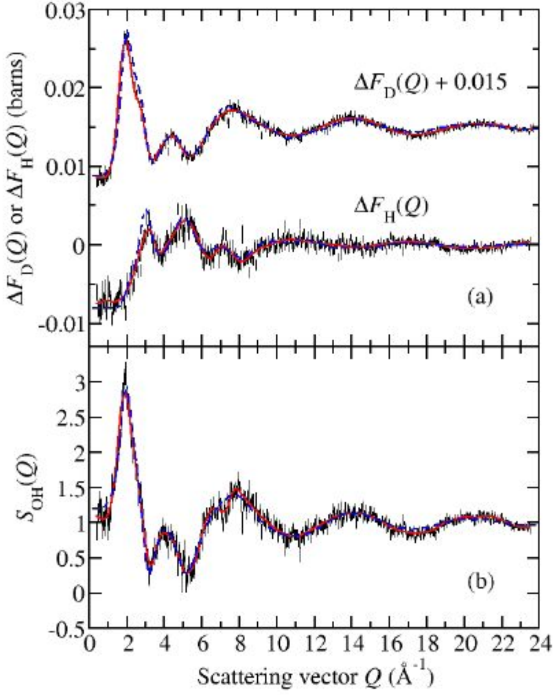
A. Zeidler, et al, *Phys. Rev. Lett. 107 (2011) 145501* (**38** *citations*).

Use Path-Integral Molecular Dynamics (PIMD) simulations to tease out the QM effects.

Recall: Egelstaff pointed out that for pair-wise additive potentials *u*(*r*) in an ergodic system (e.g. a liquid), the ***equilibrium structure*** as measured by diffraction *g*(*r*) ***is independent of atomic mass.***

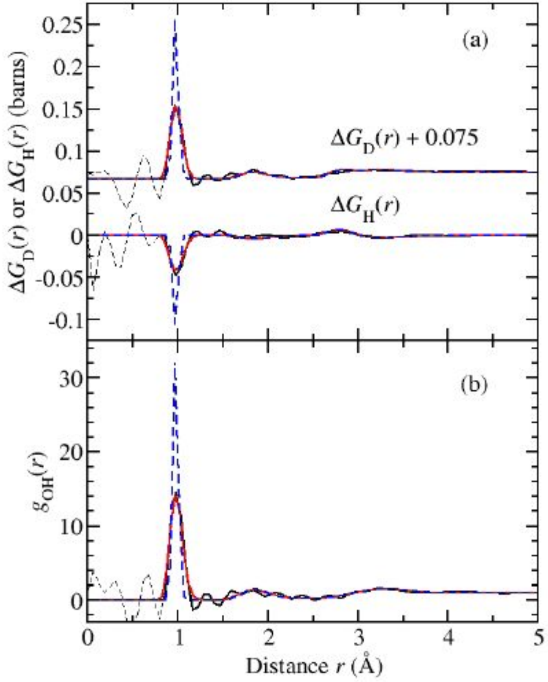
For example, any difference in x-ray diffraction data for H_2O vs D_2O at the same *T,P* is necessarily due to QM effects, as is their difference in freezing and boiling points.

Eliminate D-D and H-H:



— Spline fit to data
- - Model including QM effects

FT to R-space (O-D vs O-H):



— Model including QM effects
- - Same model, no QM effects

Our results show the intramolecular $r_{O-D}/r_{O-H} = 0.995$, in contrast to a value of 0.97 found by others, and in agreement with our molecular dynamics simulations (PIMD) that incorporate *competing QM effects.*

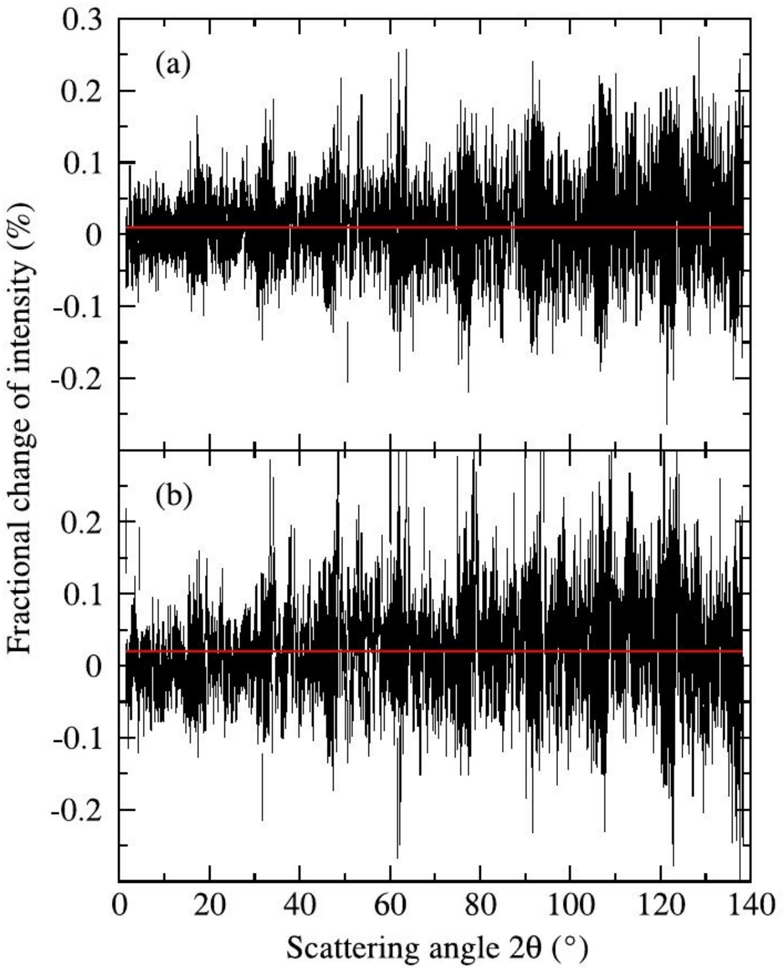
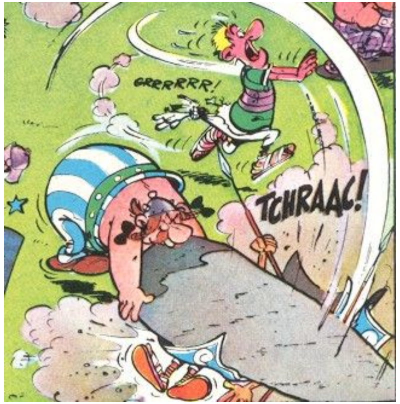
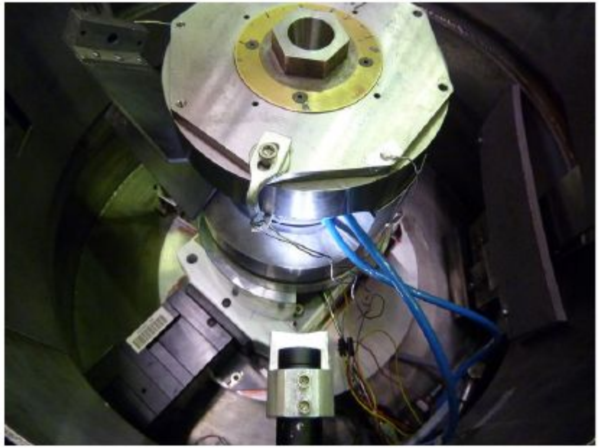
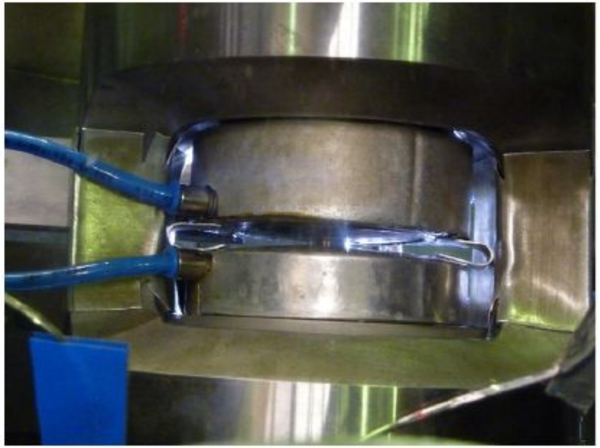


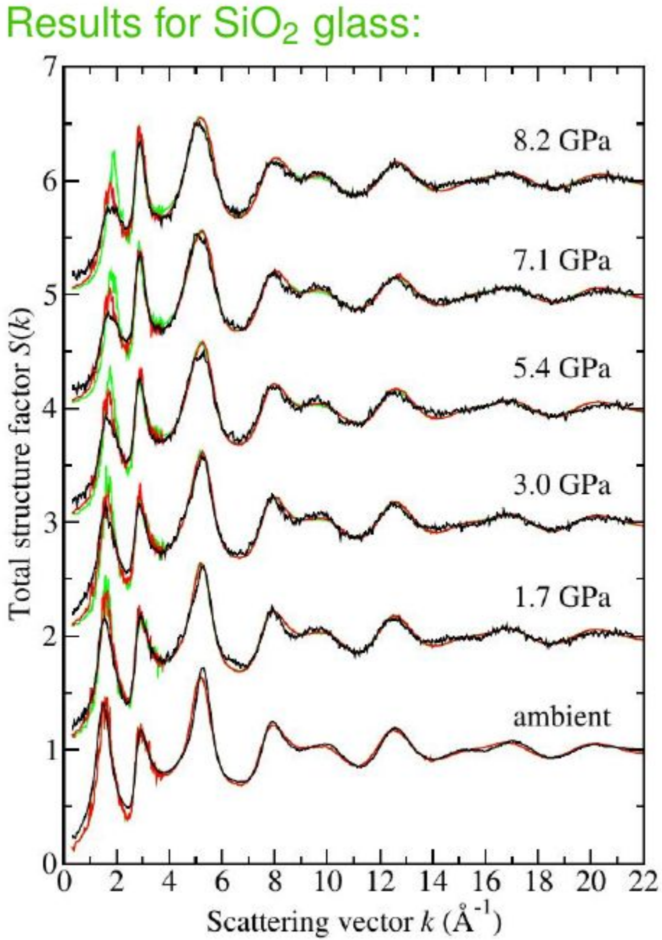
Figure 1. The stability and reproducibility of the diffraction patterns measured by using D4c [43, 44]. (a) The stability was assessed from the fractional change of intensity between the first and second halves of the data acquisition for the same mounting of a given sample. The data taken for a sample of $H_2^{18}O$ in its vanadium container are illustrated. The acquisition times for the first and second-half intensities, $I_{sc,1}(\theta)$ and $I_{sc,2}(\theta)$, were ~13.5 h each and the function $(I_{sc,2}(\theta) - I_{sc,1}(\theta))/(I_{sc,1}(\theta) + I_{sc,2}(\theta))$ is plotted as data points with vertical error bars. The fractional change of intensity shows no 2θ dependence within the statistical uncertainty and the fit to a constant level (horizontal solid light (red) curve) gives a value of 0.009(1)%. (b) The reproducibility was assessed from the fractional change of intensity between the diffraction patterns measured for two different mountings of a sample of $H_2^{nat}O$ in the same vanadium container. The acquisition times for the intensities $I'_{sc,1}(\theta)$ and $I'_{sc,2}(\theta)$ measured in the first and second mountings were ~10 and ~15.5 h, respectively, and the function $(I'_{sc,2}(\theta) - I'_{sc,1}(\theta))/(I'_{sc,1}(\theta) + I'_{sc,2}(\theta))$ is plotted as data points with vertical error bars. The fractional change of intensity shows no 2θ dependence within the statistical uncertainty and the fit to a constant level (horizontal solid light (red) curve) gives a value of 0.020(1)%. In (a) and (b) the data points show eight bands of higher density error bars in 2θ which correspond to the overlap regions between the 9 D4c multi-detectors.

PE press may be used under ambient T,P or with heated anvils (~200 C).







