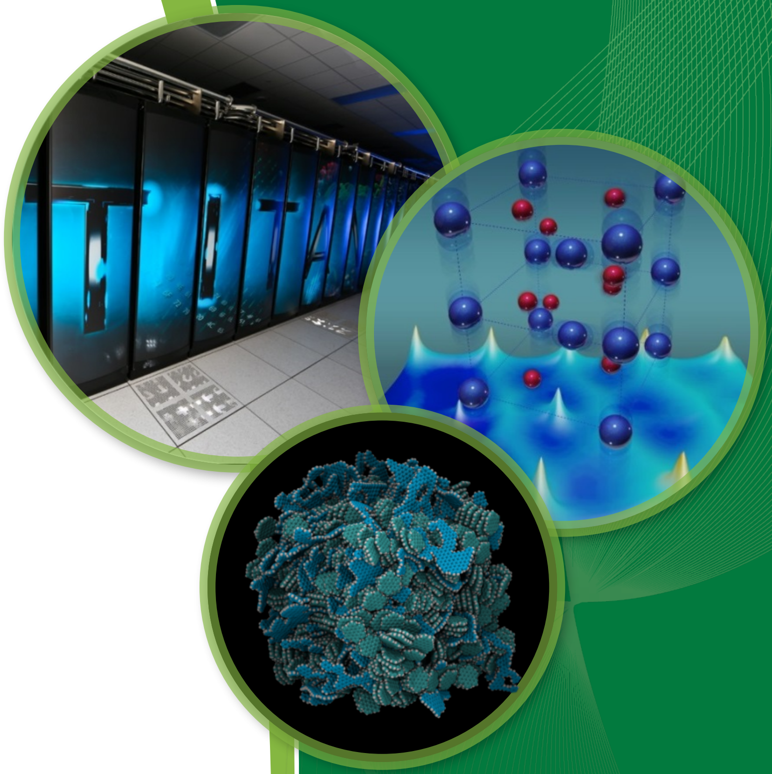
Some
– more or less random –
thoughts related to neutron spin echo spectroscopy



Georg Ehlers

Oak Ridge National Laboratory

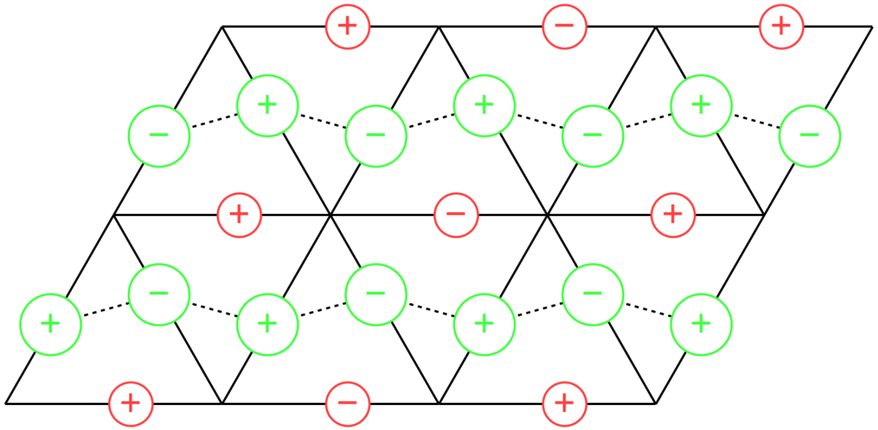
Outline

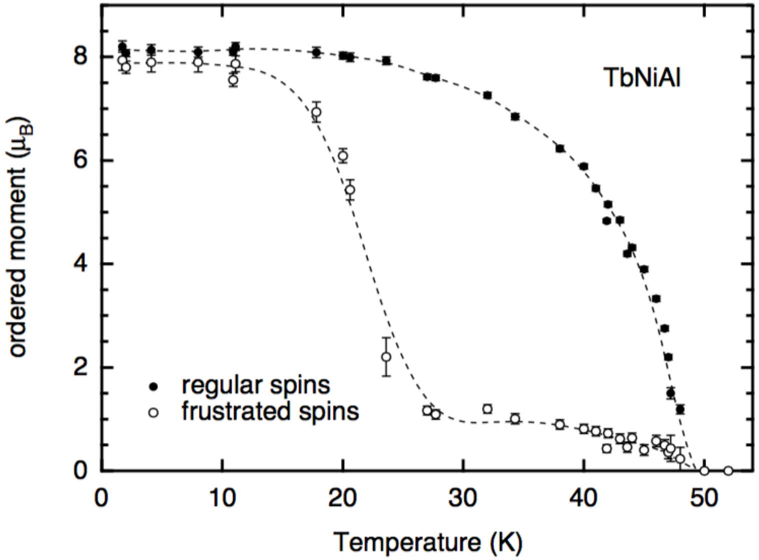
- About myself
- NSE at reactor and pulsed sources
- Complementarity with other techniques in the energy domain
- “Odd” things to measure

Career Path Step 1 – Helmholtz Zentrum Berlin

Career Path Step 1 – Helmholtz Zentrum Berlin

1993 – 1996 PhD in neutron scattering (mostly powder diffraction)





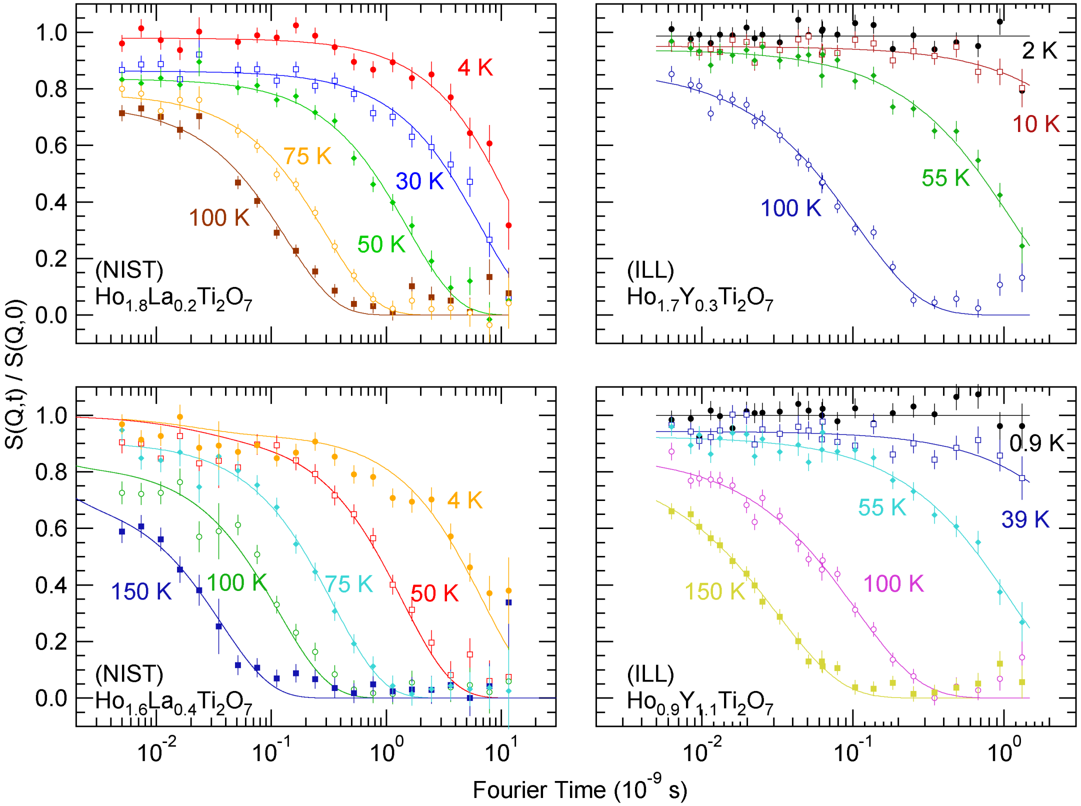
G. Ehlers et al., PRB **63** 224407 (2001)

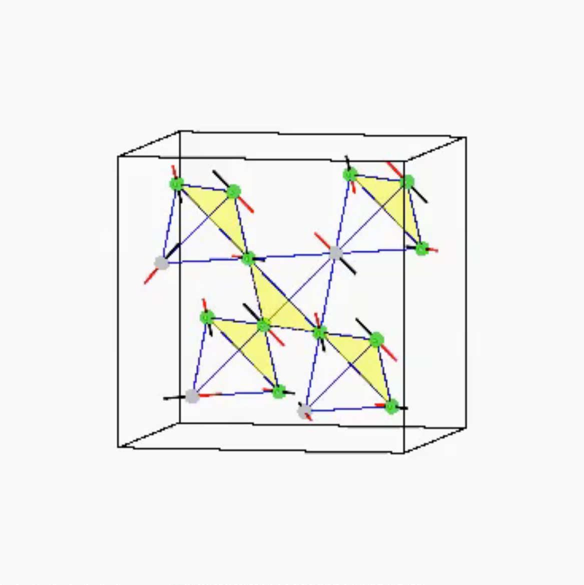
Career Path Step 2 – Institut Laue-Langevin

Career Path Step 2 – Institut Laue-Langevin

1997 – 2003

Instrument Scientist at a Spin Echo Spectrometer (IN11 & IN15)



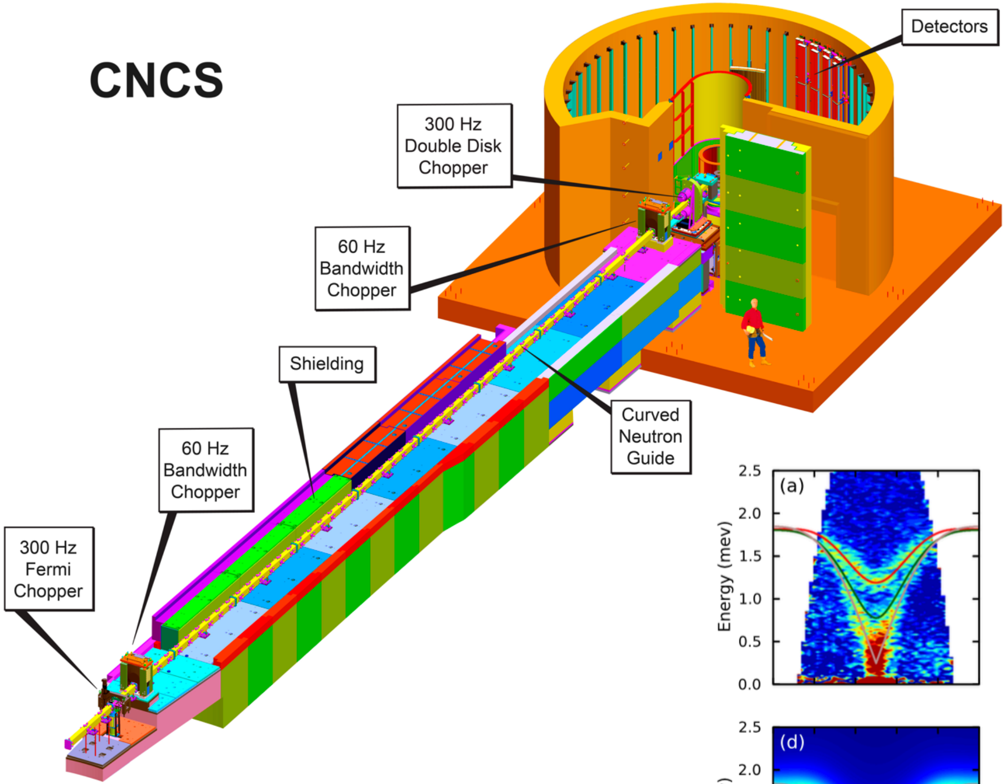


Career Path Step 3 – Oak Ridge National Lab

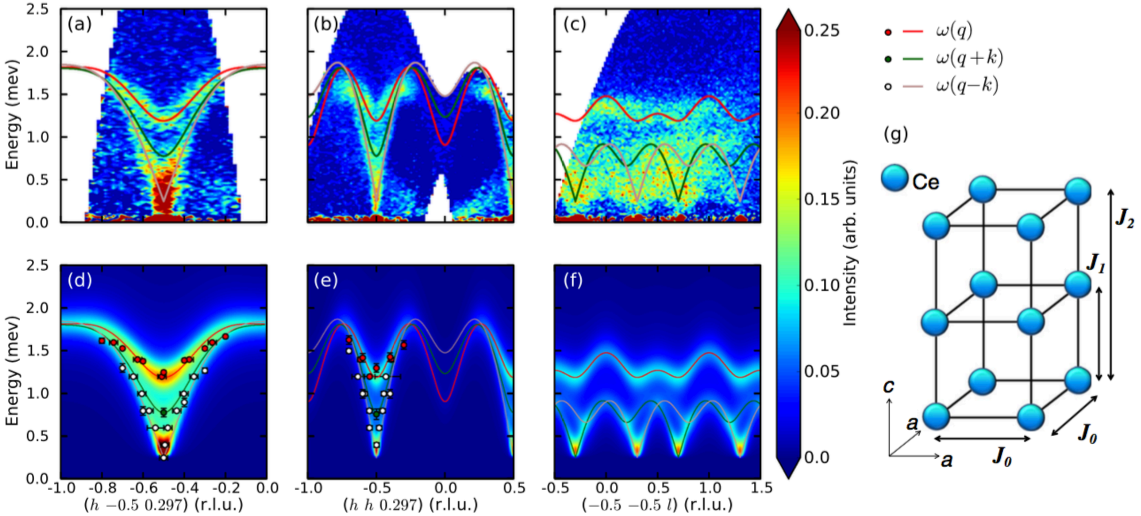
Career Path Step 3 – Oak Ridge National Lab

2003 – present	Instrument Scientist at a Time-Of-Flight Spectrometer Built and Operate CNCS at ORNL

CNCS



P. Das et al., PRL **113** 246403 (2014)



NSE as a FOURIER technique

In the scattering process the neutron exchanges momentum and energy with the sample. These are picked up as changes in direction and speed. The strong property of neutron scattering is that these two quantities are measured simultaneously (as opposed to, for example, local probes).

In order to measure a change, conventional wisdom dictates that these quantities need to be established independently before and after scattering (we want to calculate a difference). The finer the difference desired, the less intensity one is left with.

NSE as a FOURIER technique

The basic idea of spin echo is to use the neutron spin to intrinsically encode direction or speed on both sides of the sample. The difference then can be determined, on average for the beam ensemble, without knowing the actual numbers for a neutron individually.

This can be done for the energy (speed) and momentum (direction).

In my talk I will essentially only consider the former case, but both cases will be dealt with by others.

NSE as a FOURIER technique

The benefit is that the reciprocal correlation between resolution and intensity is broken. One can have high intensity and high resolution at the same time.

The drawback is that polarized beam is required, which costs intensity in itself and adds complexity.

We will spend one week here of discussing the ins and outs of all this.

