Neutron scattering as a tool to understand quantum magnetism: Magnetism and the European Spallation Source

Pascale Deen.
ESS: Senior Scientist
Adjunct associate professor: Niels Bohr Inst., Copenhagen University

Overview

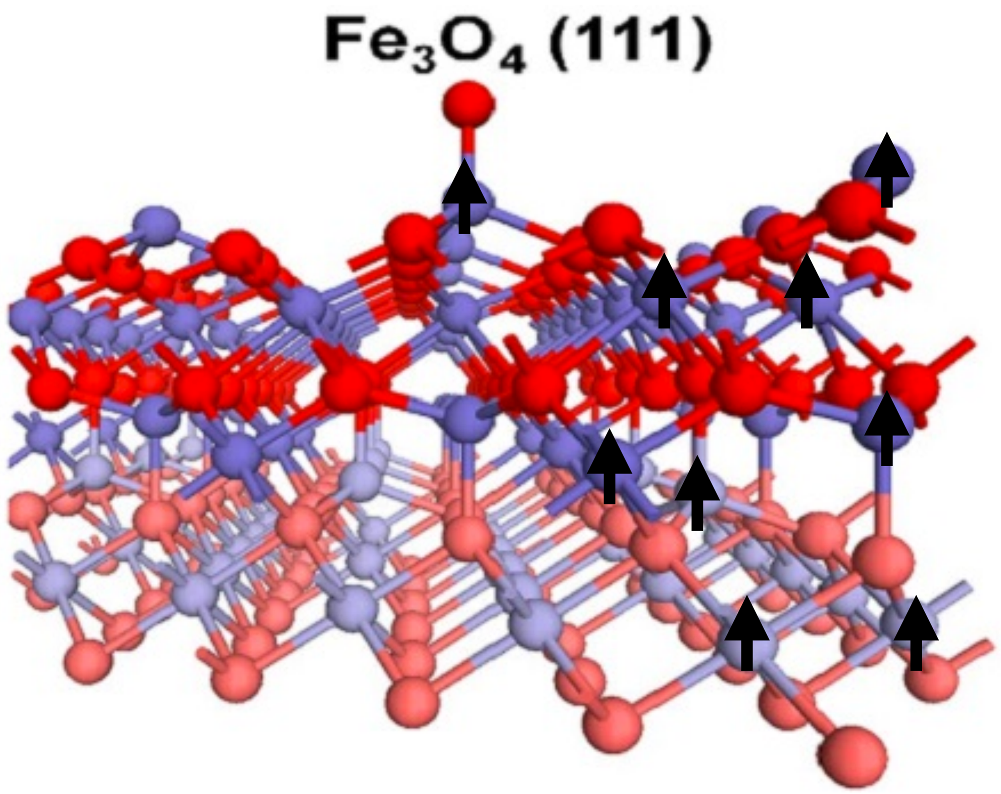
- Magnetism : a very basic overview
- Neutron scattering and magnetism
  - Diffraction
  - Inelastic neutron scattering
  - Polarisation analysis
- Recent examples of magnetic states of matter (Quantum behaviour)
- Overview of some instruments at ESS relevant to magnetism

Overview

- **Magnetism : a very basic overview**
- Neutron scattering and magnetism
  - Diffraction
  - Inelastic neutron scattering
  - Polarisation analysis
- Recent examples of magnetic states of matter (Quantum behaviour)
- Overview of some instruments at ESS relevant to magnetism

Magnetism: Ferromagnetism

6th Century BC
Lodestone
Magnet: lodestones found in Magnesia, Turkey
Natural magnets
Used early for navigation (Fe3O4 + Fe2O3)