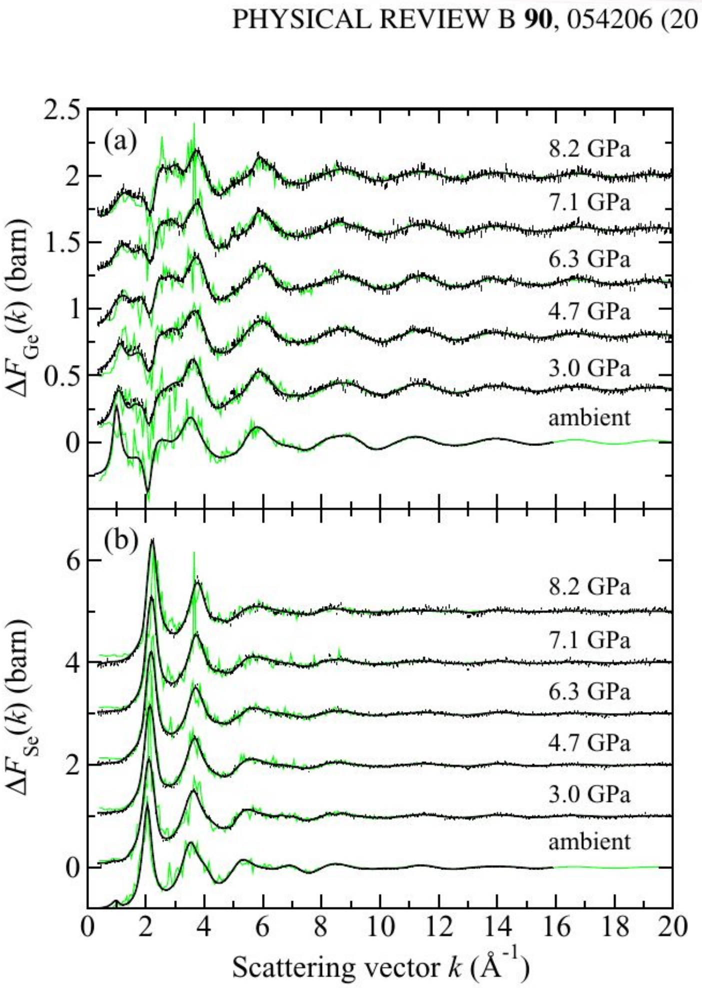


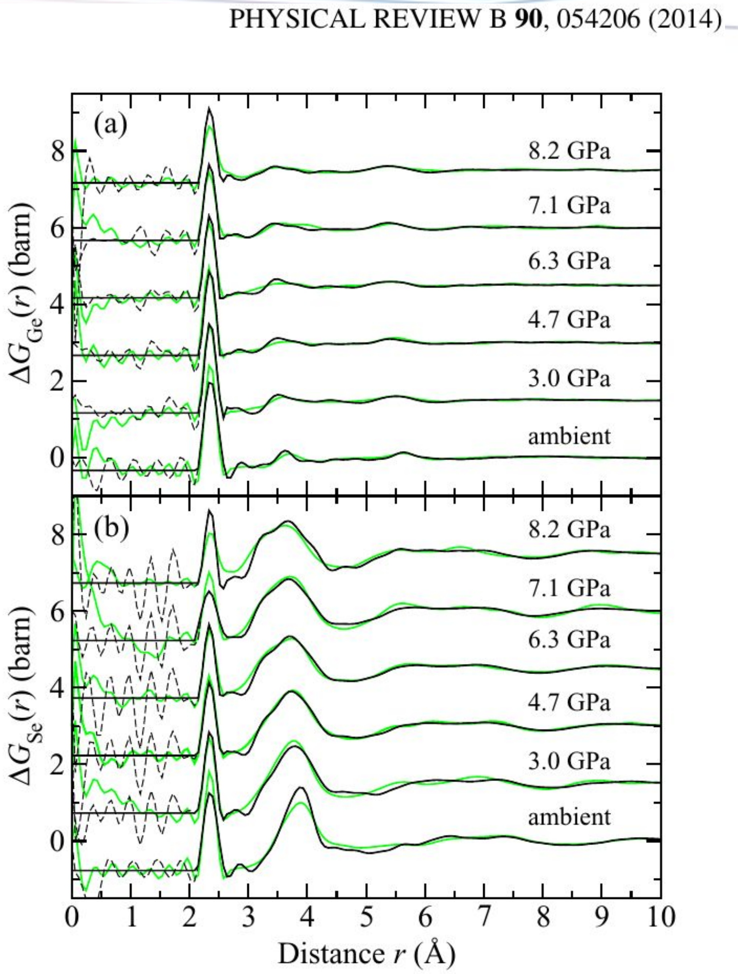
A. Zeidler, et al, *Phys. Rev. Lett.* **113** *(2014) 135501* (**45** *citations).*

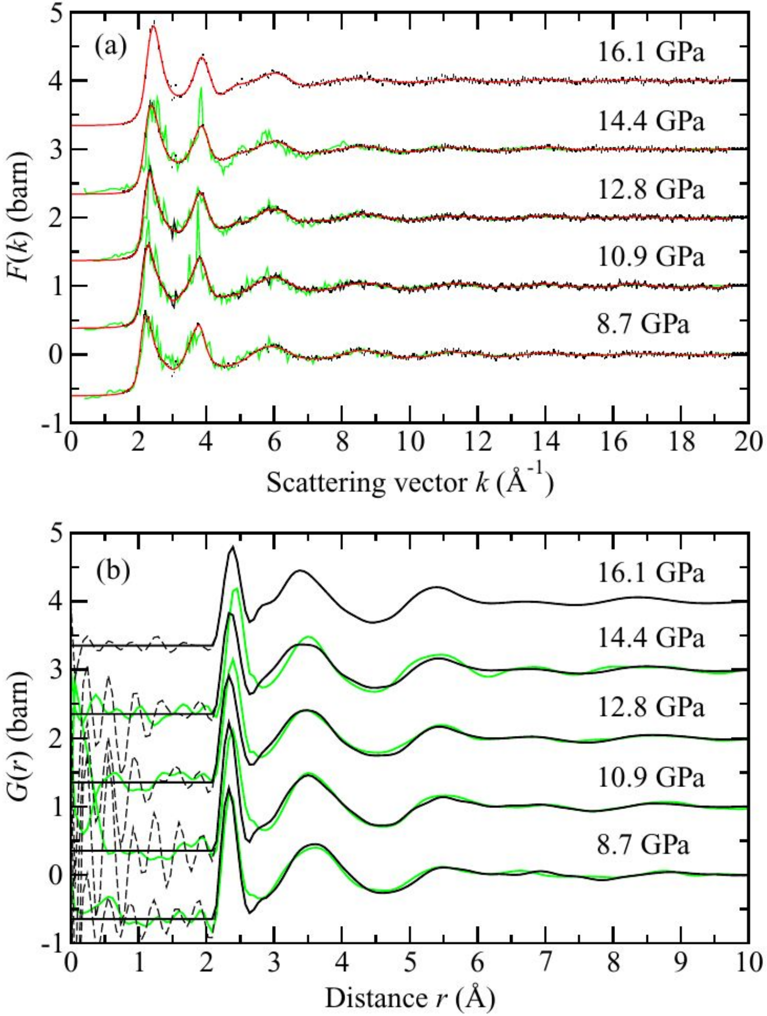
NDIS on $GeSe_2$ in PE press at D4c (ambient).

b_{natGe} = 8.185 fm.
b_{70Ge} = 10.0 fm.
b_{73Ge} = 5.09 fm.
b_{natSe} = 7.97 fm.
b_{76Se} = 12.2 fm.

Wezka *et al.*, PRB 2014.



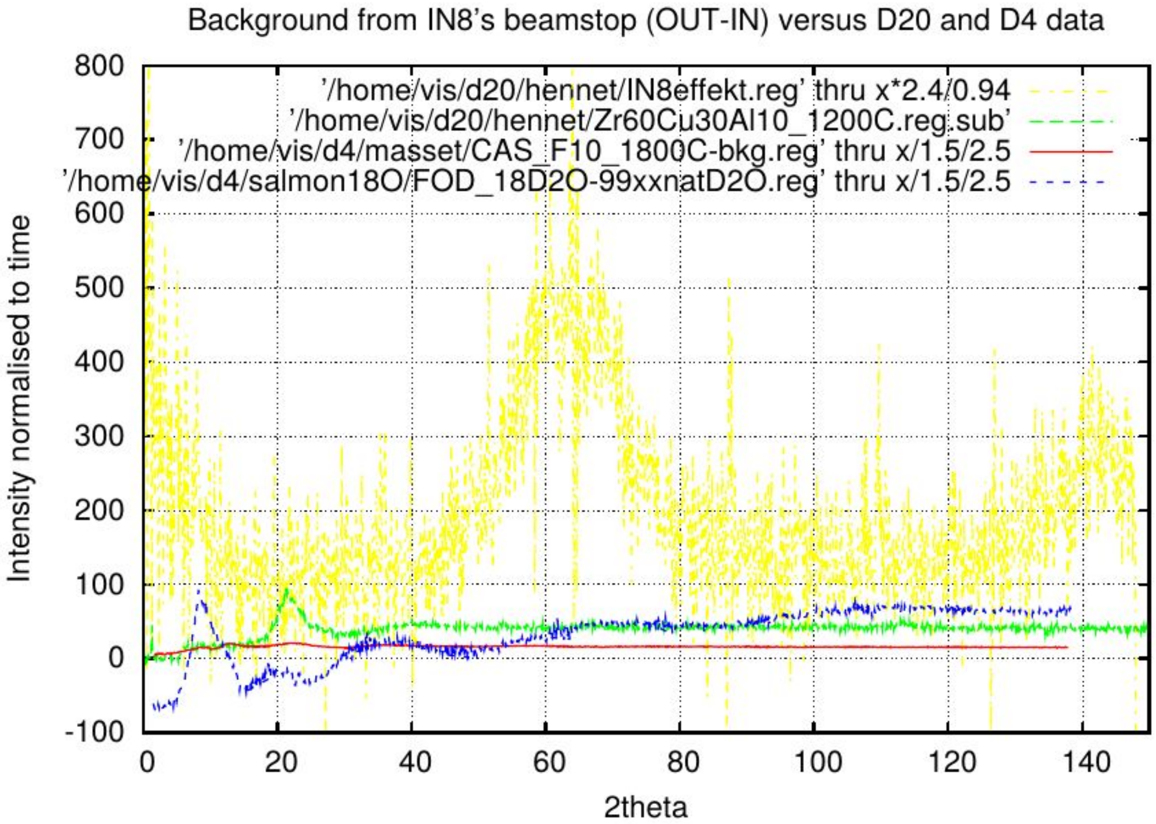




Same publication on $GeSe_2$: Only the total structure factor (i.e. no NDIS) seems to have been measurable on PEARL.

FIG. 3. (Color online) The pressure dependence of (a) the total structure factor $F(k)$ and (b) the total pair-distribution function $G(r)$ for ${}^{N}Ge^{N}Se_2$ glass. (a) The data sets measured using the PEARL diffractometer are given by the points with vertical error bars. The solid (red) curves show spline fits to these measured data sets, except when $k < 1.55$ Å^{-1} where they represent fitted Lorentzian functions because this region was not accessible in the diffraction experiments (see Ref. [31]). The solid light (green) curves show the FPMD results for pressures of 9.87, 11.56, 13.82, and 15.27 GPa. (b) The $G(r)$ functions shown by the solid (black) curves are the Fourier transforms of the measured $F(k)$ functions given in (a), and the broken (black) curves show the extent of the unphysical oscillations at r values smaller than the distance of closest approach between the centers of two atoms. The solid light (green) curves show the Fourier transforms of the simulated functions shown in (a) after applying the same maximum cutoff value $k_{max} = 19.55$ Å^{-1} as used for the neutron diffraction data.