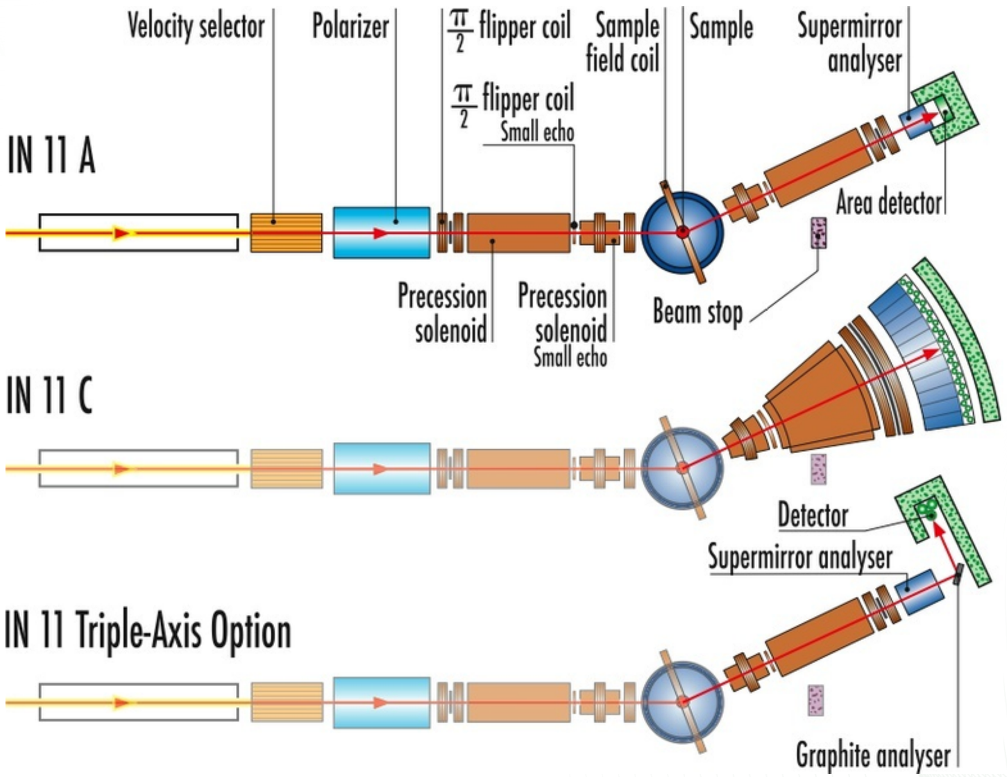
The Place of NSE in the neutron scattering landscape

Things to note:

- Energy resolution can be pushed to <1 neV but doesn't have to be (can be relaxed to 1 meV).
- Measures in the **time domain**.
- Requires polarized neutrons (that's good or bad depending on what you are after).
- In practice, **so far**, it has been proven to work best for quasi-elastic scattering. However, inelastic scattering is also possible (this is perhaps a bigger area for **future growth**).

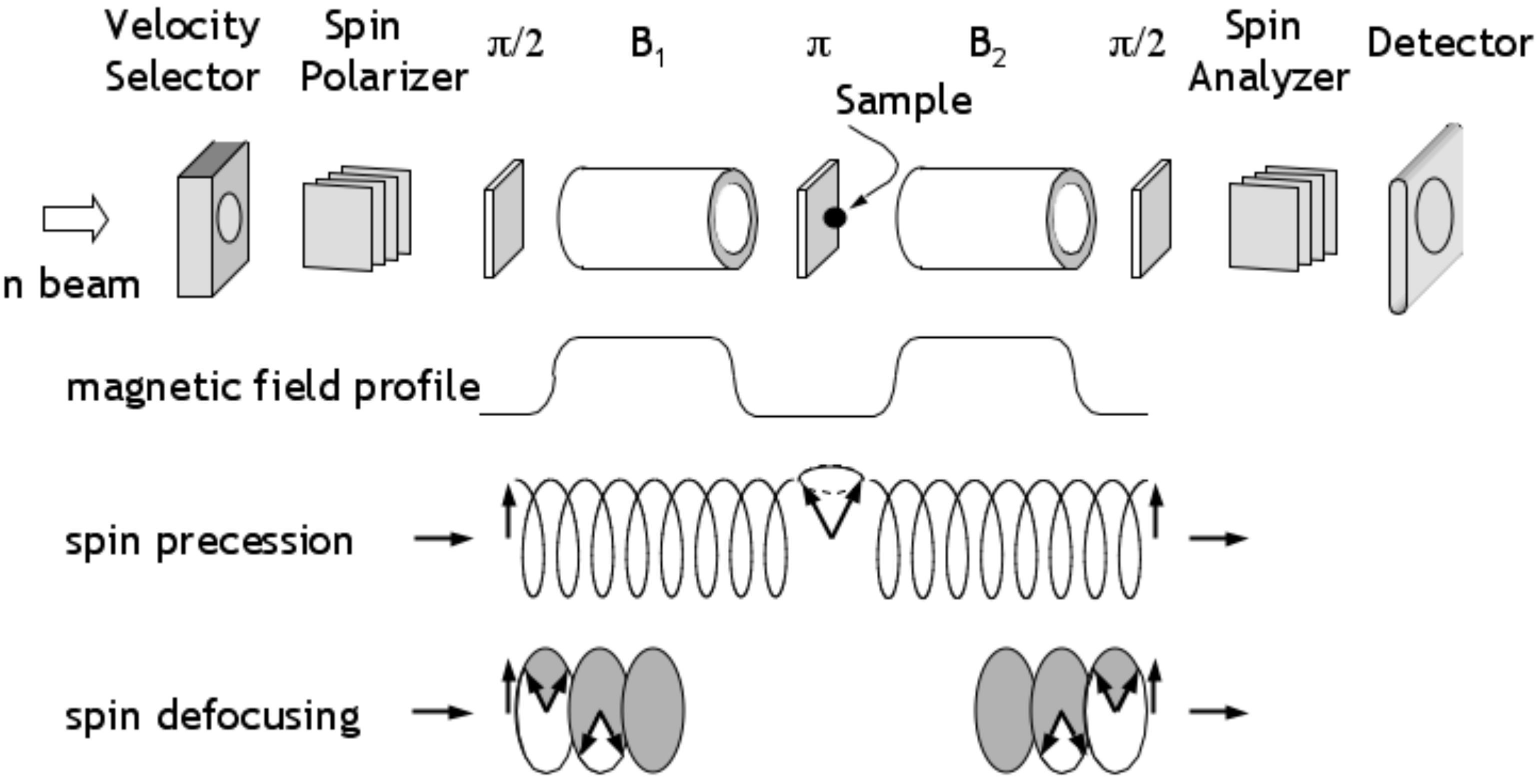
Reactor vs Pulsed Source

NSE at reactor source

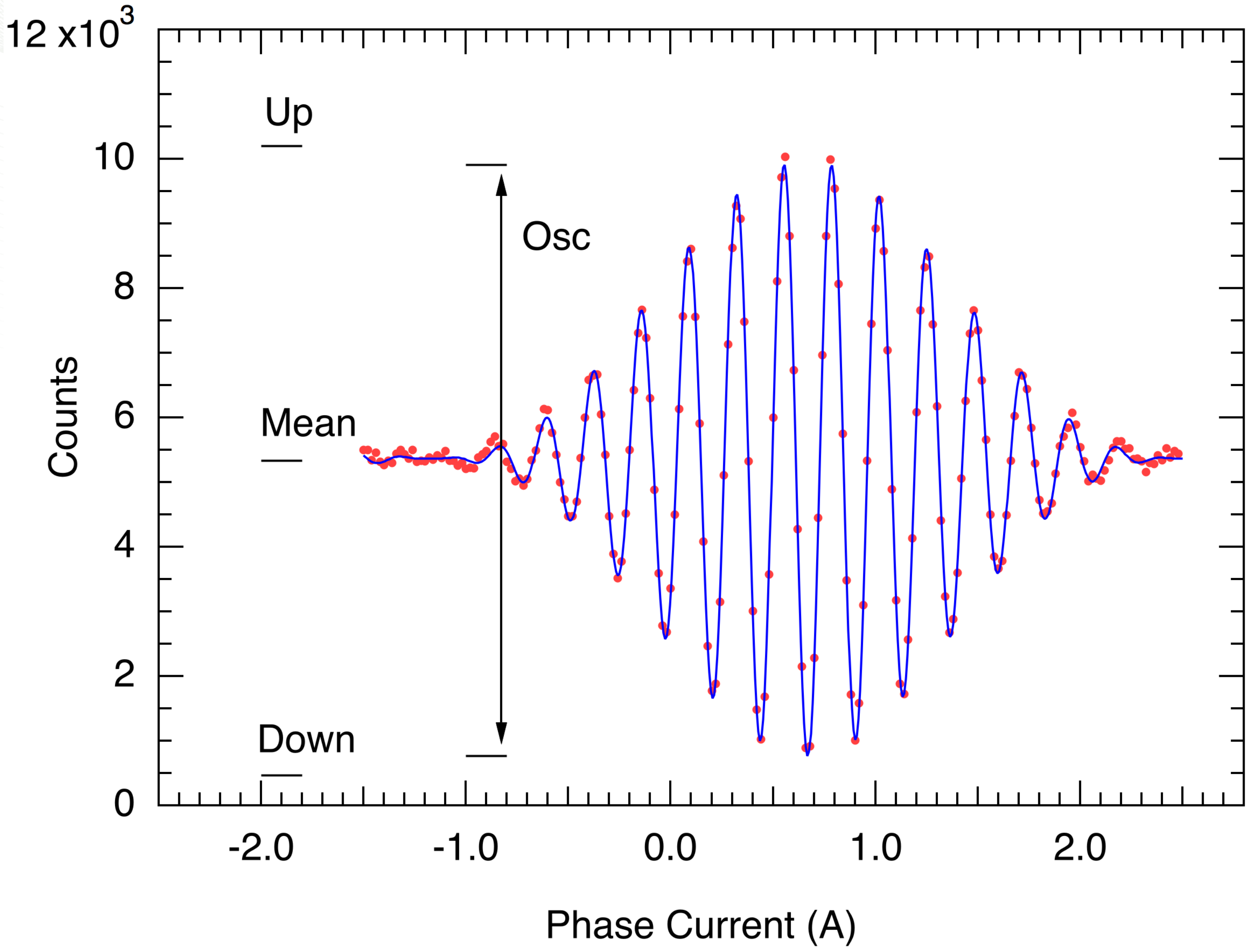


https://www.ill.eu

NSE as it was first realized



Echo Function with monochromatic beam



NSE at reactor source (NIST)

NSE at reactor source (HZB)

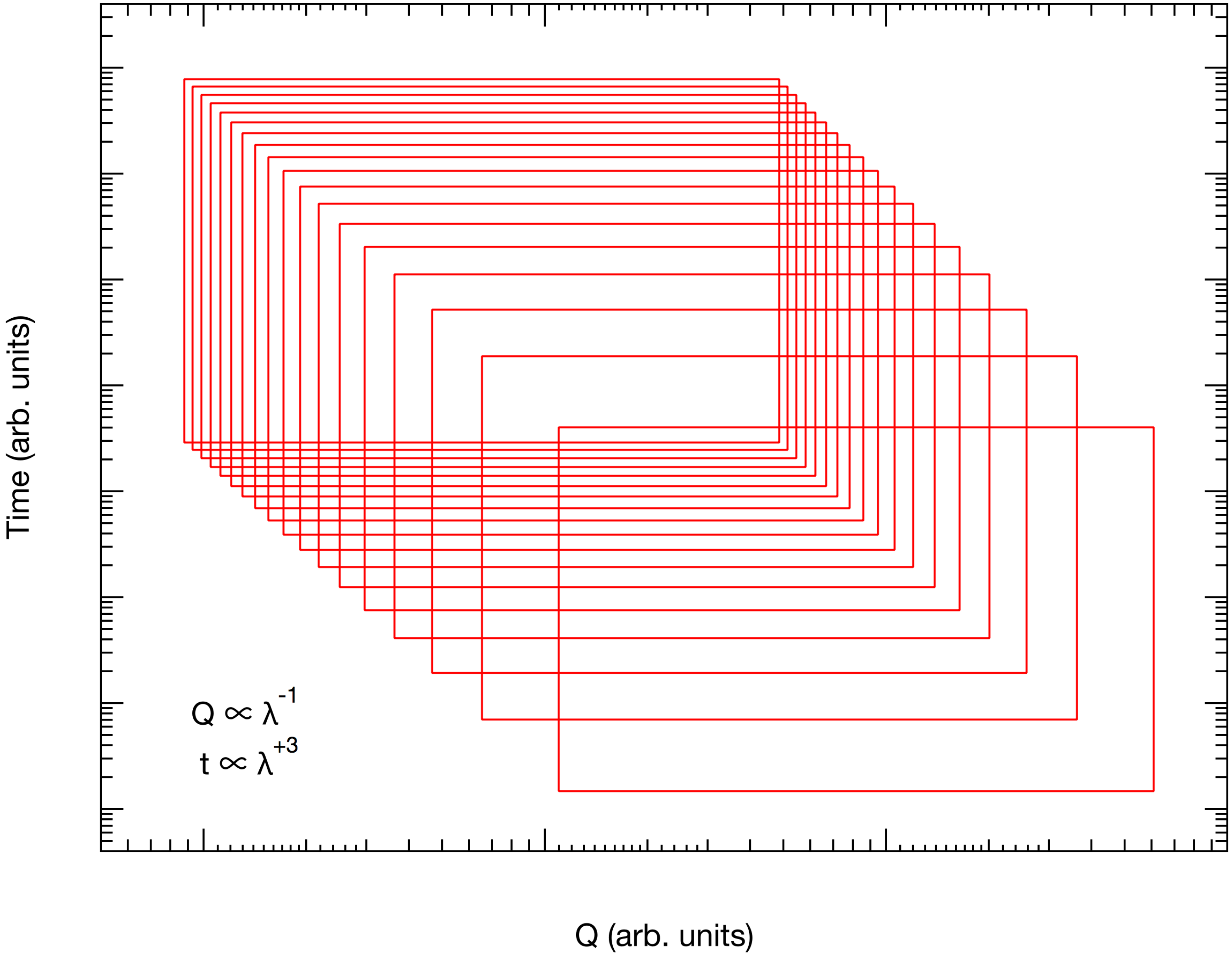
Spin Echo Spectrometers at Reactor Sources

- IN11, IN15 @ILL (IN20 used to have an option, WASP is being built)
- JNS, RESEDA @FRM-II
- MUSES @LLB
- NG-5 @NIST
- iNSE @JRR-3M
- TRISP, MIRA @FRM-II
- FLEXX @HZB (SPAN has been decommissioned)