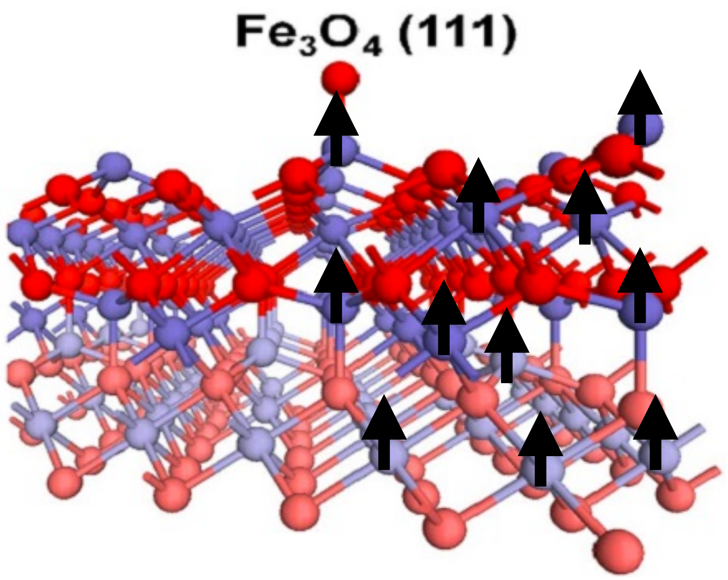


Ferromagnet

electromagnets, electric motors, generators, transformers, magnetic storage, credit cards .....