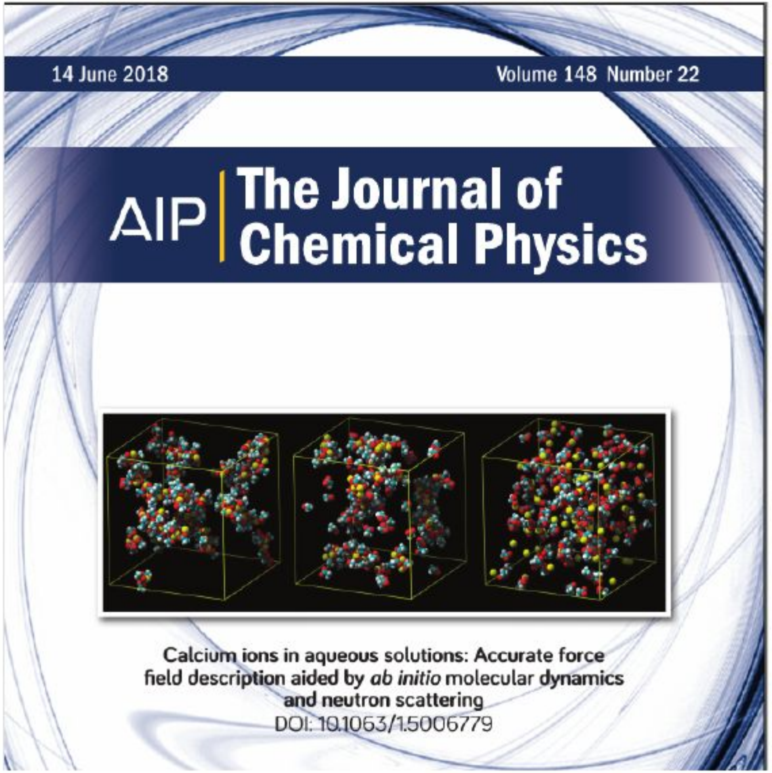
The 3 preceding NDIS experiments would not have been possible, even at the ILL with its intense and stable reactor source, if D4c were located elsewhere on Niveau C.

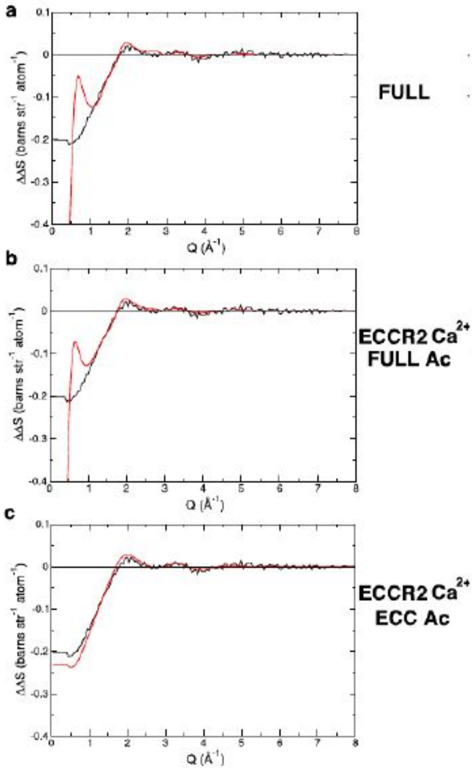


There's no place like home. D4 is threated by the persistent idea to move it to a different but full-time beamtube, which would probably be *mildly catastrophic* in terms of counting rate and background stability.

H/D substitution at D4c on aqueous solutions having applications to biological systems.

This team has performed other such "biology" experiments at D4, their S(Q) instrument of choice.





Renormalization of the ions' charges to lower values accurately emulates the polarization effects needed to prevent clustering in the simulations. Proposal 8-03-807 performed 11–15 Sept 2014 at D4, published as: *T. Martinek*, et al, *J. Chem. Phys.* **148** *(2018) 222813.*

FOR SOCIETY

Calcium ions in aqueous solutions: Accurate force field description aided by *ab initio* molecular dynamics and neutron scattering

Tomas Martinek,[1] Elise Duboué-Dijon,[1] Štěpán Timr,[1] Philip E. Mason,[1] Katarina Baxová,[1] Henry E. Fischer,[2] Burkhard Schmidt,[3] Eva Pluhařová,[4,a)] and Pavel Jungwirth[1,a)]

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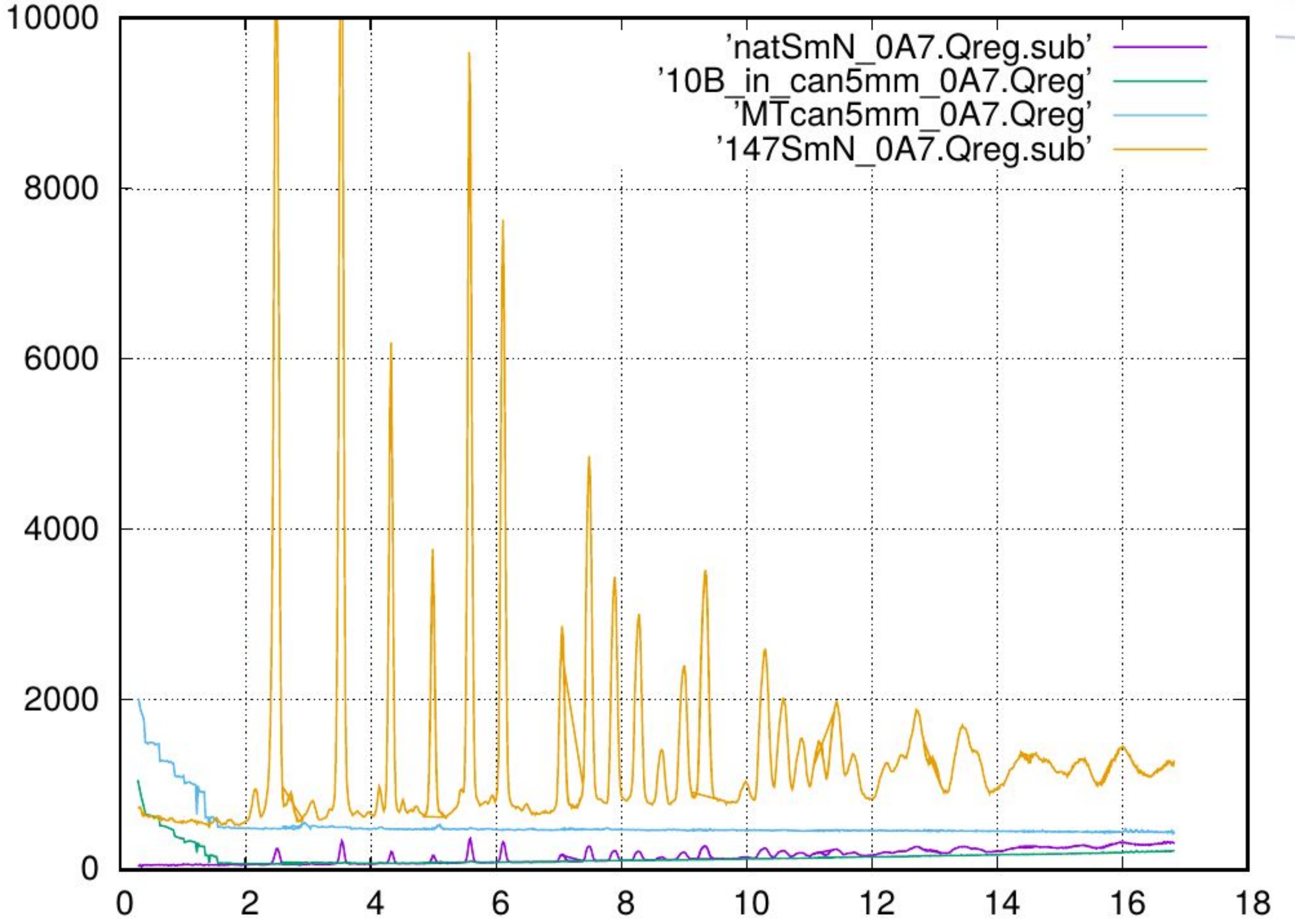
(Received 27 September 2017; accepted 2 December 2017; published online 6 March 2018)

We present a combination of force field and *ab initio* molecular dynamics simulations together with neutron scattering experiments with isotopic substitution that aim at characterizing ion hydration and pairing in aqueous calcium chloride and formate/acetate solutions. Benchmarking against neutron scattering data on concentrated solutions together with ion pairing free energy profiles from *ab initio* molecular dynamics allows us to develop an accurate calcium force field which accounts in a mean-field way for electronic polarization effects via charge rescaling. This refined calcium parameterization is directly usable for standard molecular dynamics simulations of processes involving this key biological signaling ion. *Published by AIP Publishing.* https://doi.org/10.1063/1.5006779

FOR SOCIETY

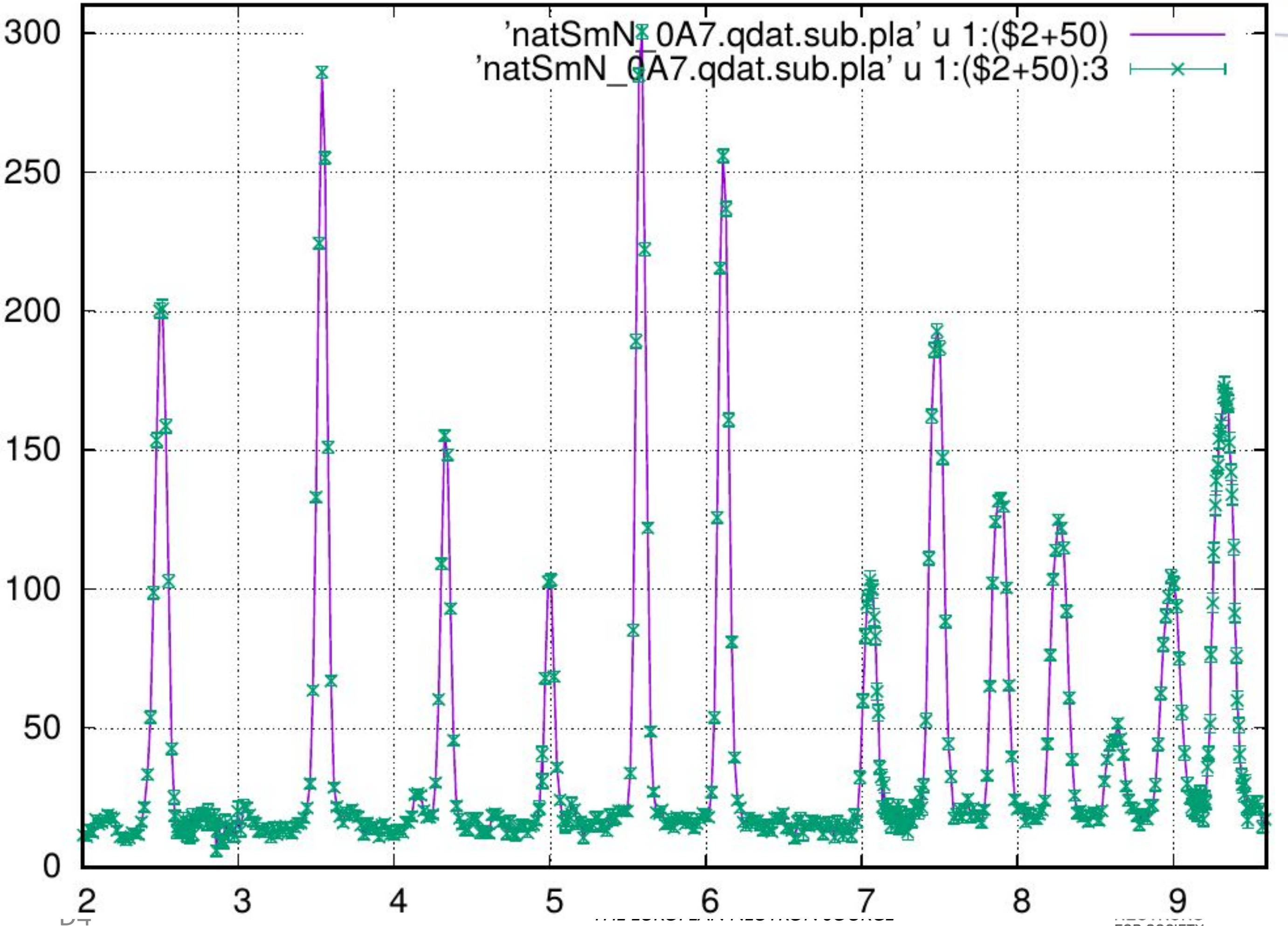
An “opportunistic” experiment example:



Even for a sample can diameter of only 5 mm, the mean transmission of the ^{nat}SmN sample at 0.7 Å is only ~ 6 %.