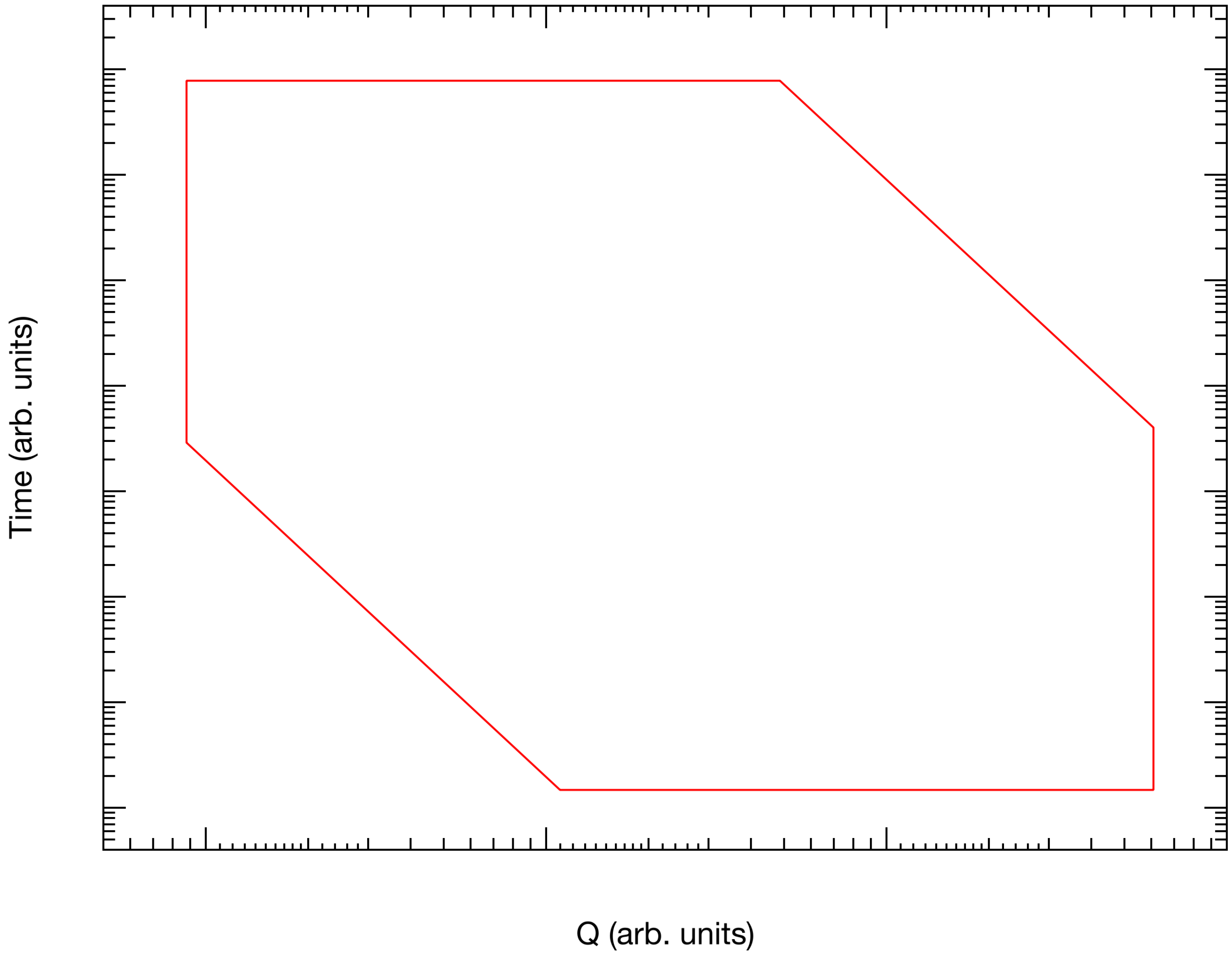
Spin Echo Spectrometers at Pulsed Sources

- SNS-NSE @SNS
- VIN-ROSE @J-PARC
- OFFSPEC & LARMOR @ISIS

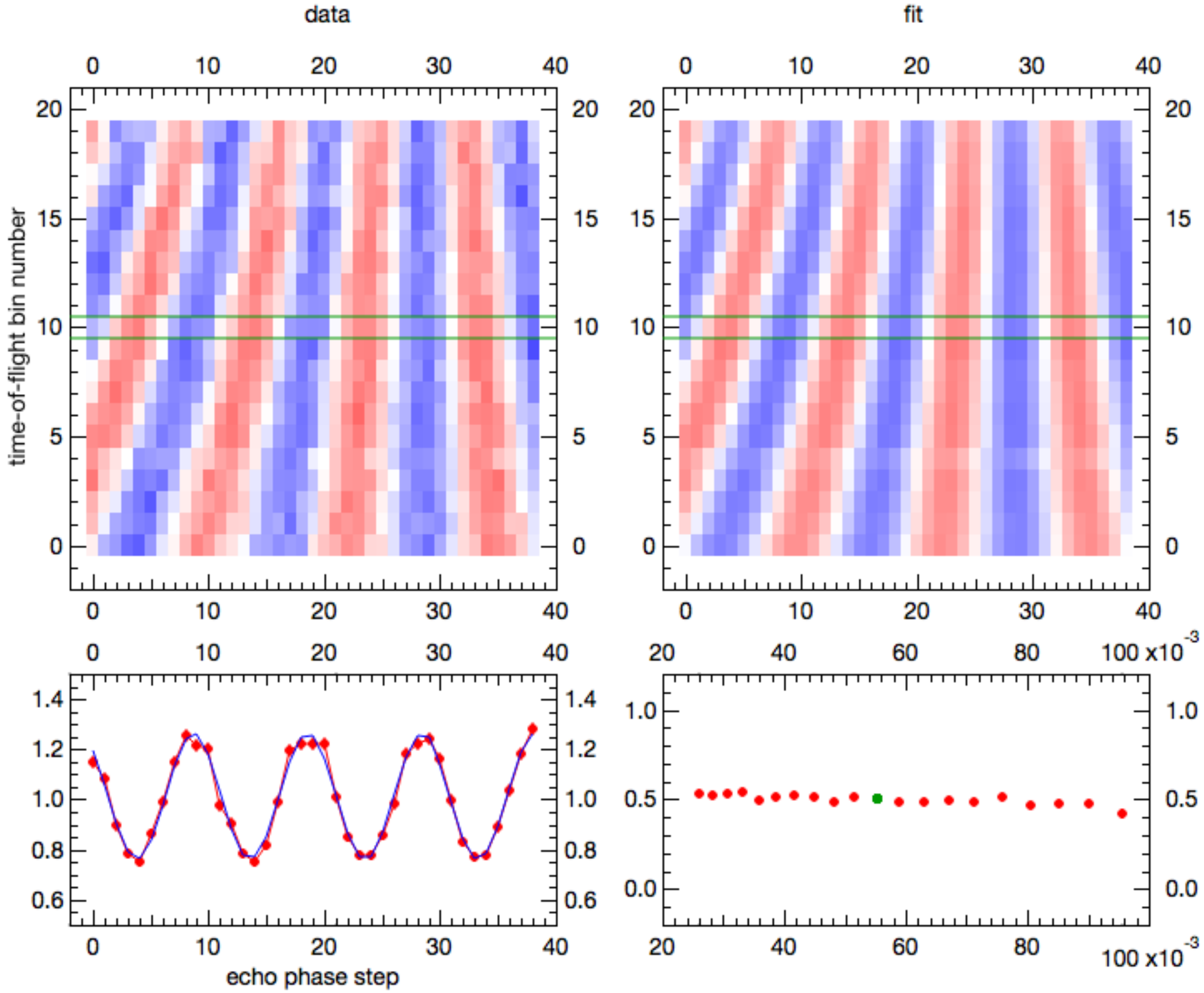
S(Q,t) area coverage at a reactor source



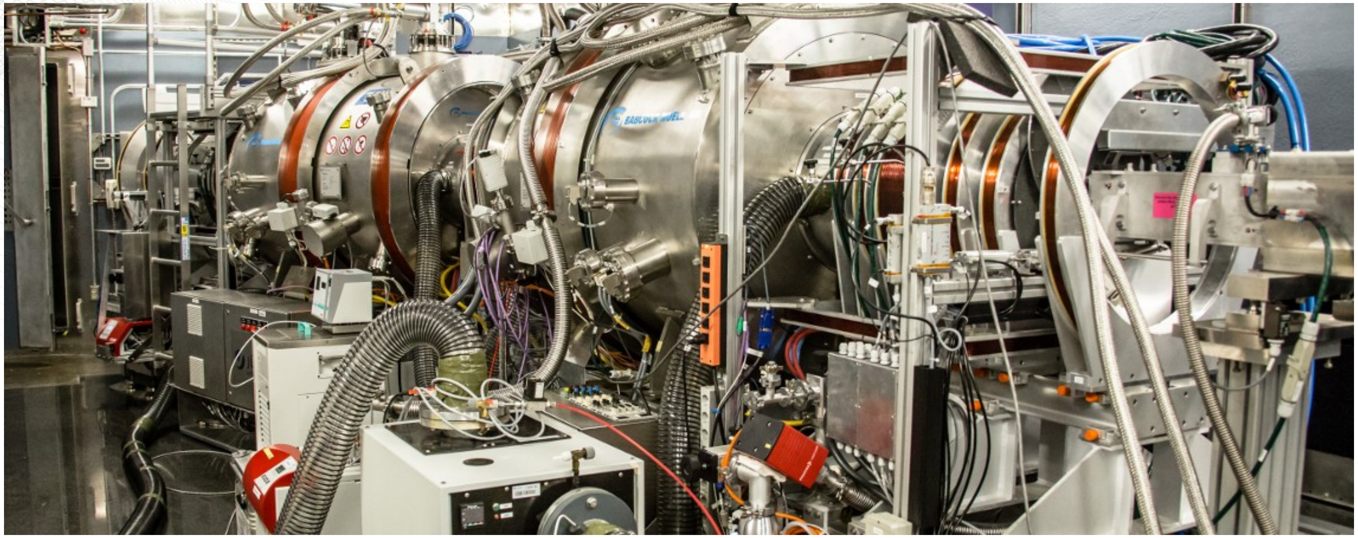
S(Q,t) area coverage at a pulsed source



Echo Function with polychromatic beam



Such installations exist …



http://neutrons.ornl.gov/nse

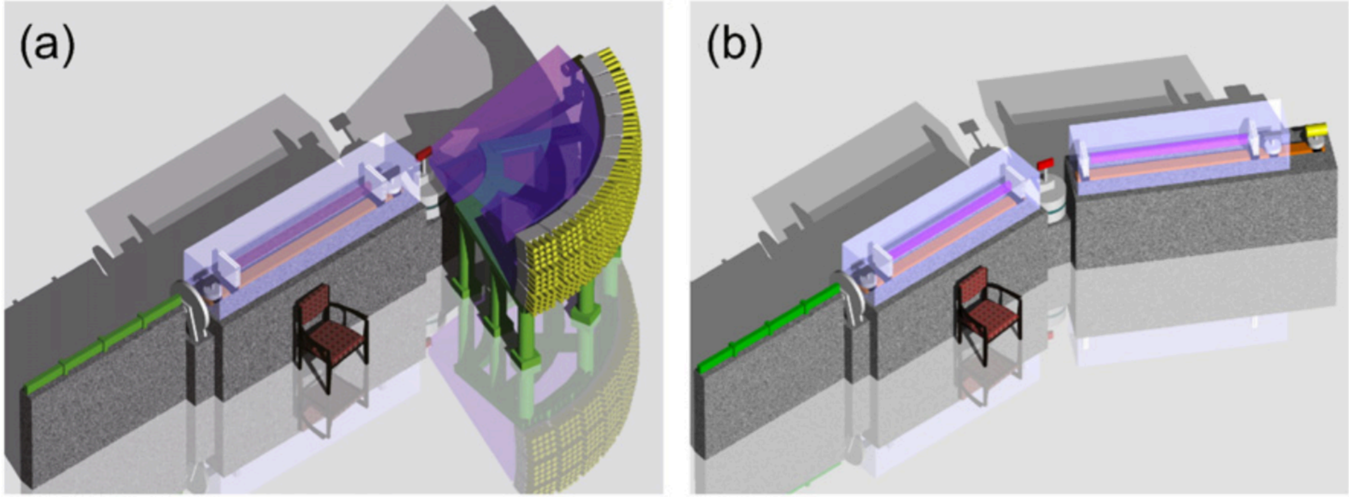
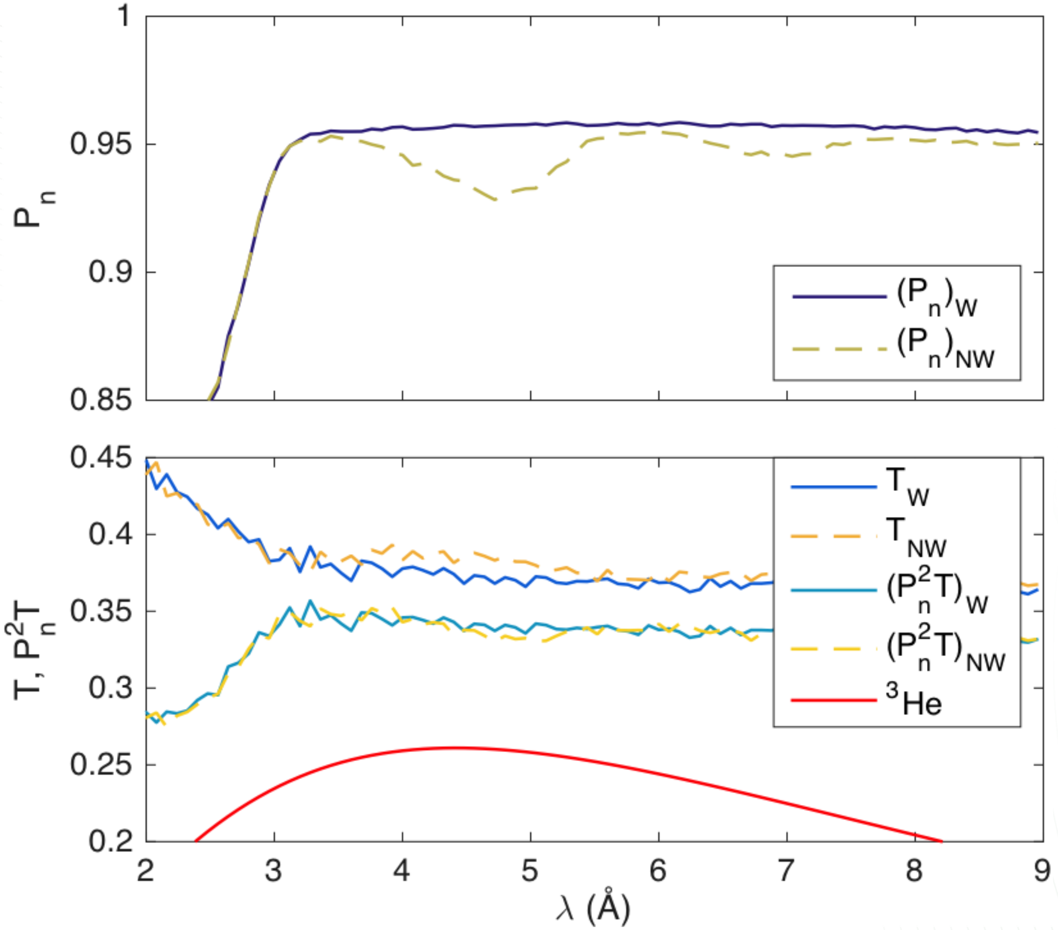


Fig. 13. Schematic view of VIN ROSE. (a) MIEZE type, (b) NRSE type.