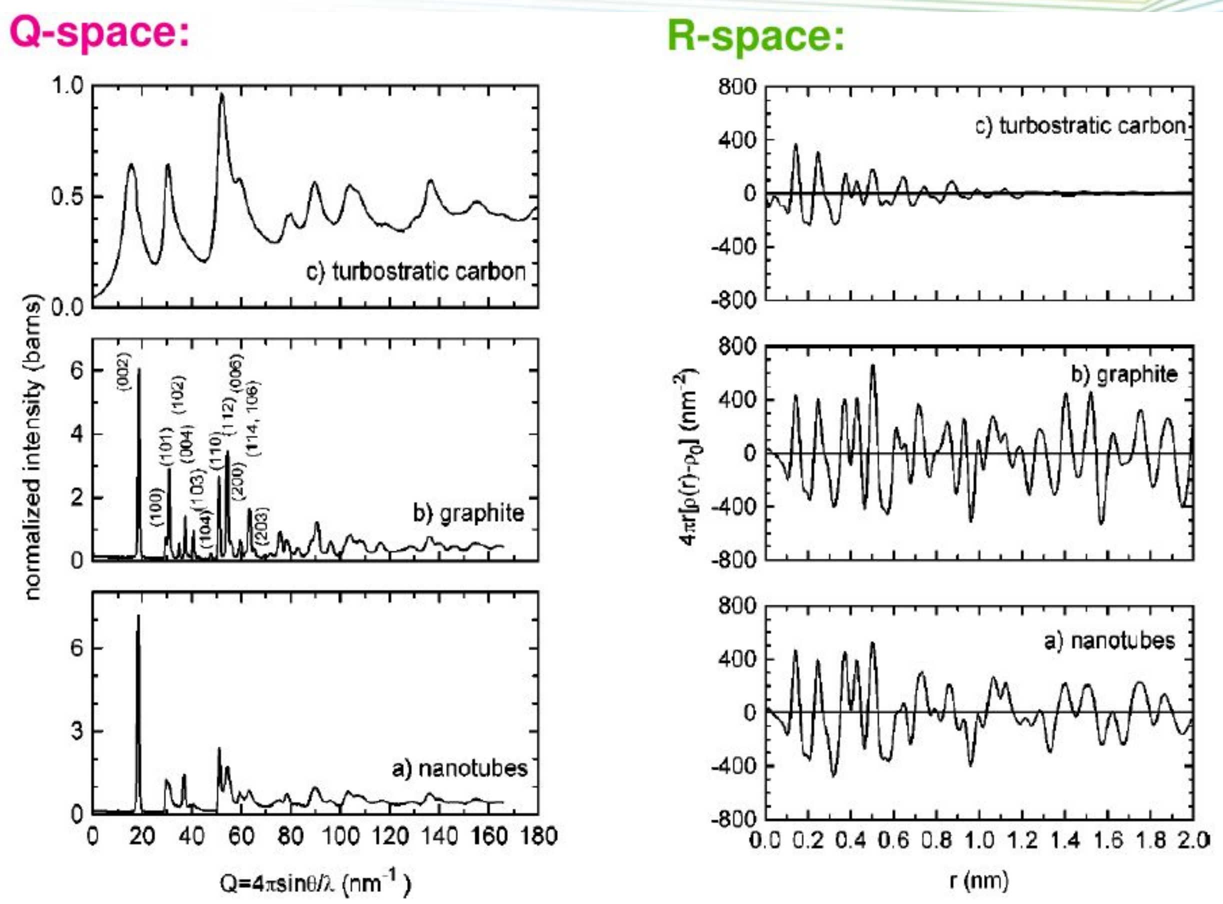
It’s diffraction intensity seem close to the same level as for ^{10}B powder, and considerably less that than of the empty container.

But after subtraction of the measured backgrounds, the final counting statistics are quite good after a couple hours counting.



First D4 results using “standard” atomic/nuclear PDF-analysis.

Here with 0.7 Å (Q_{max} ~ 17 $Å^{-1}$) instead of the now usual 0.5 Å (Q_{max} ~ 24 A^{-1}).



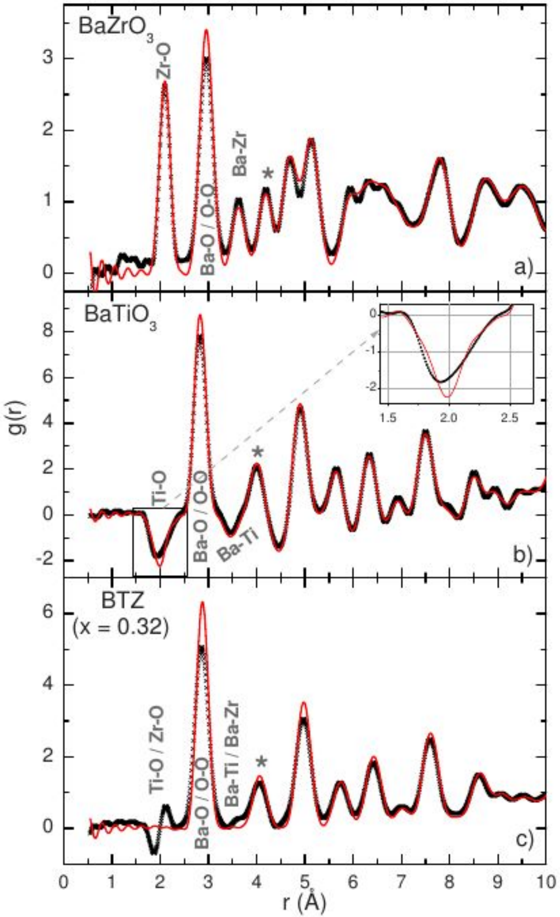
A. Burian, J.C. Dore, H.E. Fischer and J. Sloan, *Phys. Rev. B* **59** *(1999) 1665–8* (**60** *citations).*

Despite Vegard's Law, one can show that big atoms take up more space than small atoms !!

Such an effect cannot be seen with Rietveld that reveals structure only as averaged over space+time.

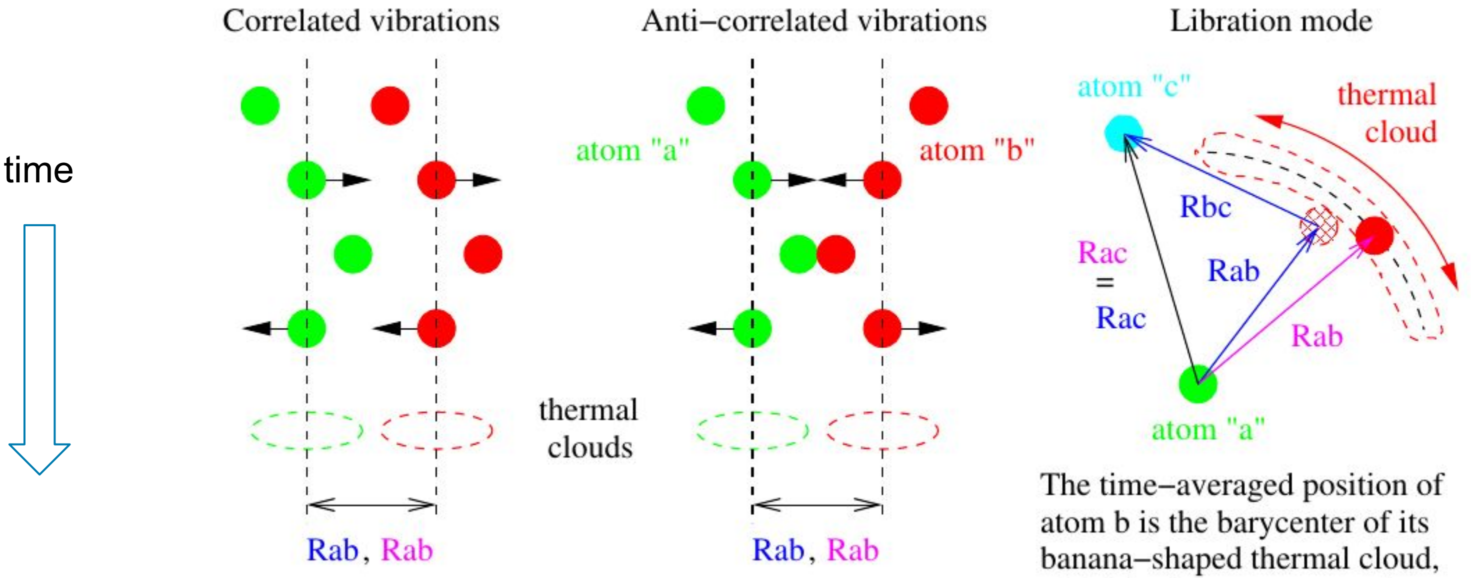
Bragg peak refinement shows that $BaTi_{1-x}Zr_xO_3$'s crystallographic structure is ABO_3 cubic perovskite for $x = 0$ and over the relaxor ferroelectric range ($0.25 \leq x \leq 0.5$) which includes the null-alloy composition $x = 0.32$. As charge disorder is minimized by the isovalent substitution Ti^{4+}/Zr^{4+}, it can be hypothesized that the long-range ferroelectric order is impeded by local structural distortions resulting from the large difference in the two cationic radii.

$\Rightarrow$ PDF-analysis using $\lambda = 0.5$ Å gave unambiguous evidence that the Ti and Zr atoms do not occupy equivalent octahedral sites as expected from the crystallographic structure, but rather the Ti atoms are displaced along [111].



C. Laulhé, et al, *Phys. Rev. B* **79** *(2009) 064104.*

Whereas Rietveld refinement gives time-averaged distances between atomic pairs, PDF-analysis sees an ensemble-average of quasi-instantaneous atomic positions and relative distances:



Rietveld−refined Rab = PDF−analysed Rab
for both correlated and anti−correlated vibrations,
but Rietveld's time−averaged thermal clouds
cannot distinguish between the two cases.
PDF(r) will however show a broader peak for
the a−b atomic pair in the anti−correlated case.

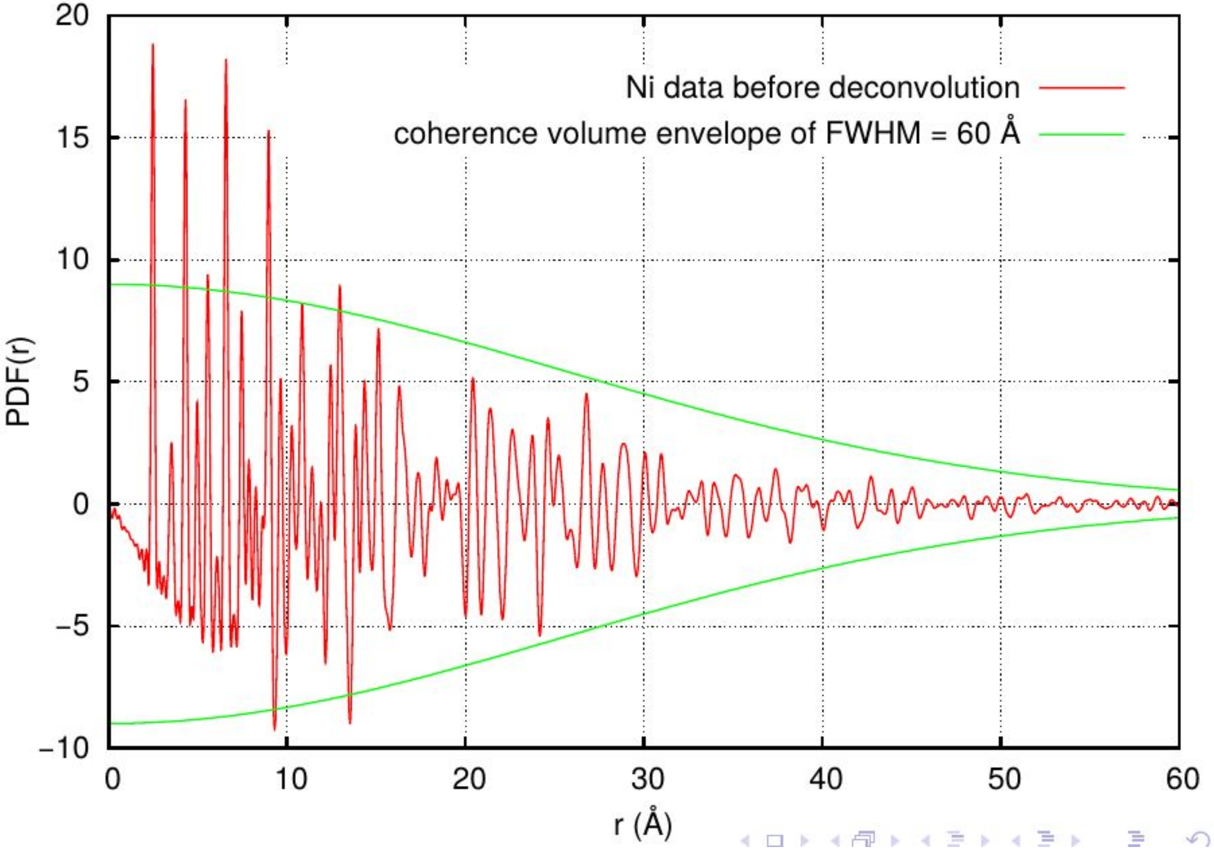
The time−averaged position of
atom b is the barycenter of its
banana−shaped thermal cloud,
which is closer to atom a than
any instantaneous position:

Rab (too short) < Rab (correct)

PDF(r) will show a sharp peak
for the a−b and a−c atomic pairs
but a very broad peak for b−c.

At short interatomic distances the peaks in $\mathrm{PDF}(r)$ are sharper and taller (conserving area $\propto$ coordination number) as compared to the neutron coherence volume's $\mathrm{FWHM} \sim 60$ Å for the D4c diffractometer:

0.5 Å @ D4c



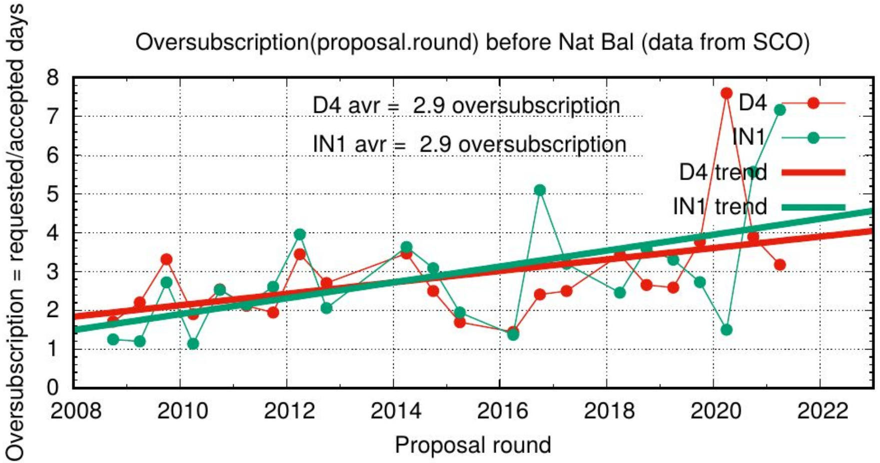
Magnetic PDF-analysis (mPDF):

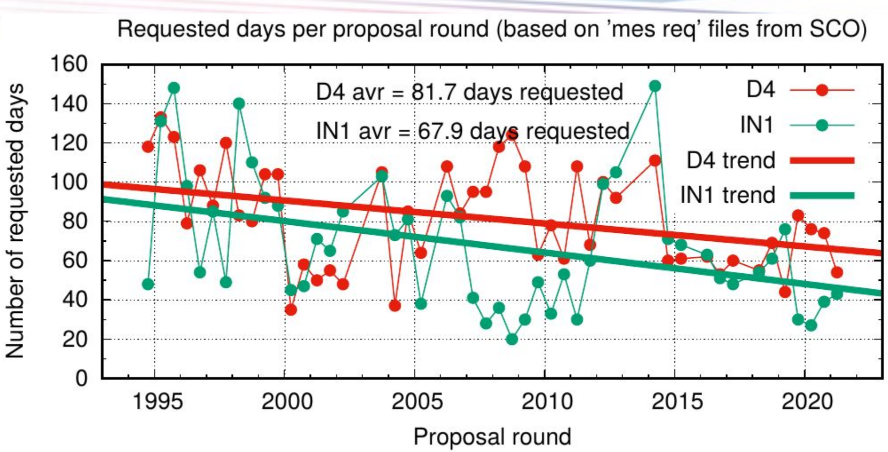
A pedagogical descriptive of magnetic PDF-analysis, along with examples of D4c data and preliminary results, can be found here:

https://www.ill.eu/fileadmin/user_upload/ILL/2_News_Press_Events/SciCo/Sci-Co-Public-Talks/SciCo101_Fischer_2019-11-08.pdf

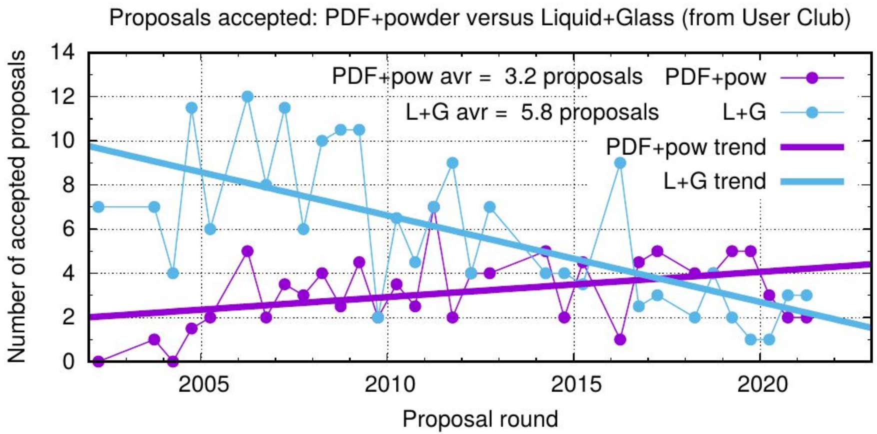
Statistics

Beamtime Proposals:





User base:

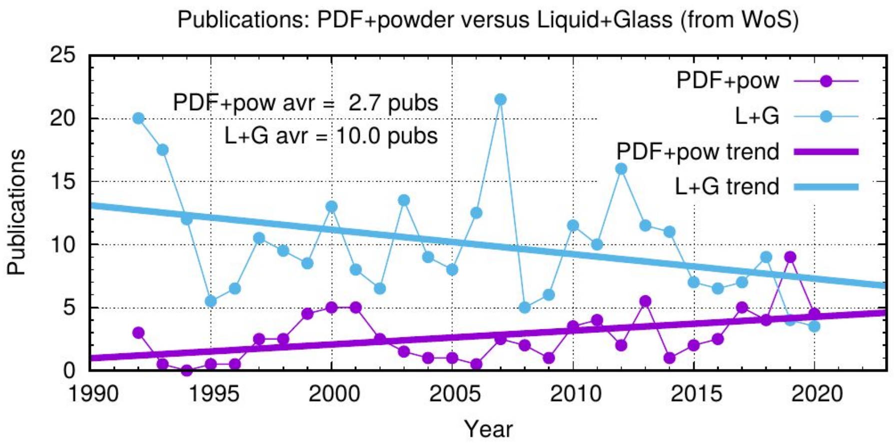


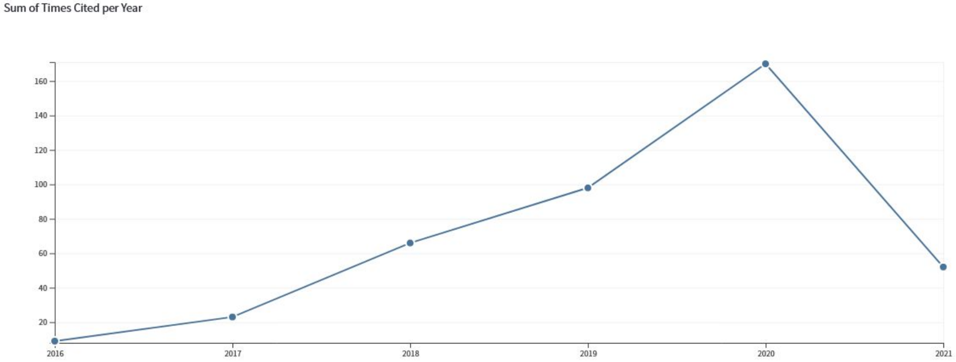
Quite balanced in nationality (de-anglosaxonized).

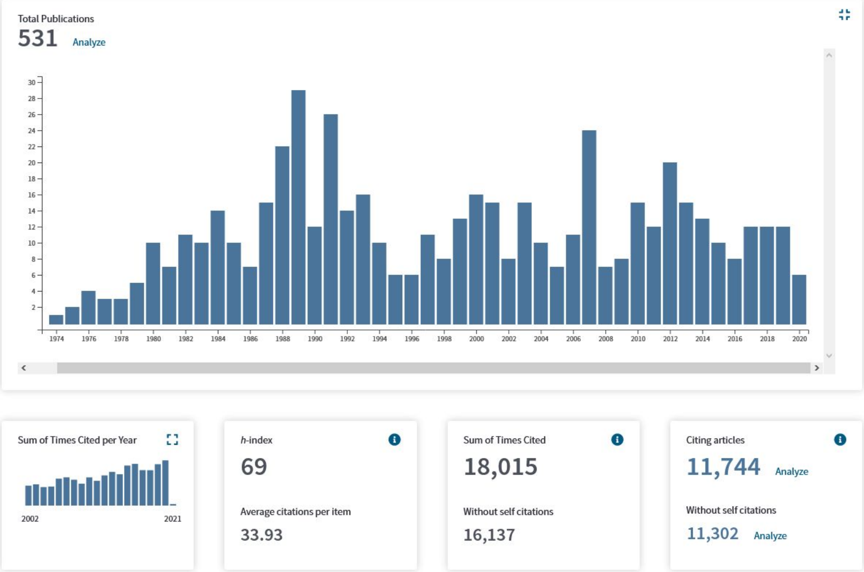
New/casual/inexperienced users, and increased EASY+DDT time, has lead to a bottleneck in the proposal/expt/data-analysis/publication pipeline.

Statistics

Publications and citations







D4's present H-index = 69, or ~98 when scaled to a full-time instrument (the highest at the ILL in 2017!), and D4's 10-year M-index = 2.83 (6th at ILL).

5-year impact factor of 3.4 (170 cites / 50 articles).