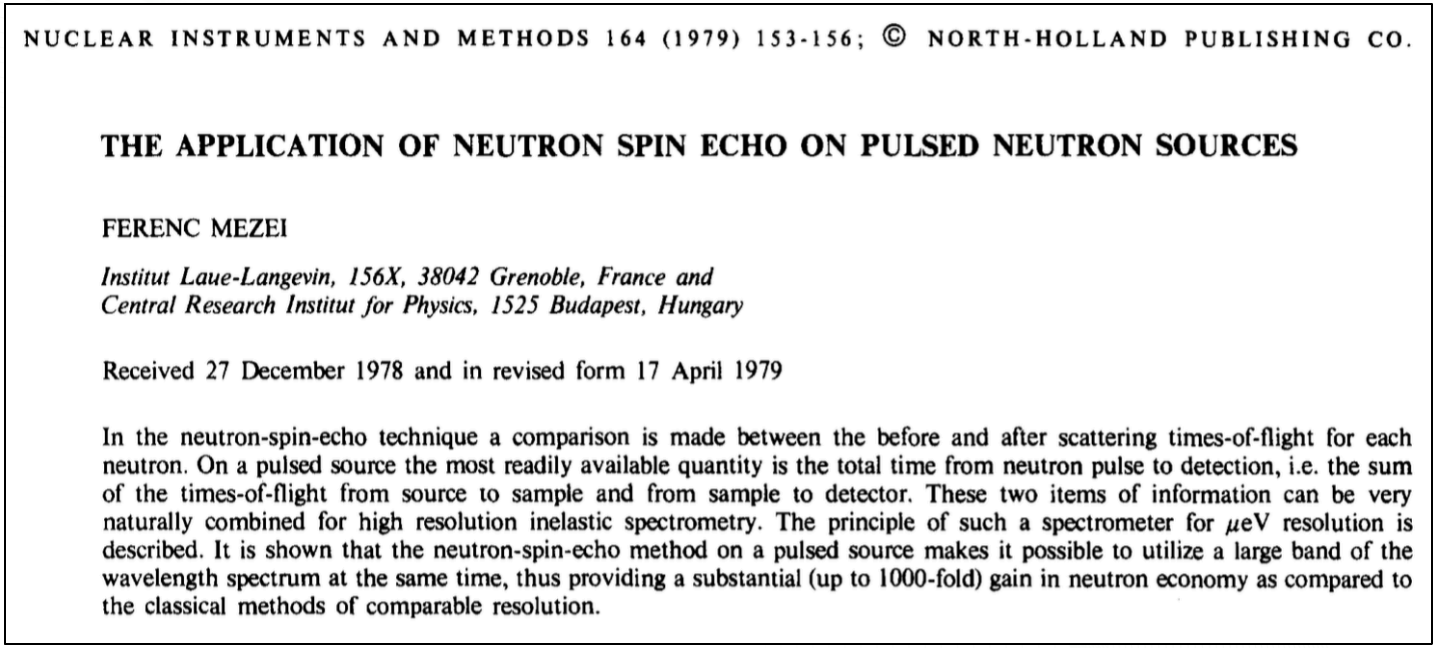
H. Seto et al., BBA **1861** 3651 (2017)

Wide-band polarizer performance



Courtesy: G. Nilsen

NSE at a pulsed source first proposed in 1979



NUCLEAR INSTRUMENTS AND METHODS 164 (1979) 153-156; © NORTH-HOLLAND PUBLISHING CO.

THE APPLICATION OF NEUTRON SPIN ECHO ON PULSED NEUTRON SOURCES

FERENC MEZEI

Institut Laue-Langevin, 156X, 38042 Grenoble, France and
Central Research Institut for Physics, 1525 Budapest, Hungary

Received 27 December 1978 and in revised form 17 April 1979

In the neutron-spin-echo technique a comparison is made between the before and after scattering times-of-flight for each neutron. On a pulsed source the most readily available quantity is the total time from neutron pulse to detection, i.e. the sum of the times-of-flight from source to sample and from sample to detector. These two items of information can be very naturally combined for high resolution inelastic spectrometry. The principle of such a spectrometer for μeV resolution is described. It is shown that the neutron-spin-echo method on a pulsed source makes it possible to utilize a large band of the wavelength spectrum at the same time, thus providing a substantial (up to 1000-fold) gain in neutron economy as compared to the classical methods of comparable resolution.

NSE at a pulsed source

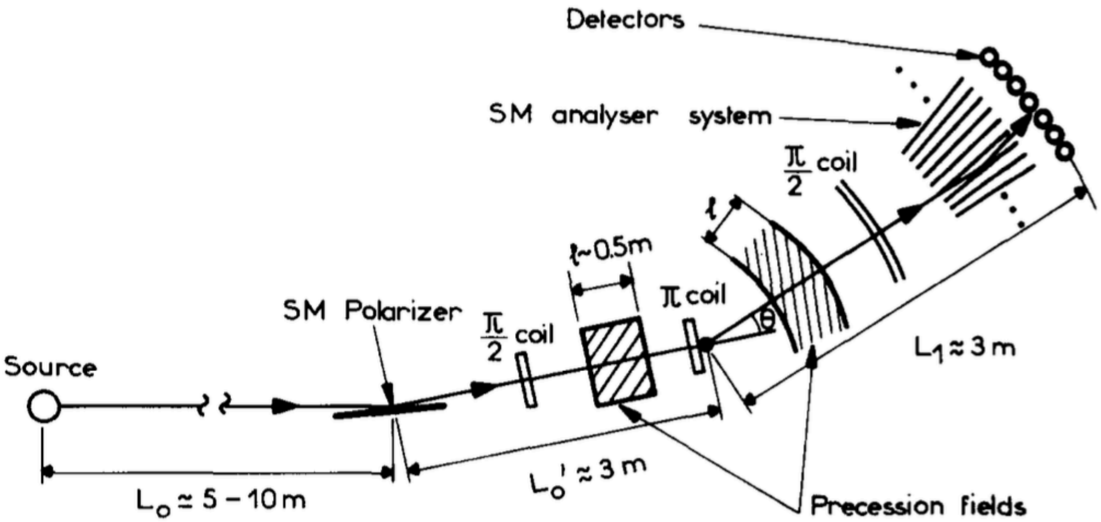


Fig. 1. Schematic lay-out of a neutron spin echo high resolution inelastic spectrometer on a pulsed neutron source.

Mezei, 1979

Inelastic TOF NSE

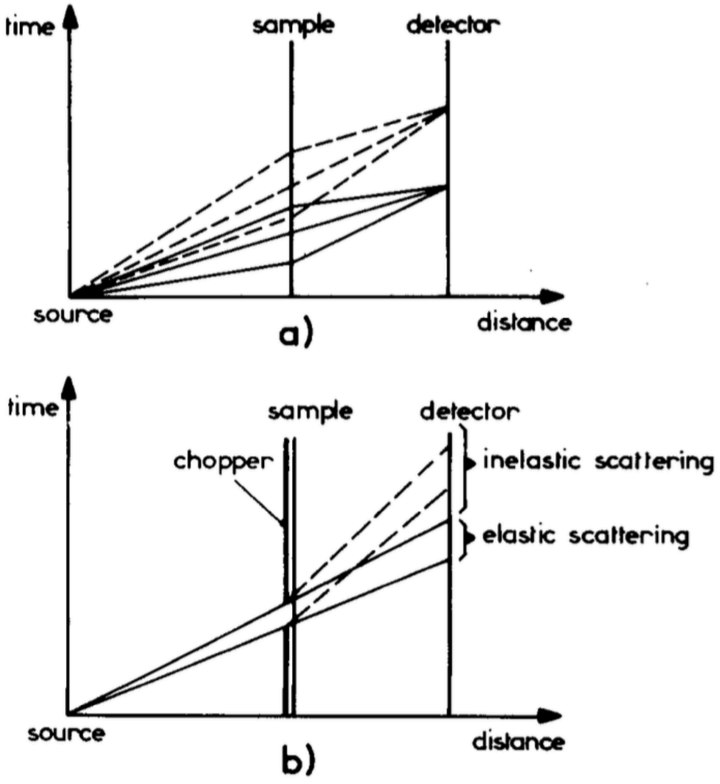
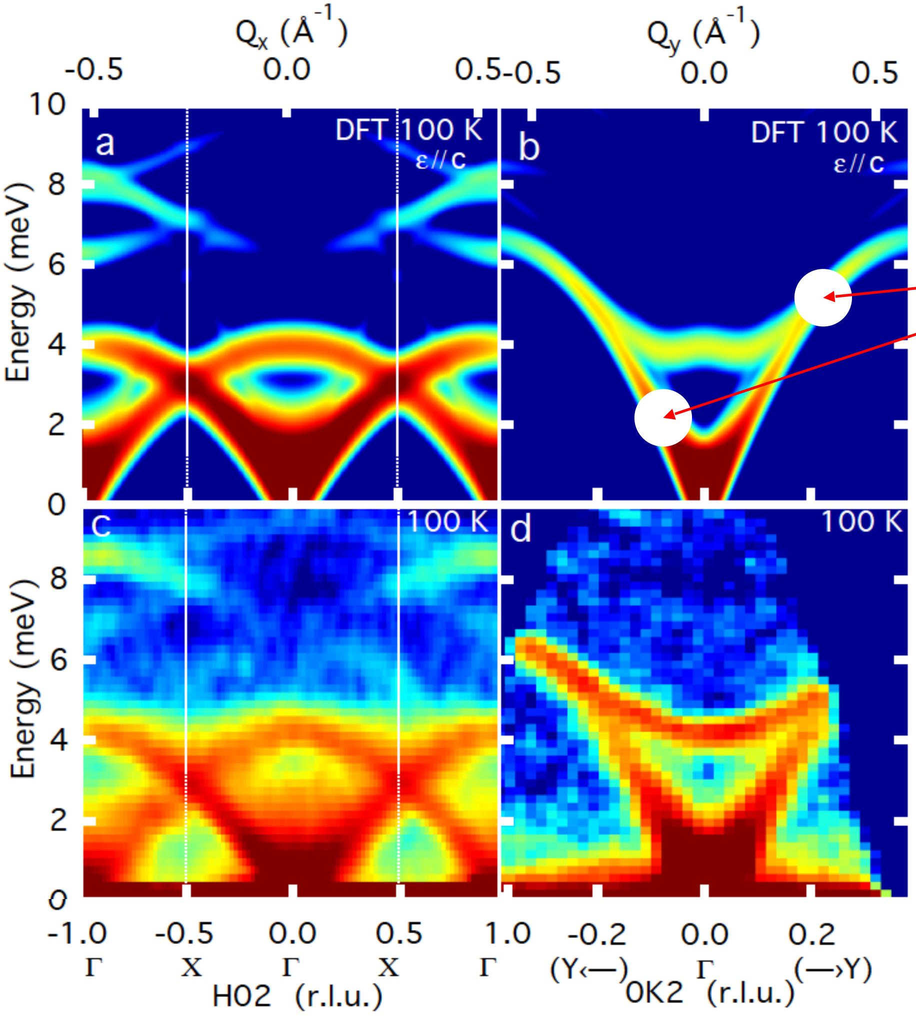
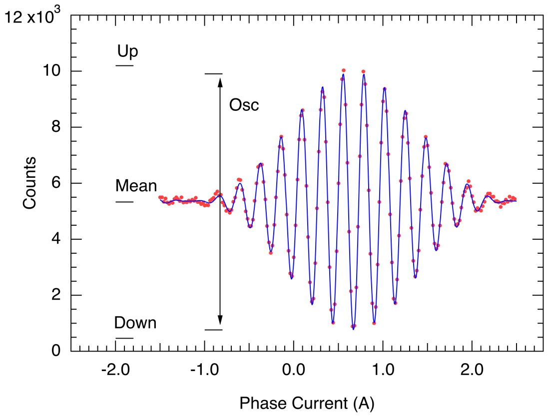


Fig. 2. (a) The time-of-flight diagram for pure NSE type of operation. (b) The principle of time-of-flight filtering for NSE-TOF type of operation.

Mezei, 1979

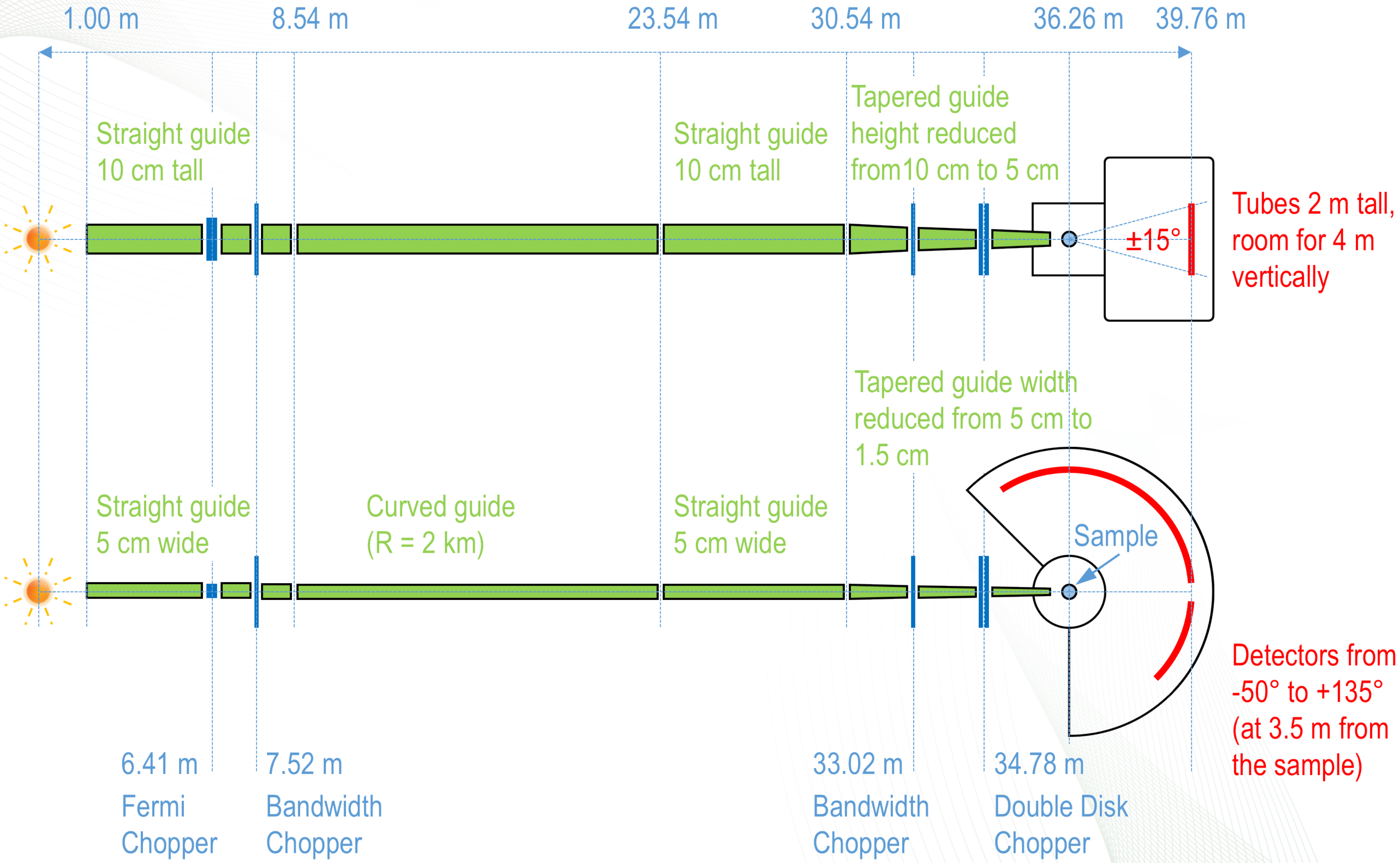
Inelastic NSE on top of TOF – is it possible?