Brainstorming ideas

In the past, such ideas have generally not been motivated to improve D4's performance, but rather had other priorities and consequences that would have been very detrimental to D4's operation:

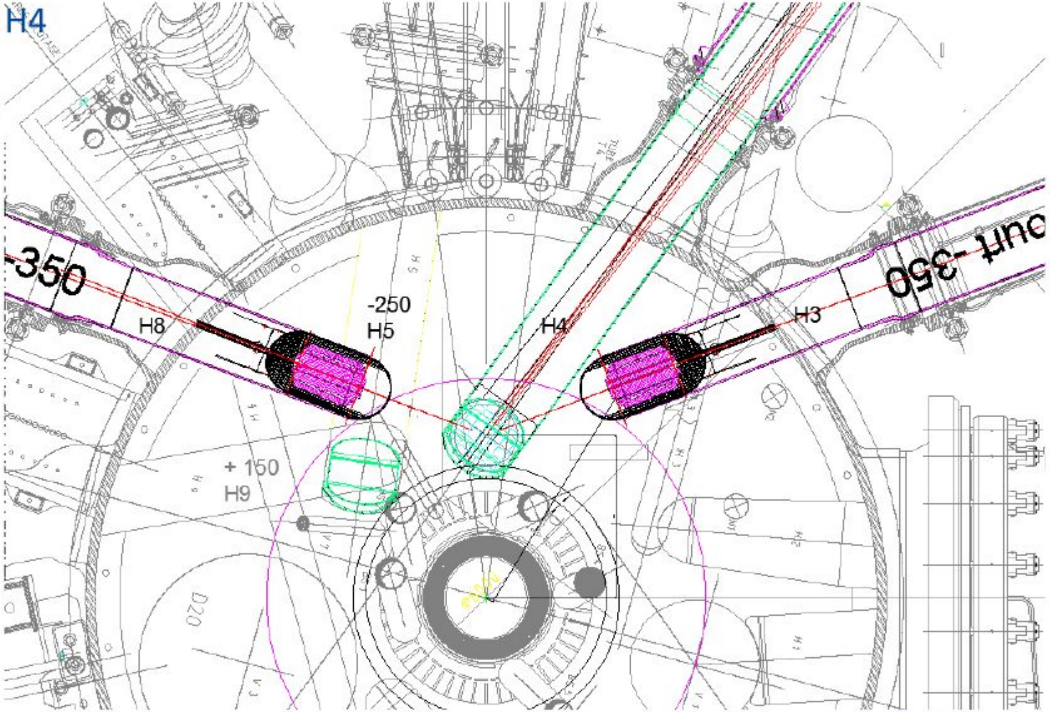
- (2002): Discussion of combining D4 with D9 as some kind of general-purpose hot-neutron diffractometer - it was never very clear what that meant in practice.
- (2005): A push by the Direction to consider moving D4 to the IH2 inclined beam tube.
- (2008): Overly imaginative+enthusiastic discussion of replacing the D4c microstrip detectors with a stretched-wire banana-geometry detector, which would have far less counting-rate stability.
- (2010): Consideration of returning to beam-sharing mode with IN1, which had been discarded in 1983 due to catastrophic background counts.
- (2006 - 2010): Promotion of the HiQ proposal for a PDF-analysis instrument on the IH2 inclined beam tube that would eventually replace D4.
- (2015): A brief/vague consideration of moving D4 to IH3 (or IH2?) once Brisp was dismounted.
- (2017): Proposal of a 3rd cold source that would remove the hot source as the first step.

Fortunately for D4, all of these ideas were unsuccessful in making it to the execution stage, perhaps because the instrument responsables of the hot-source instruments were not adequately consulted.

3rd Cold Source:

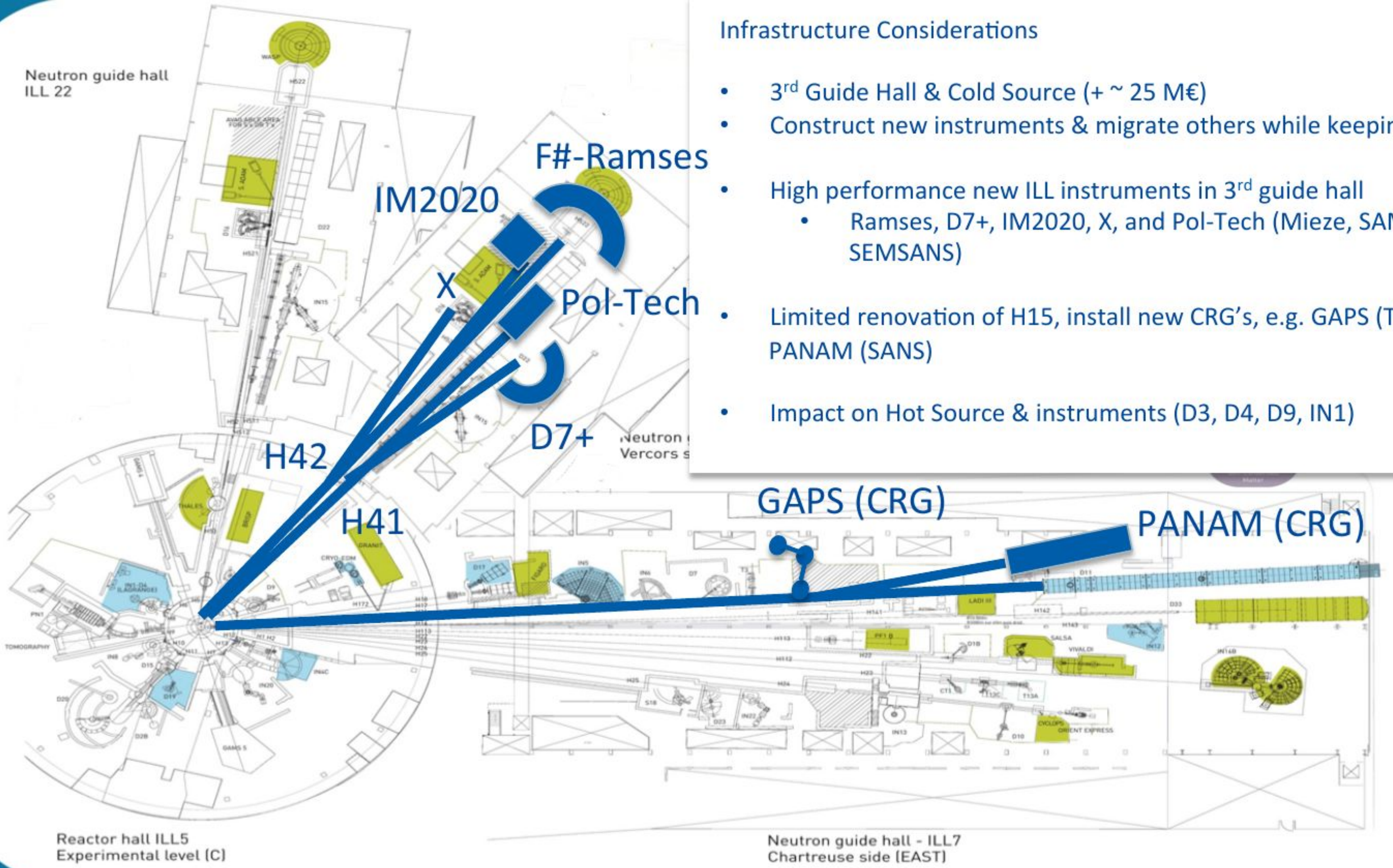
- Remove the Hot Source
- Copy-paste H5 like beam-tube cold HCS into H4
- Use H8 & H3 with in-beam-tube hot sources for D3, D4, IN1, D9
- Looks feasible
  .....but not fully worked out yet

Source: Charles Dewhurst's presentation to the ILL Scientific Council in spring 2017.



Infrastructure Considerations

- 3rd Guide Hall & Cold Source (+ ~ 25 M€)
- Construct new instruments & migrate others while keeping ILL7 on-line
- High performance new ILL instruments in 3rd guide hall
  - Ramses, D7+, IM2020, X, and Pol-Tech (Mieze, SAM, Mollie, SEMSANS)
- Limited renovation of H15, install new CRG's, e.g. GAPS (TAS) and PANAM (SANS)
- Impact on Hot Source & instruments (D3, D4, D9, IN1)



NEUTRONS FOR SCIENCE

Why not a 2nd hot source for the ILL?

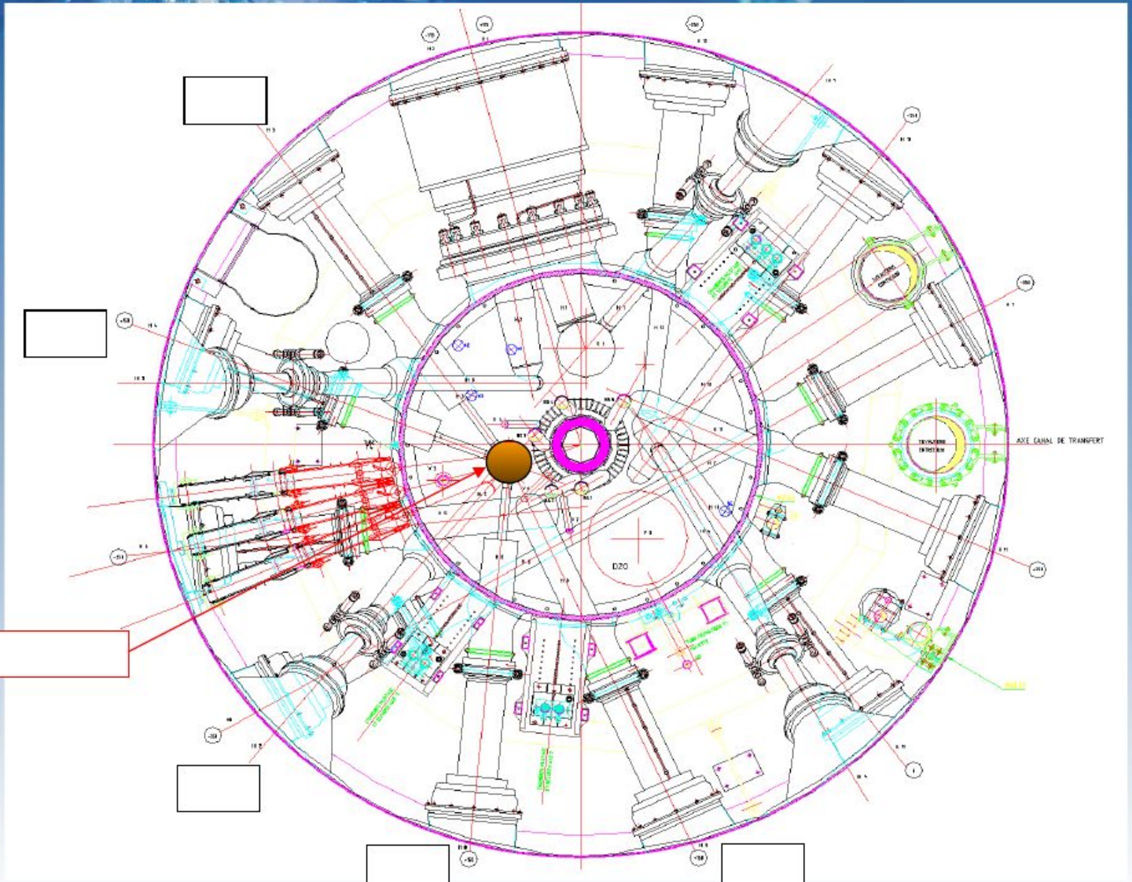
Benchmarking of ILL instruments with respect to the ESS