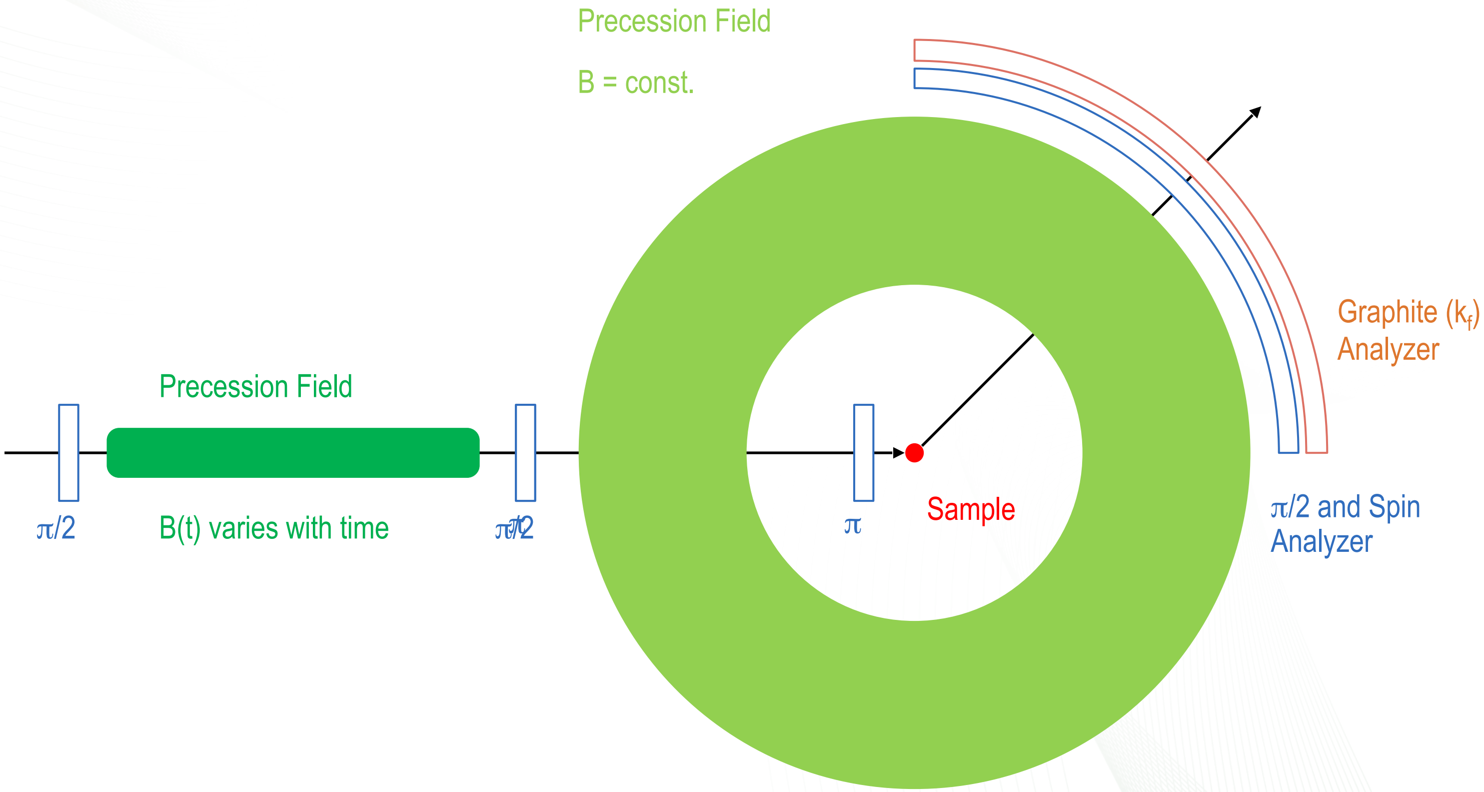


C. W. Li et al., Nat. Phys. **11** 1063 (2015)

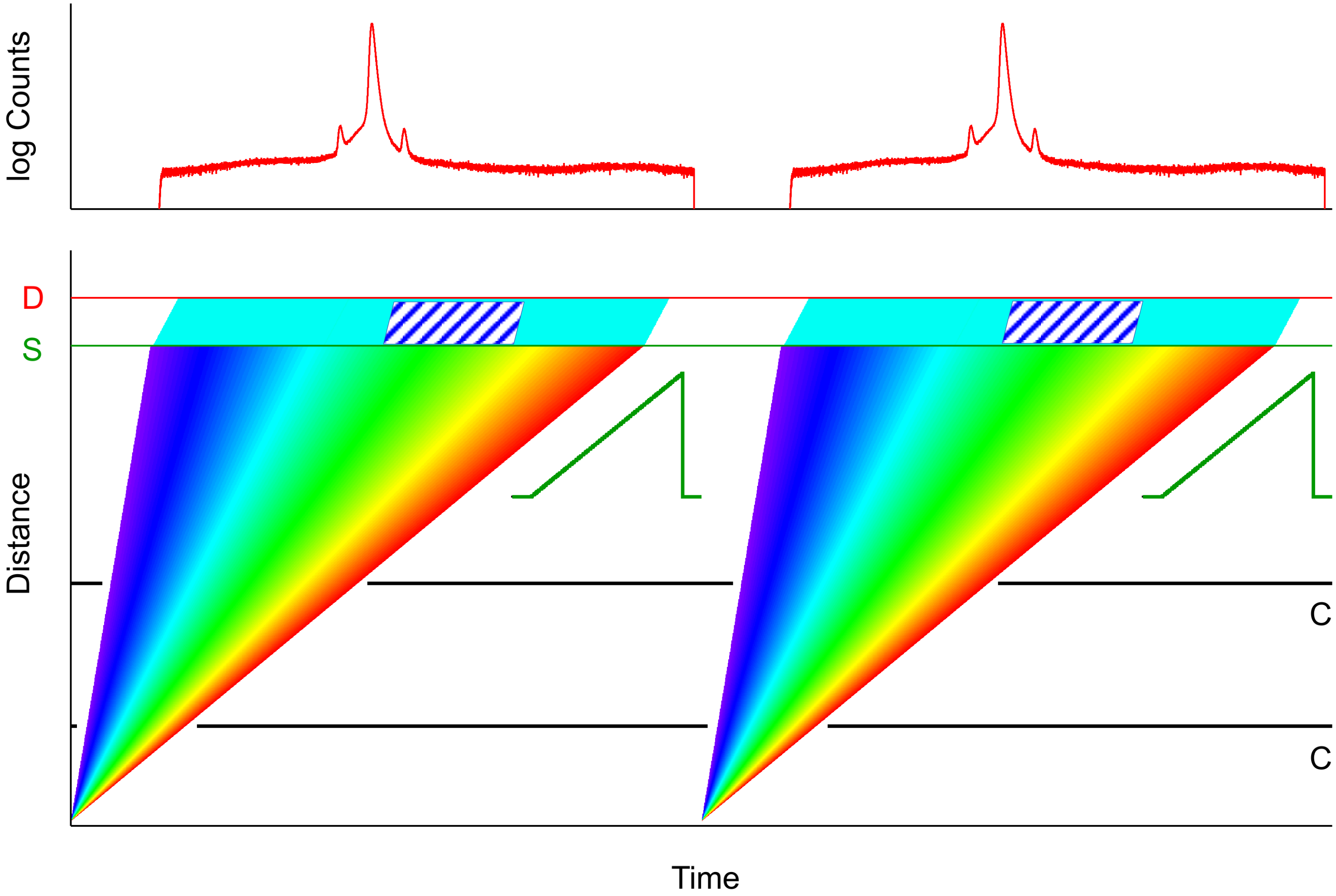
Inelastic NSE on top of TOF – is it possible?



Inelastic NSE on top of TOF “host” – is it possible?



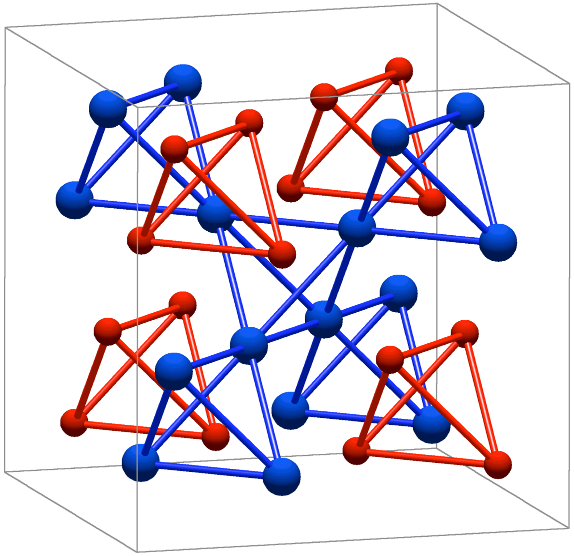
Inelastic TOF NSE – on top of Backscattering Host?



Complementarity with other techniques in the energy domain

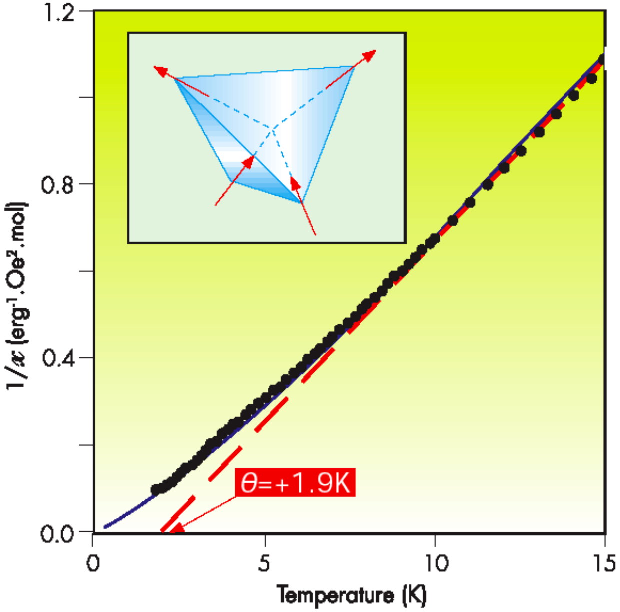
Spin Ice

Spin Ice Crystal Structure



Stoichiometry

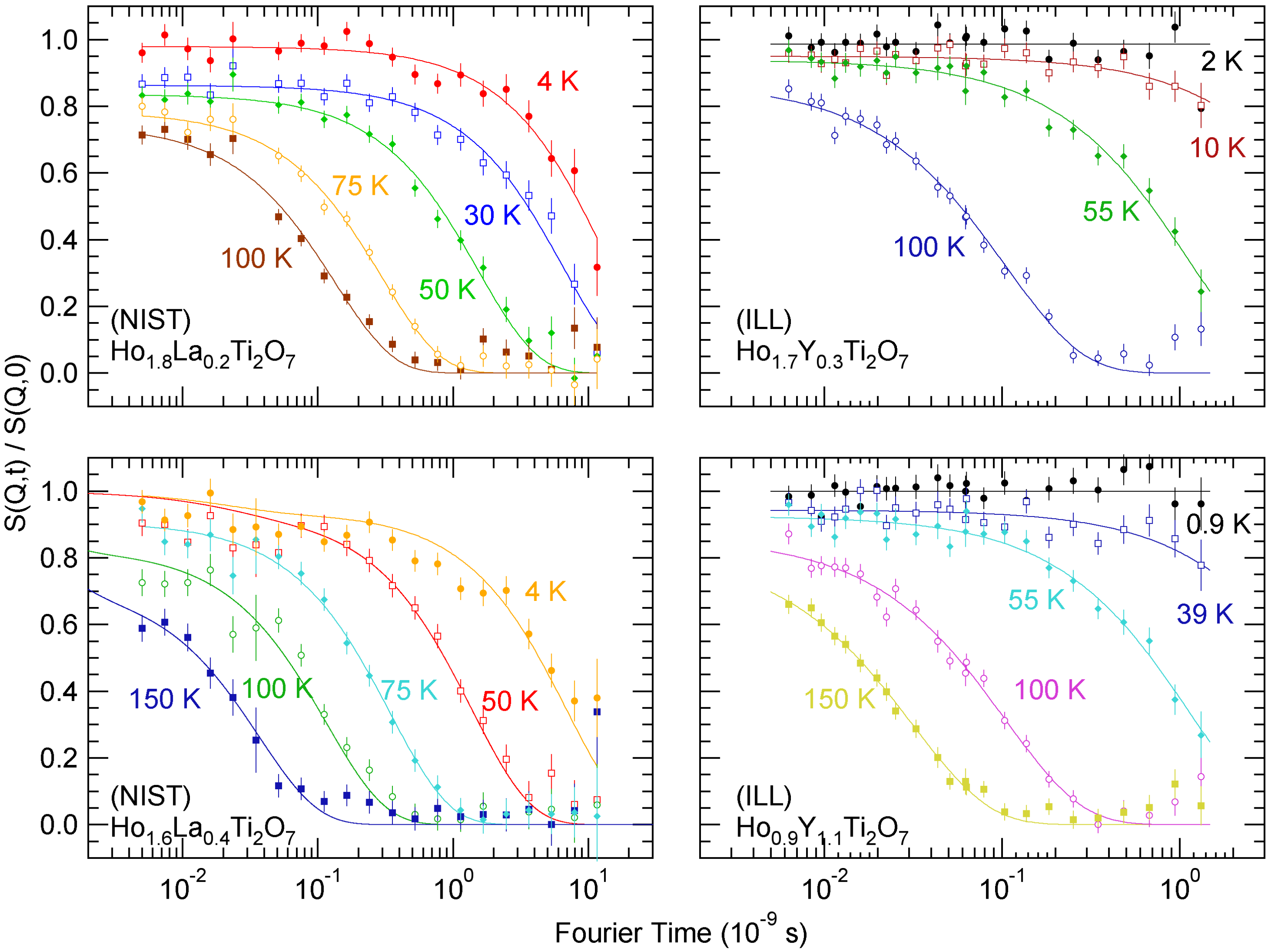
$Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$, ($Ho_2Sn_2O_7$, $Dy_2Sn_2O_7$)



$$\chi(T) = \frac{n}{3k_B} \cdot \frac{\mu_{\text{eff}}^2}{T - \Theta}$$

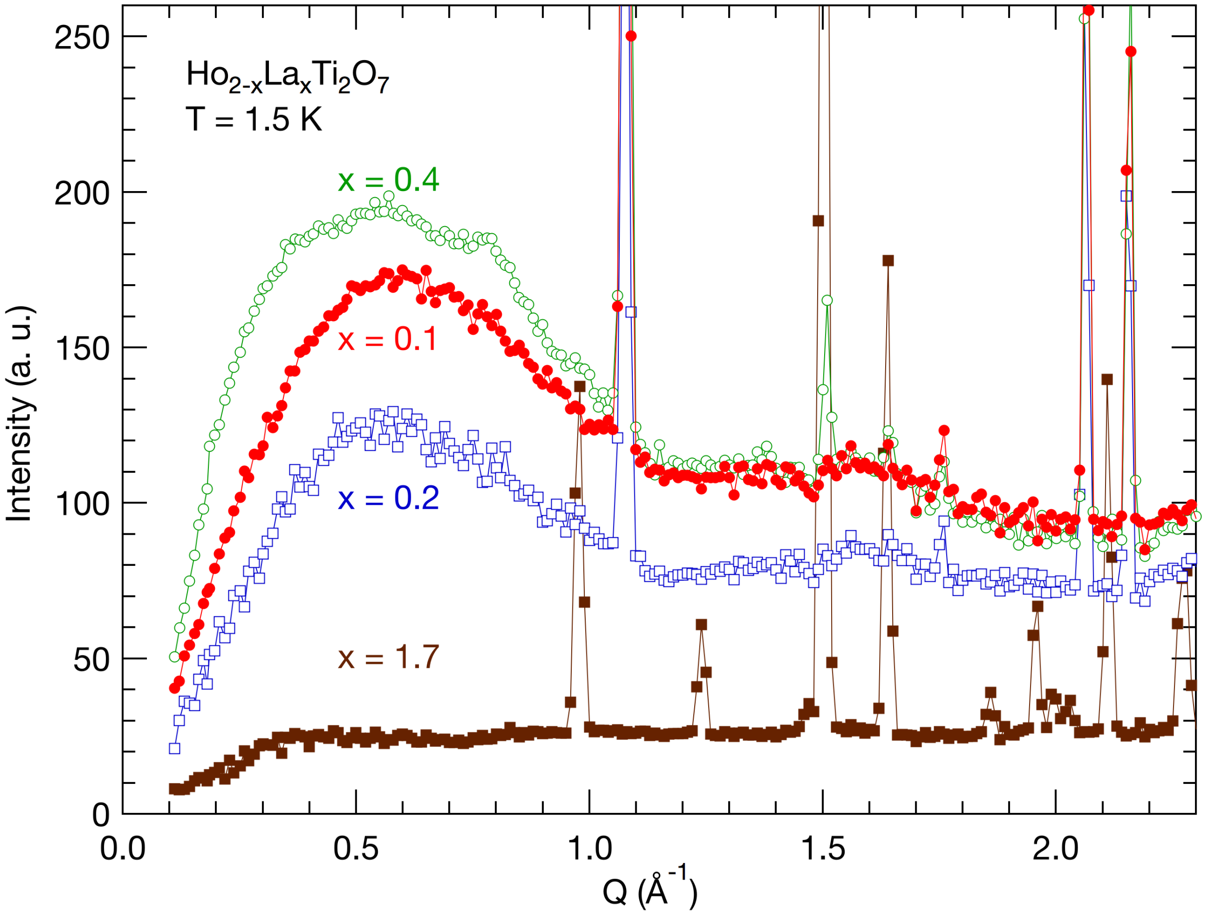
$$\mu_{\text{eff}} \approx 9\mu_B \quad \Theta \approx +1\text{K}$$