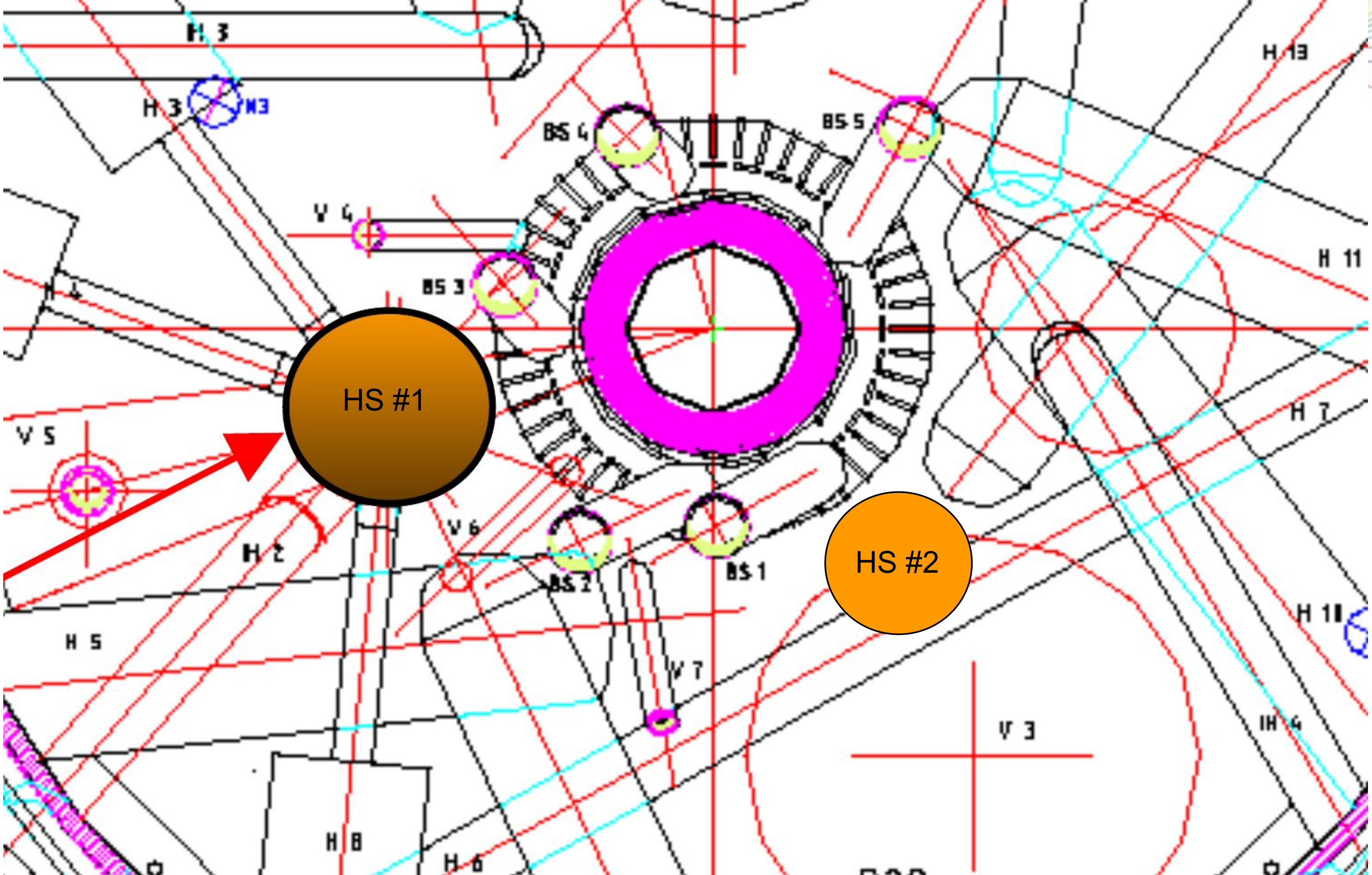
Version 19/11/09, compiled by Ken Andersen, Helmut Schober, Roland Gähler with input from many others

One of the deliverables of the ILL 20/20 program, funded by ESFRI, is the benchmarking of ILL instruments with respect to similar machines at future and present neutron sources. Such a document is also being requested by some of our associates. In particular, we have been asked to compare the performance of the ILL with the expected performance from the ESS. This document is intended to evolve with time. Critical input is highly encouraged.

Liquids diffractometer D4C	< 1	ESS has no hot source and therefore has about 10x lower time-integrated flux than the ILL at 0.5 Å. Building a crystal-monochromator instrument is therefore not interesting. Alternatively, a TOF instrument could try to benefit from the peak flux, which however is only about 3x that of the ILL for hot neutrons. That would require a pulse-shaping chopper. However, in order to fill the time frame between proton pulses, a large (>10) use of WFM would be required and most of the additional wavelength ranges would be much longer than the presently used wavelength range of 0.35 – 0.7 Å. Less than 1/3 of the time frame could be filled usefully, reducing the gain factor to below 1. The ILL hot-neutron instruments are those which are least favourable for moving to the ESS: there is no hot source, giving a strongly reduced peak flux compared to the thermal energy range, the repetition period is too long and WFM is not effective.
Hot-neutron single X D3, D9	< 1	Similar arguments as for D4.
Hot triple-axis IN1	<1	The TAS gain factor goes further down as the wavelength is decreased. In addition, as the ESS has no hot source planned, the time-integrated brightness falls considerably below that of the ILL. See description of D4C.

OPTIMISATION OF THE HOT NEUTRON SOURCE AND ITS BEAM TUBES H3-H4-H8



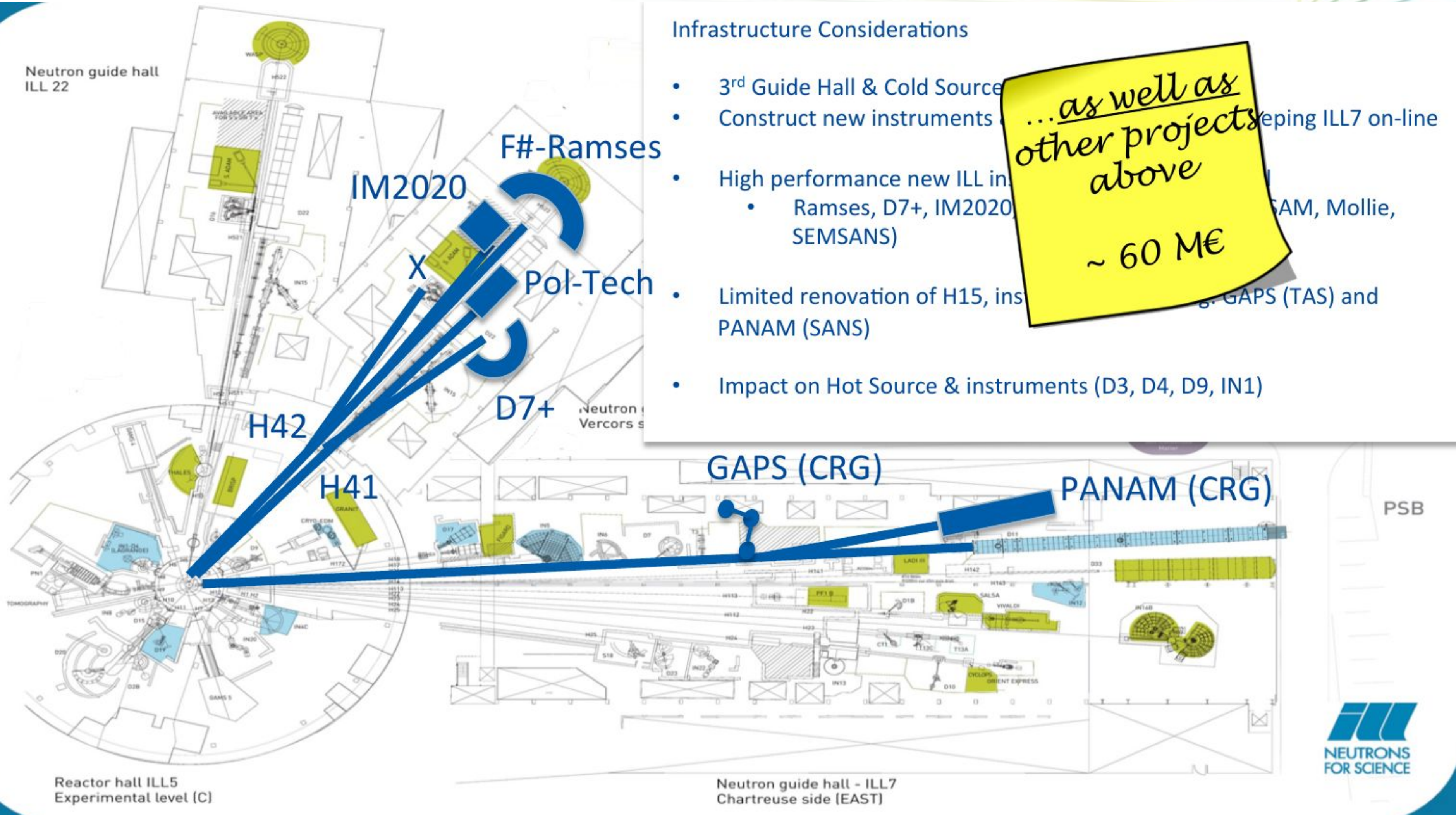

HS #1
HS #2
H 3
H 13
BS 4
BS 5
V 4
BS 3
H 11
V 5
H 7
V 6
H 2
BS 2
BS 1
H 5
V 7
V 3
H 8
H 6

Infrastructure Considerations

- 3rd Guide Hall & Cold Source
- Construct new instruments … eping ILL7 on-line
- High performance new ILL ins
    - Ramses, D7+, IM2020, … AM, Mollie, SEMSANS)
- Limited renovation of H15, ins … GAPS (TAS) and PANAM (SANS)
- Impact on Hot Source & instruments (D3, D4, D9, IN1)

…as well as other projects above

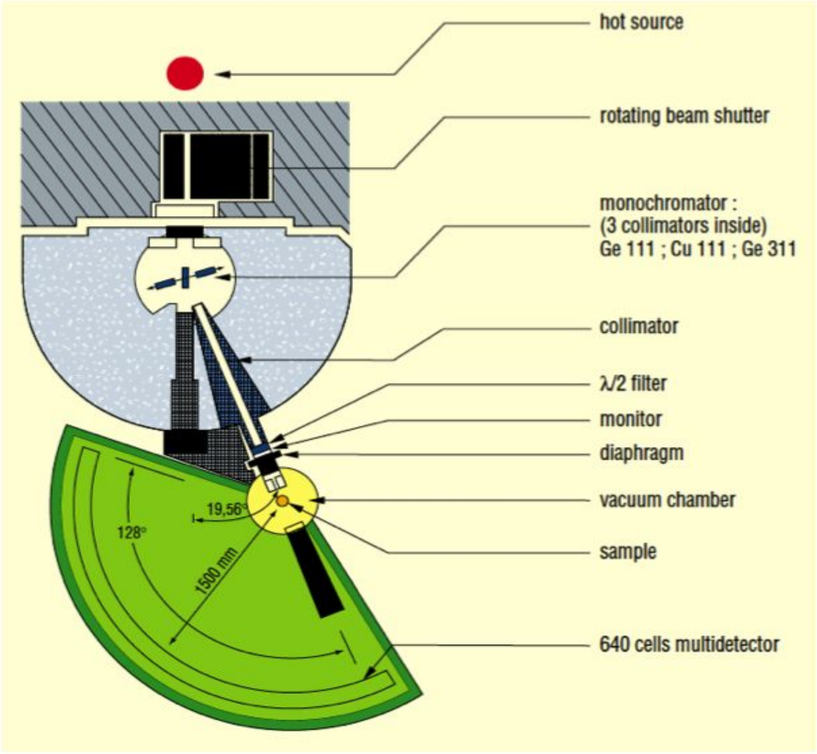
~ 60 M€

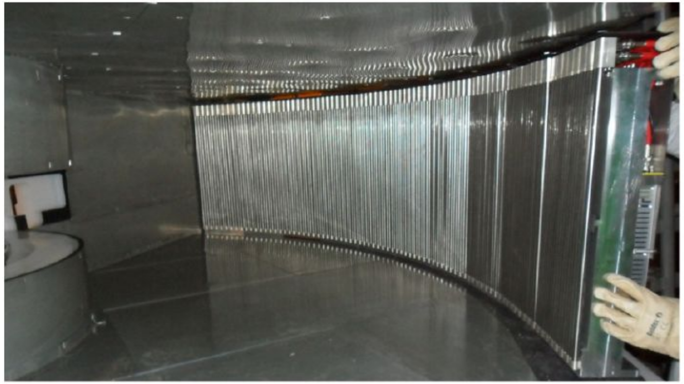


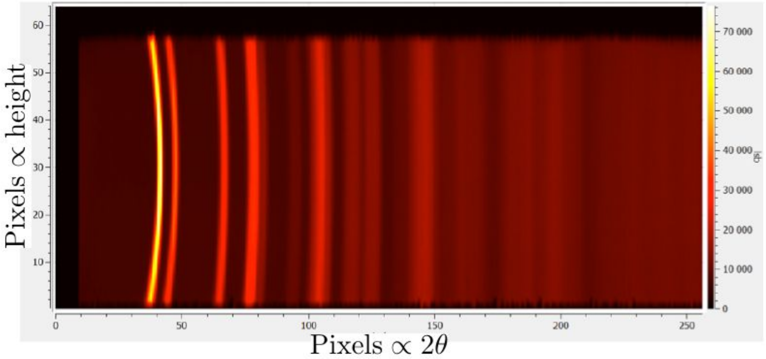
Or a ^{3}He-tubes detector à la 7C2 with narrower tubes for better 2θ-definition?

7C2, the new neutron diffractometer for liquids and disordered materials at LLB

G. J. Cuello[1], J. Darpentigny[2], L. Hennet[2,3], L. Cormier[4], J. Dupont[2], B. Homatter[2], B. Beuneu[2]







Brainstorming ideas (cont'd)

New high-temperature gas flow cell

A new "banana-like" detector could allow in situ experiments as those performed with this kind of reaction cells

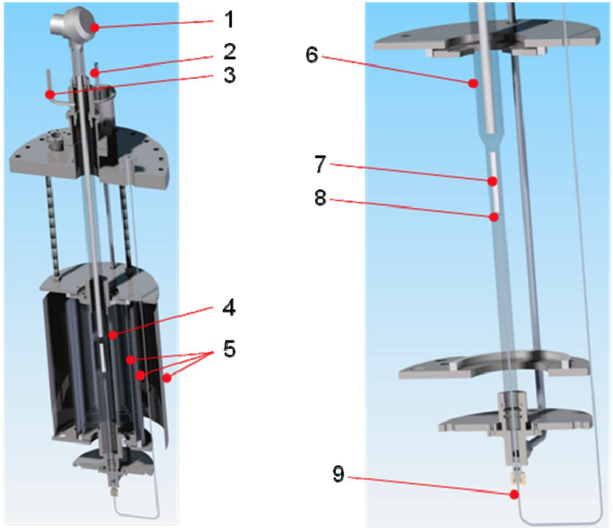


Figure 1. The schematic diagram of the flow-through gas cell (with the right hand portion highlighting the central portion of the left hand figure), showing (1) oxygen and temperature sensor; (2) gas inlet; (3) gas outlet; (4) heater element; (5) heat shielding; (6) quartz sample holder; (7) sample; (8) quartz frit; (9) thermocouple.

International Conference on Neutron Scattering 2009 IOP Publishing
Journal of Physics: Conference Series **251** (2010) 012090 doi:10.1088/1742-6596/251/1/012090

New high temperature gas flow cell developed at ISIS

R Haynes[1], S T Norberg[1,2], S G Eriksson[2], M A H Chowdhury[1], C M Goodway[1], G D Howells[1], O Kirichek[1], and S Hull[1]

[1] ISIS Facility, STFC Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire, OX11 0QX, UK
[2] Department of Chemical and Biological Engineering, Chalmers University of Technology, SE-412 96 Gothenburg, Sweden

E-mail: oleg.kirichek@stfc.ac.uk

Abstract. A flow-through quartz gas cell, together with a gas flow control and monitoring system, has been designed and constructed at ISIS. This equipment allows neutron powder diffraction data to be collected on samples at temperatures up to around 1300 K when exposed to user chosen mixtures of O_2, Ar, CO_2, and CO. By exploiting the sensitivity of neutrons to the presence of light atoms such as oxygen, it is possible to probe the crystal structure of oxide materials as a function of oxygen partial pressures down to $\log_{10}p(O_2)$ of about -20. The resultant structural information can then be correlated with the bulk properties of the materials, whose research and technological interests lie in fields such as energy production, storage materials, catalysis, and earth science.

Many thanks for your attention!

INSTITUT LAUE LANGEVIN

NEUTRONS FOR SOCIETY

APPENDICES

SUMMARY OF SCIENTIFIC ACTIVITY

Strengths and weaknesses of the science program and user base + 2 highlights from 2020

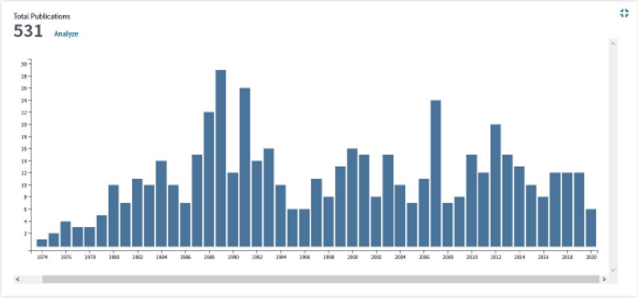
- For decades, D4 has seen a continued turnover of new, good science, and new users.
- Frequently new, unanticipated applications of total scattering techniques.
- The high counting rate and superb stability in background counts.
- The impressively increasing fraction of PDF-analysis experiments (nano/disordered crystals).
- In particular over the past 5 years, Magnetic PDF-analysis has developed a lot at D4.

- Increasing number of new/inexperienced users requiring very much time+help+effort.
- Much more administrative workload (e.g. PhD recruitment, sending samples, contracts, etc).
- New, stricter safety rules, further consuming the time of instrument responsibles.
- More EASY/DIR experiments, and now doing the experiments alone during Covid.

- Magnetic PDF-analysis on the magnetically non-ordering $NaYbO_2$, showing very interesting results for the dynamic local spin-spin correlations between Yb ions.
- Total scattering on amorphous materials $(PhSn)_4S_6$ exhibiting white light emission. This emitted light is highly directional and has a brilliance comparable to a Laser.

STATISTICS

Overload factors, experiments performed and publication output for the last 5 years

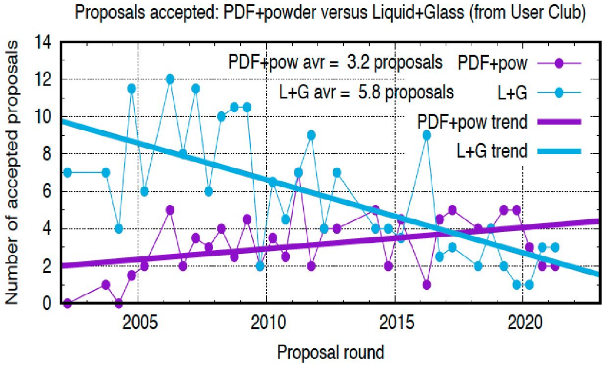


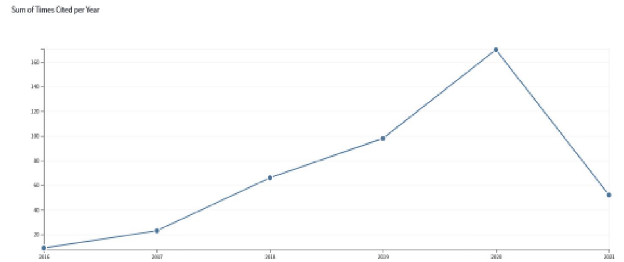
H-index = 69 – highest at ILL, when corrected for D4 as ½-time instr.

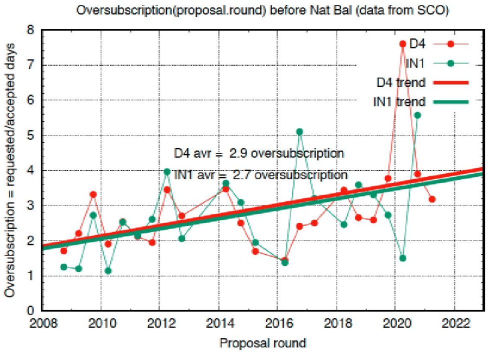
Average cites/pub = 34

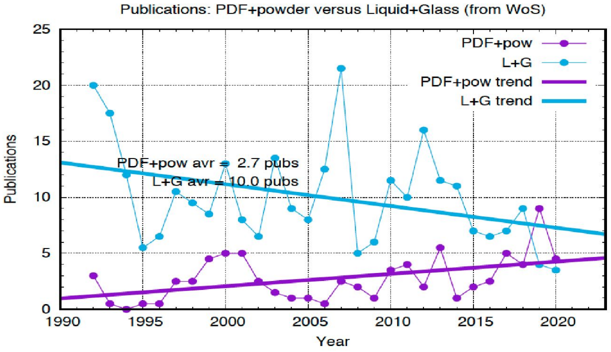
5-yr impact factor = 3.4

Average pubs/y = 10









FAILED EXPERIMENTS or UNPUBLISHED DATA

Number of and reasons for failures and bottlenecks for scientific output

- **Lack of sufficient scientific man-hours competent in doing experiments at D4 and in analysing the Total-Scattering data using Fourier techniques**:

  The Graduate School takes up ~50% of Henry's time, Gabriel as the only other scientist competent to do PDF-analysis also does many experiments at other ILL instruments, and the increasing, now majority, fraction of new or inexperienced users at D4 (see written report) requires very much time/effort/help from Henry+Gabriel. Finally, new/inexperienced users also tend to publish less, largely due to lack of familiarity or sufficient confidence with Fourier techniques in diffraction.

- **Up until ~ 4 years ago, D4 could claim the following publication pipeline throughput:**

~ 95% of D4 experiments are successful, i.e. produce good raw data.

~ 85% of the raw data are fully analyzed (normalization, MS+attenuation corrections, etc).

~ 85% of the fully analyzed data are finally submitted for publication (~ all finally accepted).

Thus 0.95 * 0.85 * 0.85 = ~70% of D4 experiments are finally published (Δt_pipeline = 3+ years).

The present lack of D4-scientist man-hours has strongly reduced the first 0.85 factor !!!

SUMMARY OF RECENT TECHNICAL DEVELOPMENTS

Improvements/upgrades - sample environment, instrument control, data treatment, etc

- New vanadium tail (from UK) for the D4-specific cryofurnace (2016).
- Alpha1 incident beam collimation in place and tested successfully (2016).
- Isostatic mechanical support for the air-pad system under the detector bloc (2017).
- Rewiring of the instrument: detector and motor cables principally (2017).
- New FPGA detector signal processing, installed under the detectors (2017).
- Successful first ASI/DSI texts and certification of instrument (spring 2018).
- Updates+improvements to the D4-version of the data-analysis program CORRECT, making it much more user-friendly and efficient, remaining of course command-line/script driven (2019).
- Start including D4 data treatment into MANTID (2020).

FUTURE DEVELOPMENTS, UPGRADES

Projects that do not fit into the operation budget of the instrument or Endurance

- 2022: Design and installation of an incident beam collimation for the sample bell jar, similar to that which was successfully done for the D4 cryofurnace, thus reducing background and also allowing a lower Q_{min} by a factor of 1.5 or so. Depending on the cost, the opening of this collimator could be motorised.
- 2022: Design and installation of a sample changer for our cryofurnace, based on the idea of that developed for IN1 by Alain Bertoni. For D4 the system could be simpler, because we plan to have 4 to 6 positions only. This simplifies the geometrical constraints.
- 2022: Design of a sample changer for ambient conditions (i.e., room temperature and vacuum). The idea is to have a sample changer with 4 to 6 positions, adapted to a new cover plate (i.e. bell-jar lid). This will allow interchanging with the present lid for delicate experiments where small changes in the background could be annoying.

If Alain has not enough time for the three projects, the third one could be delayed to 2023.
We remind you that, ***in the best case, D4 obtains at most 50% of Alain's working time.***

Brainstorming ideas

New multidetector

VI European Conference on Neutron Scattering (ECNS2015) IOP Publishing
Journal of Physics: Conference Series **746** (2016) 012020 doi:10.1088/1742-6596/746/1/012020

7C2, the new neutron diffractometer for liquids and disordered materials at LLB

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