Defect-assisted relaxation in a spin ice



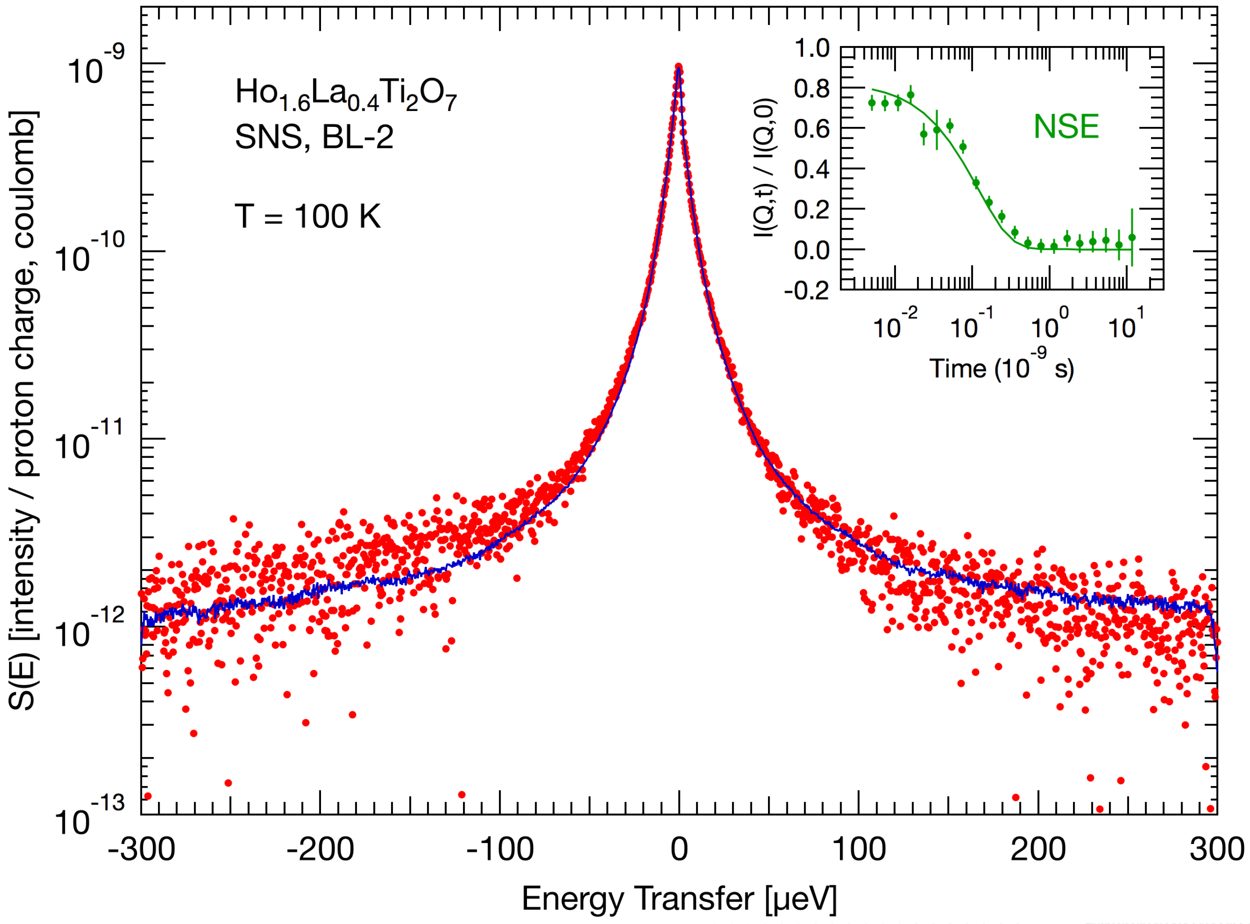
G. Ehlers et al., PRB **73** 174429 (2006)

Complementarity with techniques in the energy domain



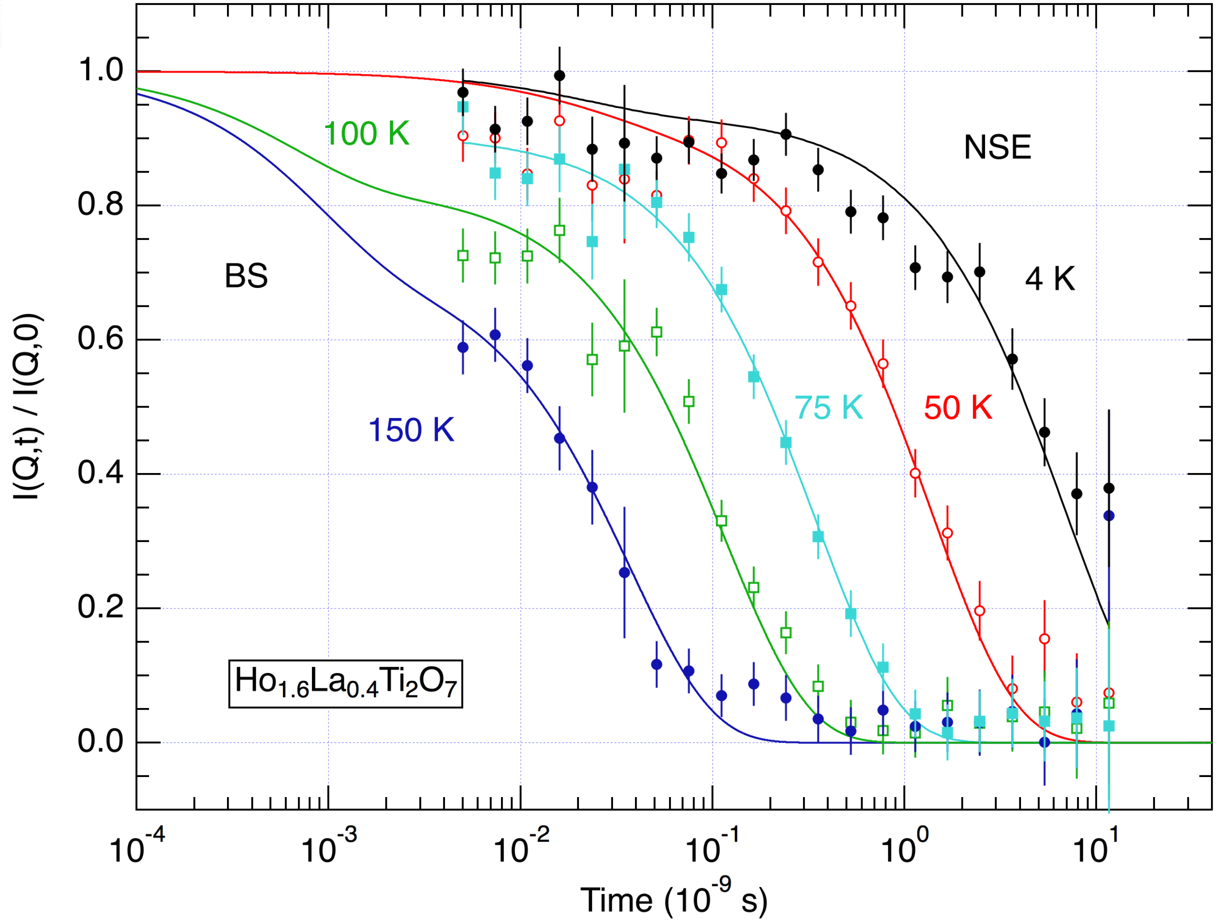
G. Ehlers et al., J. Phys. Condensed Matter **20** 235206 (2008)

Complementarity with techniques in the energy domain



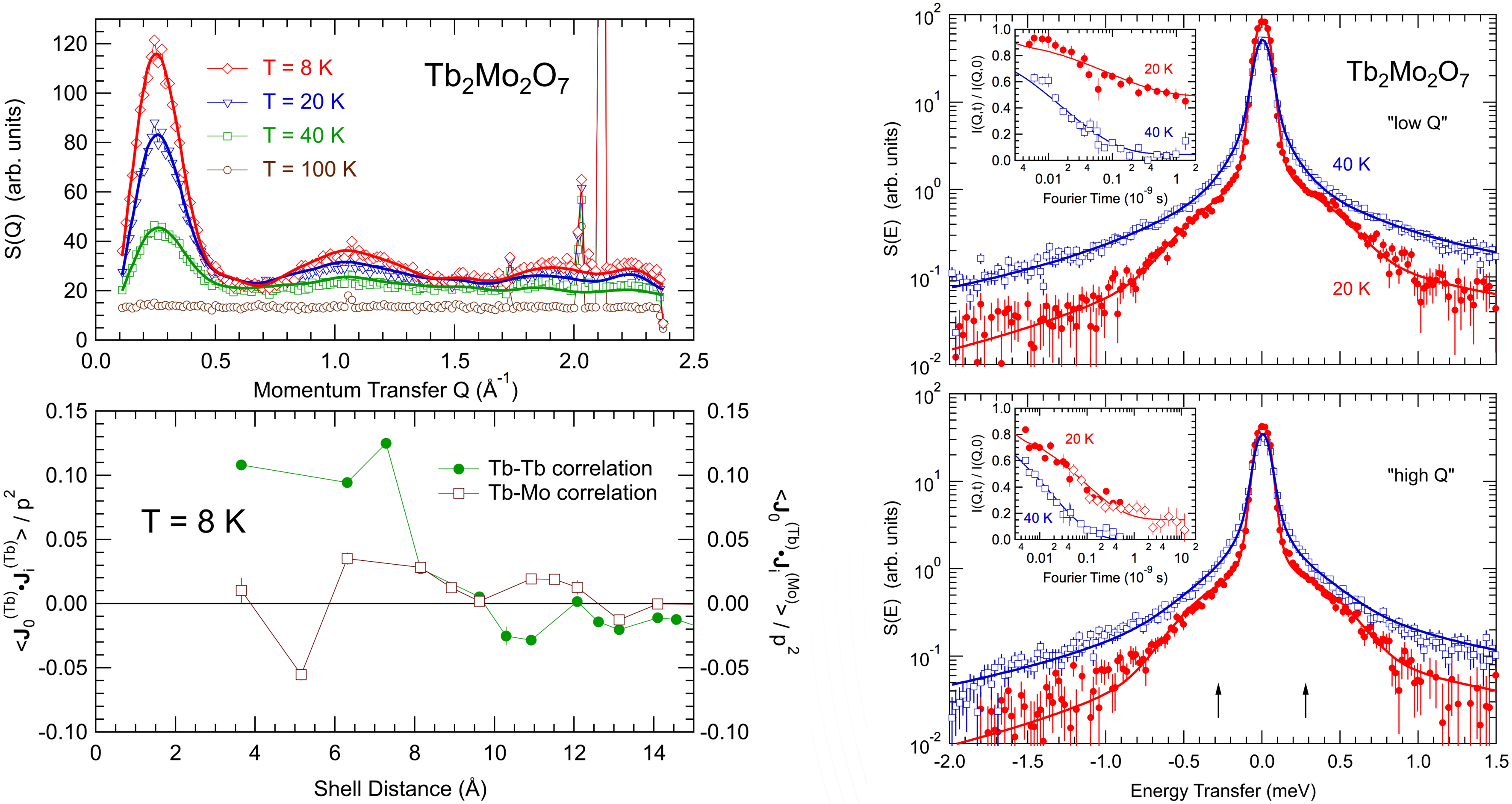
G. Ehlers et al., J. Phys. Condensed Matter **20** 235206 (2008)

Complementarity with techniques in the energy domain



G. Ehlers et al., J. Phys. Condensed Matter **20** 235206 (2008)

Complementarity with techniques in the energy domain



G. Ehlers et al., PRB **81** 224405 (2010)

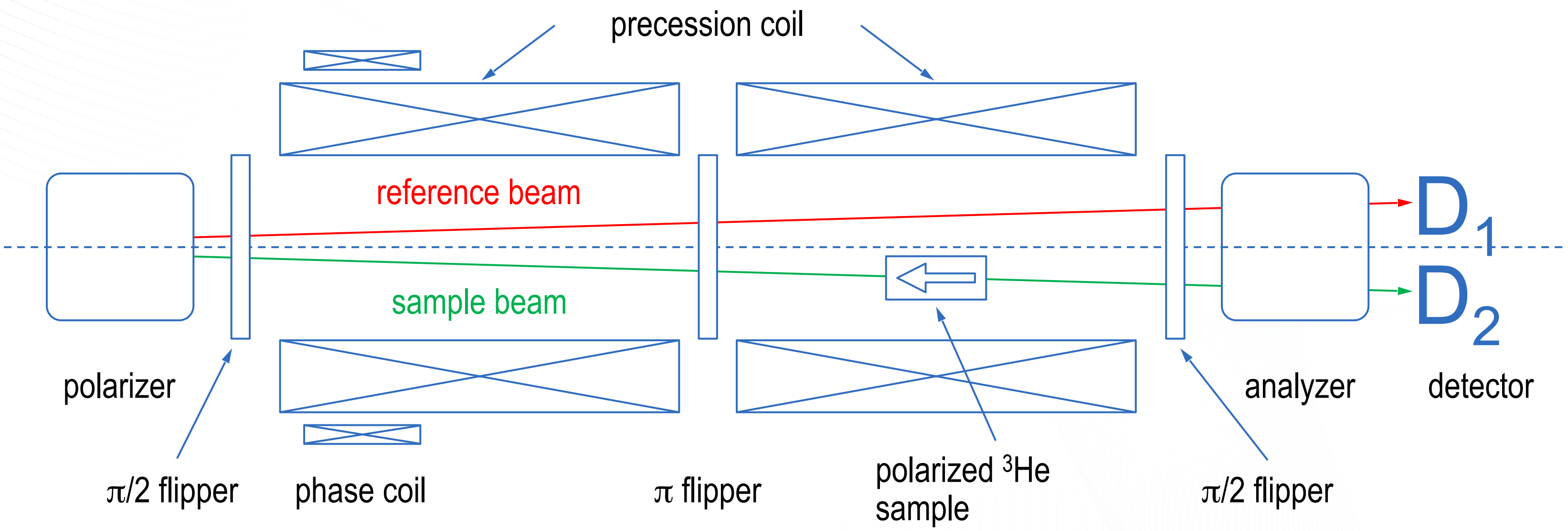
“Odd” things to measure

Fermi pseudo-Potential in Schrödinger Equation

$$V_F = \frac{2\pi\hbar^2}{m} N b_c' + \frac{4\pi\hbar^2}{m} N b_i' \sqrt{\frac{I}{I+1}} \mathbf{s} \cdot \mathbf{P} - \boldsymbol{\mu} \cdot \mathbf{B}$$

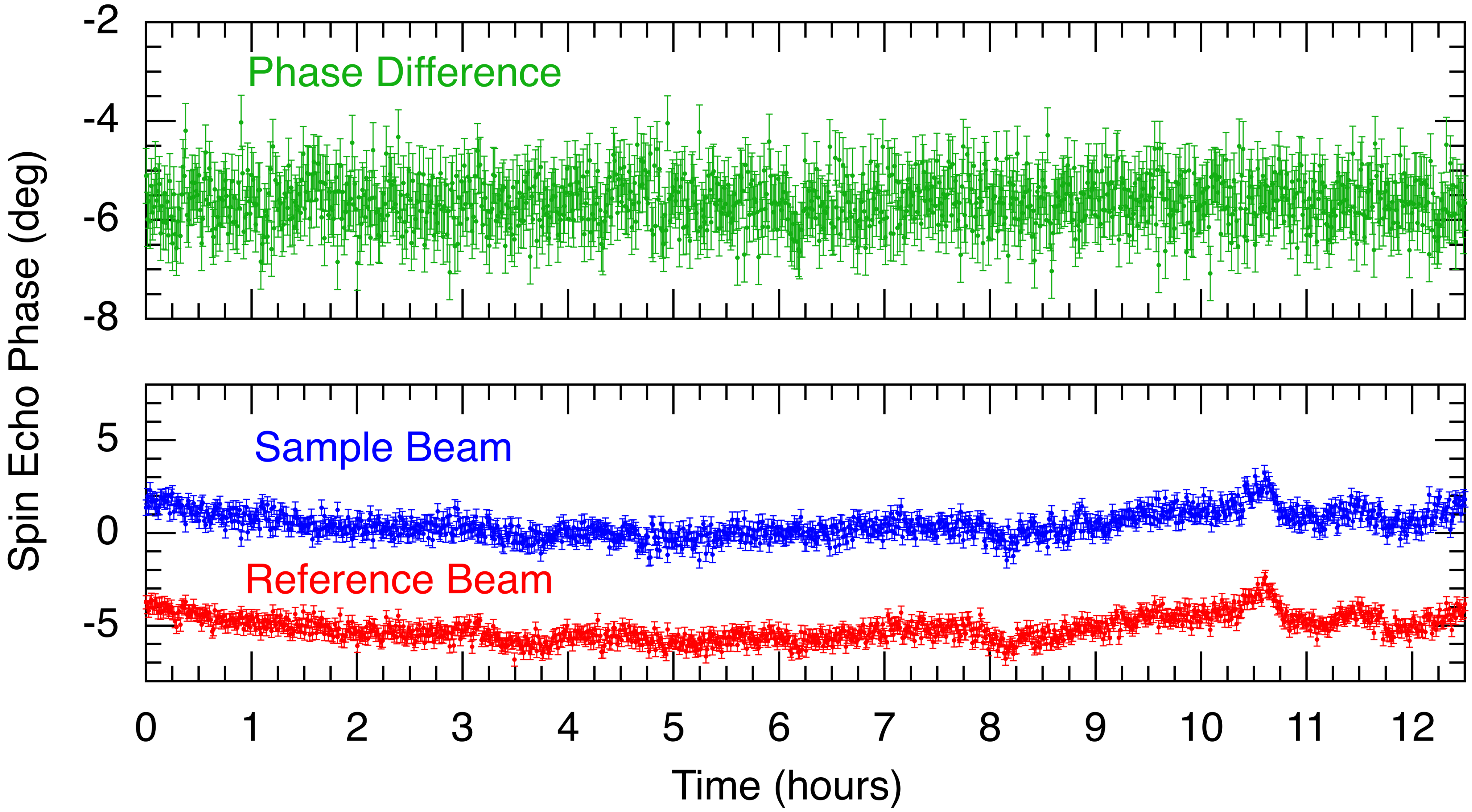
(1)	neutron refraction	$\varphi \propto \lambda^3$
(2)	pseudomagnetic field	$\varphi \propto \lambda$
(3)	magnetic field	$\varphi \propto \lambda$

Two-Beam Method @IN15



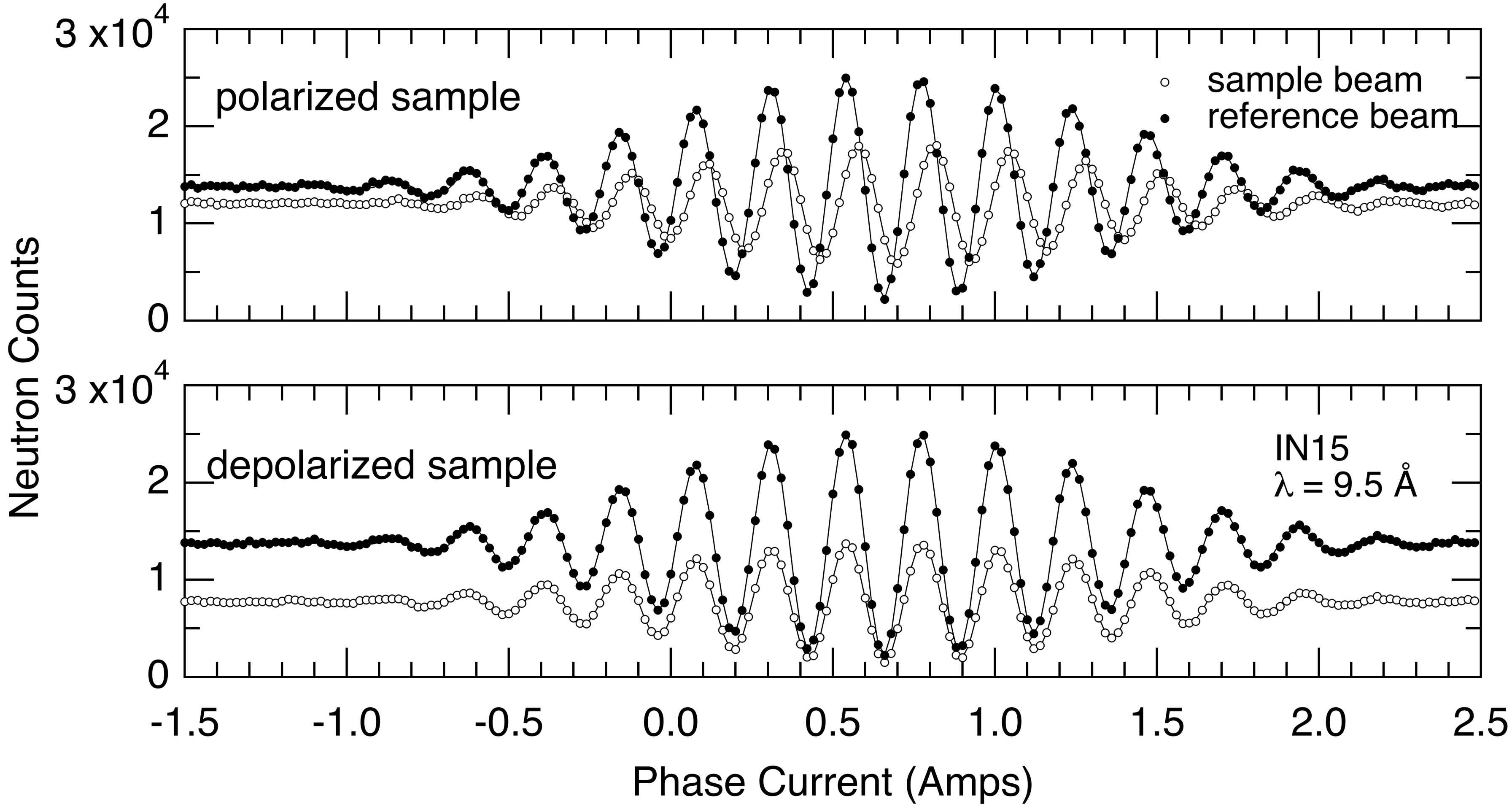
O. Zimmer et al., EPJdirect **A1** 1 (2002)

Two-Beam Method @IN15



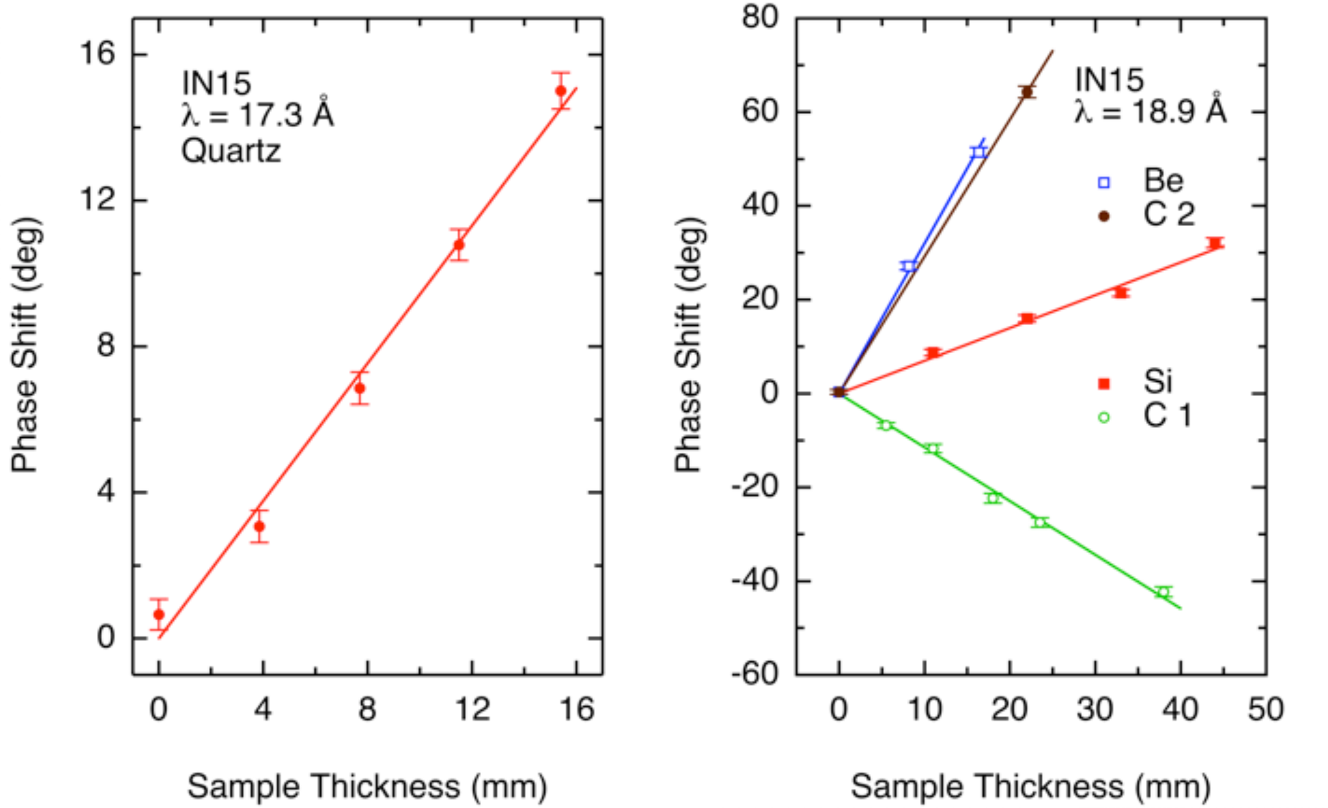
O. Zimmer et al., EPJdirect **A1** 1 (2002)

Two-Beam Method @IN15



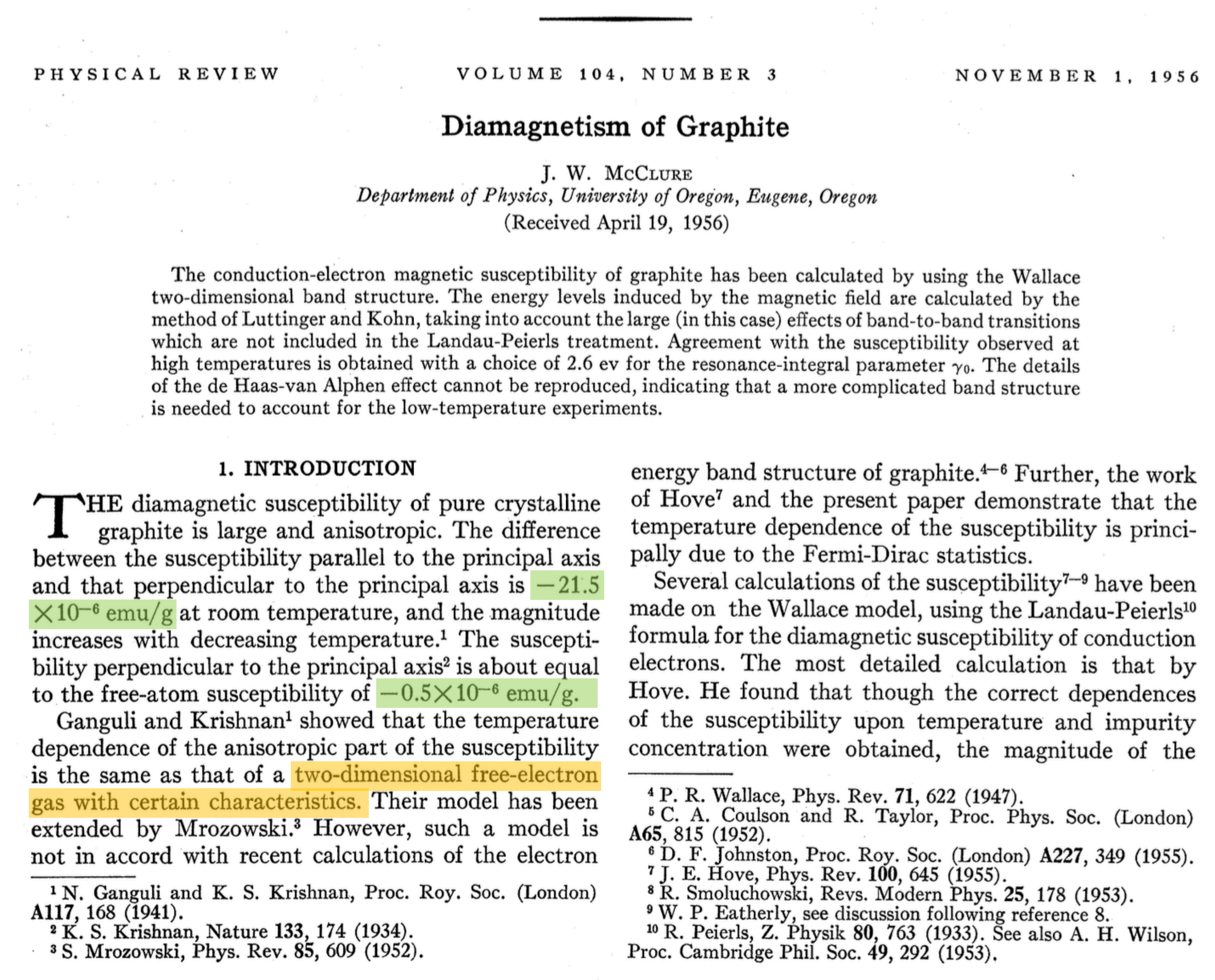
O. Zimmer et al., EPJdirect **A1** 1 (2002)

Effect from Neutron Refraction



A. I. Frank et al., Physics of Atomic Nuclei **65** 2066 (2002)

Anisotropic diamagnetic susceptibility of graphite



PHYSICAL REVIEW VOLUME 104, NUMBER 3 NOVEMBER 1, 1956

Diamagnetism of Graphite

J. W. McClure
Department of Physics, University of Oregon, Eugene, Oregon

(Received April 19, 1956)

The conduction-electron magnetic susceptibility of graphite has been calculated by using the Wallace two-dimensional band structure. The energy levels induced by the magnetic field are calculated by the method of Luttinger and Kohn, taking into account the large (in this case) effects of band-to-band transitions which are not included in the Landau-Peierls treatment. Agreement with the susceptibility observed at high temperatures is obtained with a choice of 2.6 ev for the resonance-integral parameter γ_0. The details of the de Haas-van Alphen effect cannot be reproduced, indicating that a more complicated band structure is needed to account for the low-temperature experiments.

1. INTRODUCTION

THE diamagnetic susceptibility of pure crystalline graphite is large and anisotropic. The difference between the susceptibility parallel to the principal axis and that perpendicular to the principal axis is -21.5×10^{-6} emu/g at room temperature, and the magnitude increases with decreasing temperature.[1] The susceptibility perpendicular to the principal axis[2] is about equal to the free-atom susceptibility of -0.5×10^{-6} emu/g.

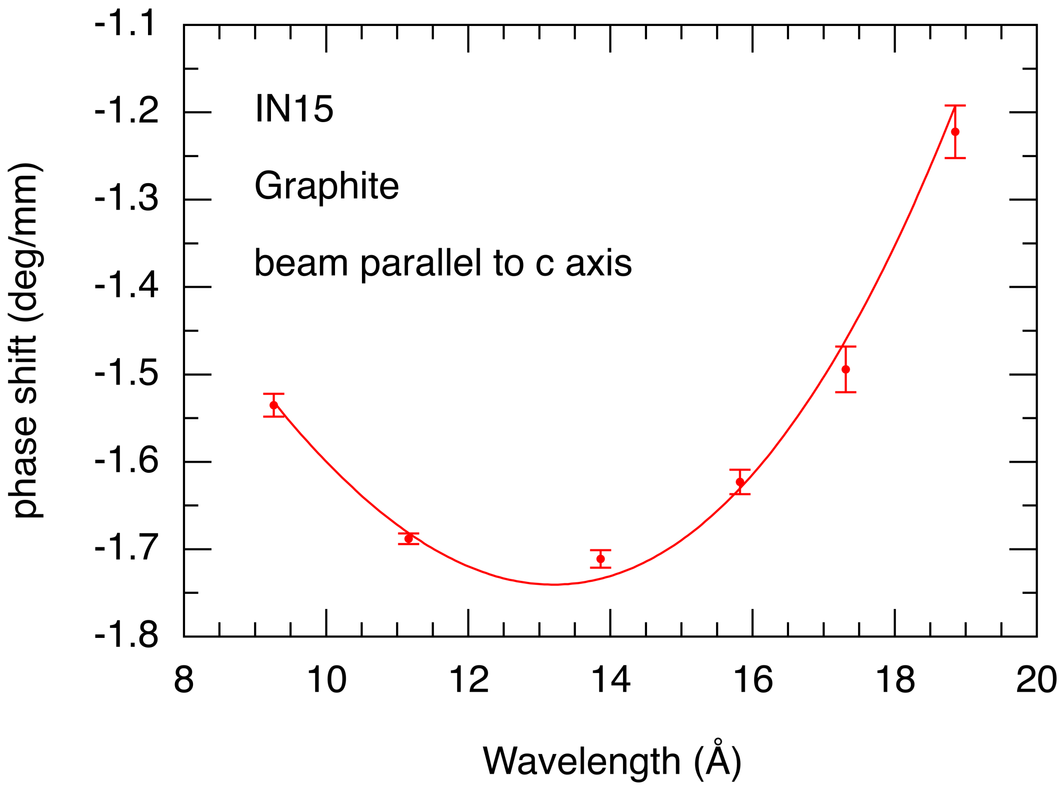
Ganguli and Krishnan[1] showed that the temperature dependence of the anisotropic part of the susceptibility is the same as that of a two-dimensional free-electron gas with certain characteristics. Their model has been extended by Mrozowski.[3] However, such a model is not in accord with recent calculations of the electron energy band structure of graphite.[4–6] Further, the work of Hove[7] and the present paper demonstrate that the temperature dependence of the susceptibility is principally due to the Fermi-Dirac statistics.

Several calculations of the susceptibility[7–9] have been made on the Wallace model, using the Landau-Peierls[10] formula for the diamagnetic susceptibility of conduction electrons. The most detailed calculation is that by Hove. He found that though the correct dependences of the susceptibility upon temperature and impurity concentration were obtained, the magnitude of the

---

[1] N. Ganguli and K. S. Krishnan, Proc. Roy. Soc. (London) A117, 168 (1941).
[2] K. S. Krishnan, Nature 133, 174 (1934).
[3] S. Mrozowski, Phys. Rev. 85, 609 (1952).
[4] P. R. Wallace, Phys. Rev. 71, 622 (1947).
[5] C. A. Coulson and R. Taylor, Proc. Phys. Soc. (London) A65, 815 (1952).
[6] D. F. Johnston, Proc. Roy. Soc. (London) A227, 349 (1955).
[7] J. E. Hove, Phys. Rev. 100, 645 (1955).
[8] R. Smoluchowski, Revs. Modern Phys. 25, 178 (1953).
[9] W. P. Eatherly, see discussion following reference 8.
[10] R. Peierls, Z. Physik 80, 763 (1933). See also A. H. Wilson, Proc. Cambridge Phil. Soc. 49, 292 (1953).

Effect from Neutron Refraction



fit to

$\varphi = A\lambda + B\lambda^3$

gives

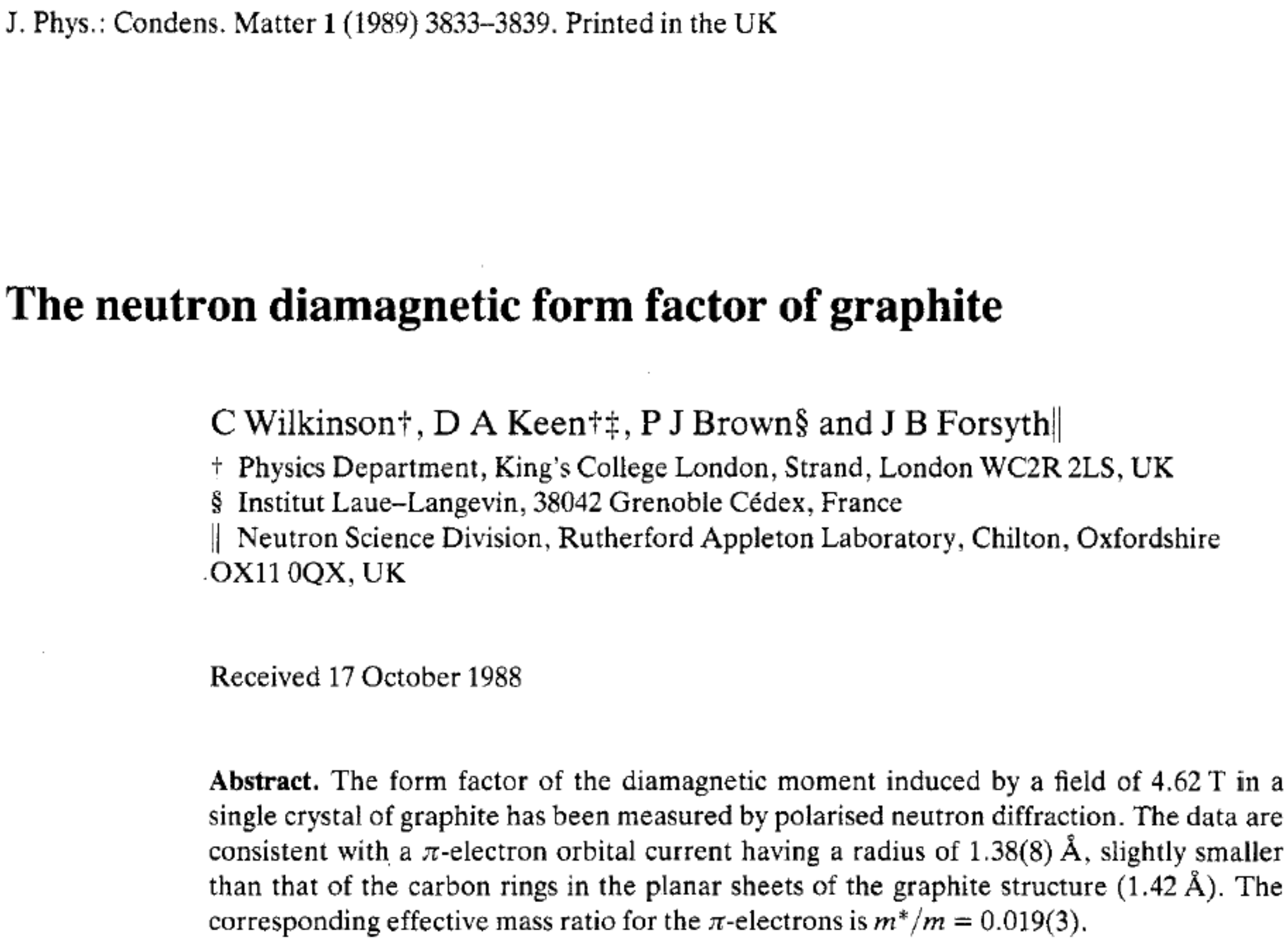
$A = (198 \pm 1) \times 10^{-3}$
$B = (396 \pm 7) \times 10^{-6}$

A. I. Frank et al., Physics of Atomic Nuclei **65** 2066 (2002)

Translation to Susceptibility

translate A to diamagnetic susceptibility	$\chi_c = B_{\mathrm{int}} / B_{\mathrm{ext}}$ $\chi_c = (-0.574 \pm 0.003) \times 10^{-3}$
measured	$\chi_c = (-0.563 \pm 0.005) \times 10^{-3}$
translate B to scattering length density	$\rho_b = (7.2 \pm 0.1) \times 10^{-6}$ Å^{-2}
table value	$\rho_b = 7.56 \times 10^{-6}$ Å^{-2}

Alternative way to measure, D3 (polarized diffraction)



J. Phys.: Condens. Matter **1** (1989) 3833–3839. Printed in the UK

The neutron diamagnetic form factor of graphite

C Wilkinson†, D A Keen†‡, P J Brown§ and J B Forsyth‖
† Physics Department, King's College London, Strand, London WC2R 2LS, UK
§ Institut Laue–Langevin, 38042 Grenoble Cédex, France
‖ Neutron Science Division, Rutherford Appleton Laboratory, Chilton, Oxfordshire OX11 0QX, UK

Received 17 October 1988

Abstract. The form factor of the diamagnetic moment induced by a field of 4.62 T in a single crystal of graphite has been measured by polarised neutron diffraction. The data are consistent with a π-electron orbital current having a radius of 1.38(8) Å, slightly smaller than that of the carbon rings in the planar sheets of the graphite structure (1.42 Å). The corresponding effective mass ratio for the π-electrons is $m^*/m = 0.019(3)$.

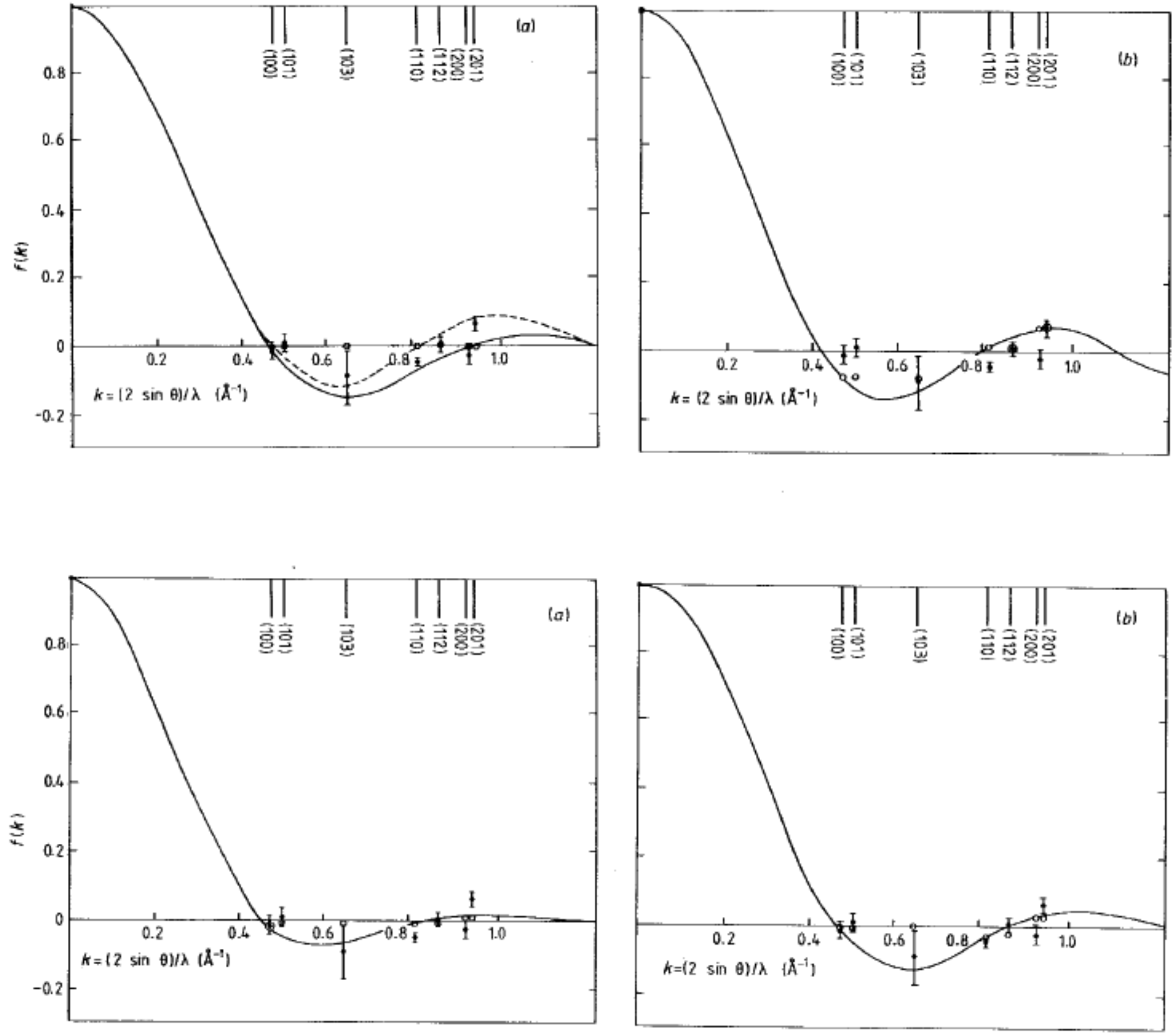
Alternative way to measure, D3 (polarized diffraction)

induced magnetic moment:

0.0056 μ_B per ring

measured at D3 (ILL)

$\lambda = 0.9$ Å
$B = 4.6$ T



OAK RIDGE National Laboratory | SPALLATION NEUTRON SOURCE

Acknowledgments

Measurements were performed in collaboration with

J. S. Gardner (ANSTO)
A. Faraone, Y Qui (NIST)
B. Farago, P. Fouquet, O. Zimmer (ILL)
J. R. Stewart (ISIS)
E. Mamontov (SNS)
A. I. Frank (Dubna)
A. Cornelius (U Nevada)
K. C. Kam (UC Santa Barbara)

DONE!

- ✓ About myself
- ✓ NSE at reactor and pulsed sources
- ✓ Complementarity with other techniques in the energy domain
- ✓ “Odd” things to measure

Questions ?