Magnetism: Ferromagnetism

Signatures



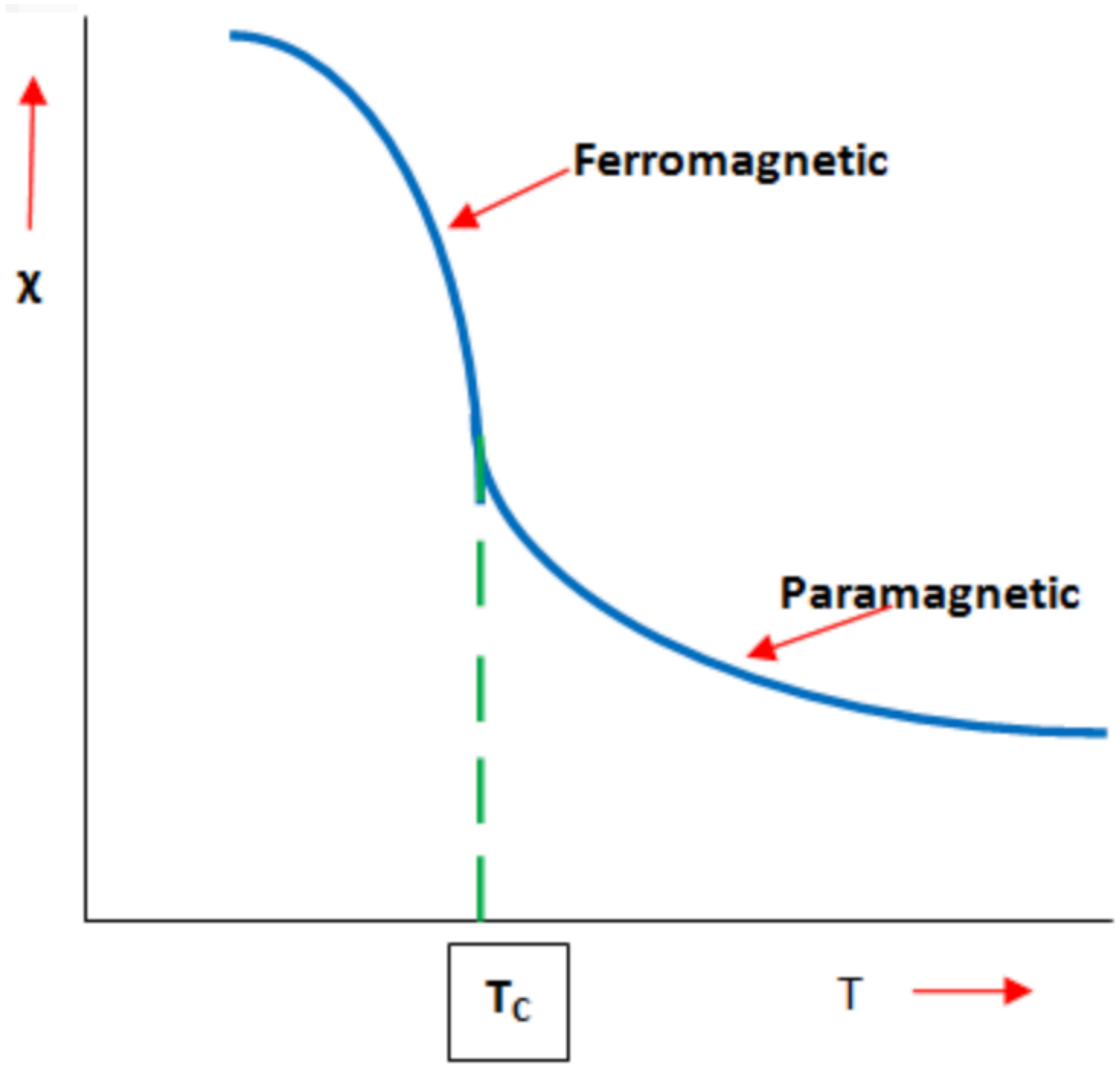


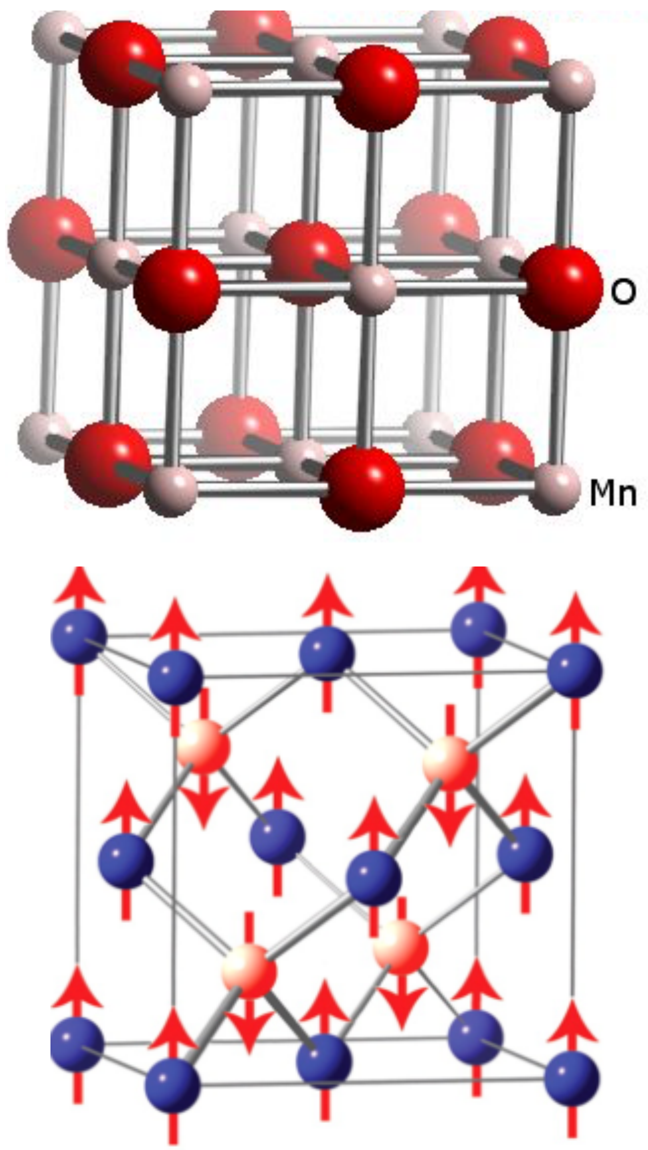
Figure 1

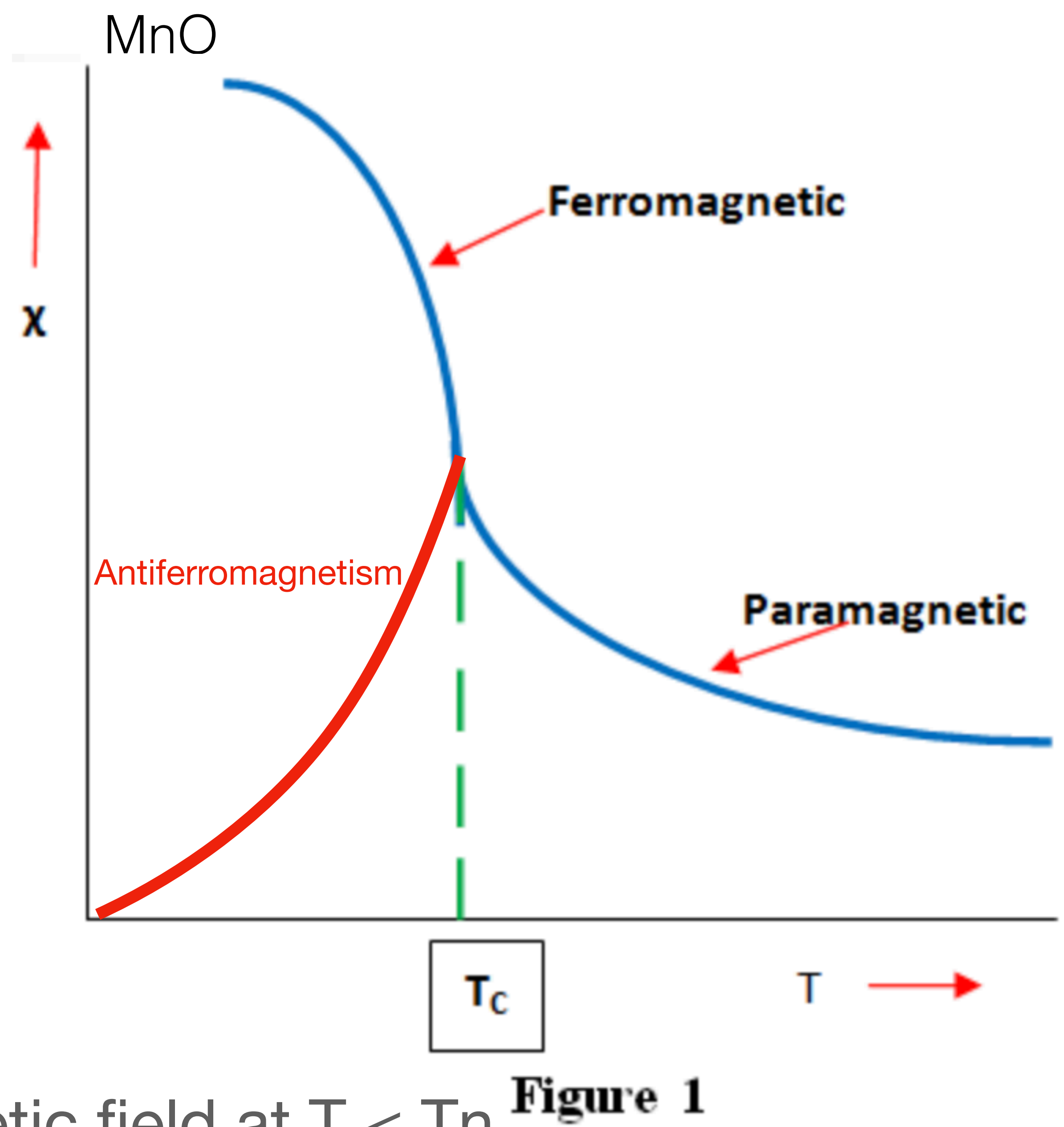
Ferromagnet

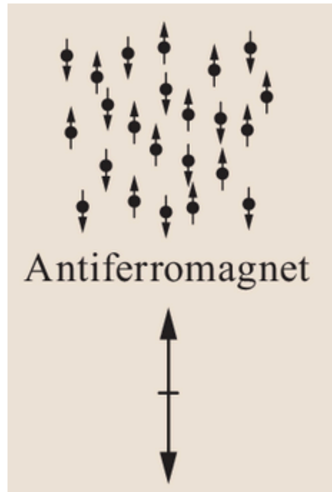
electromagnets, electric motors, generators, transformers, magnetic storage, credit cards …..

Magnetism: Antiferromagnetism, a hidden order

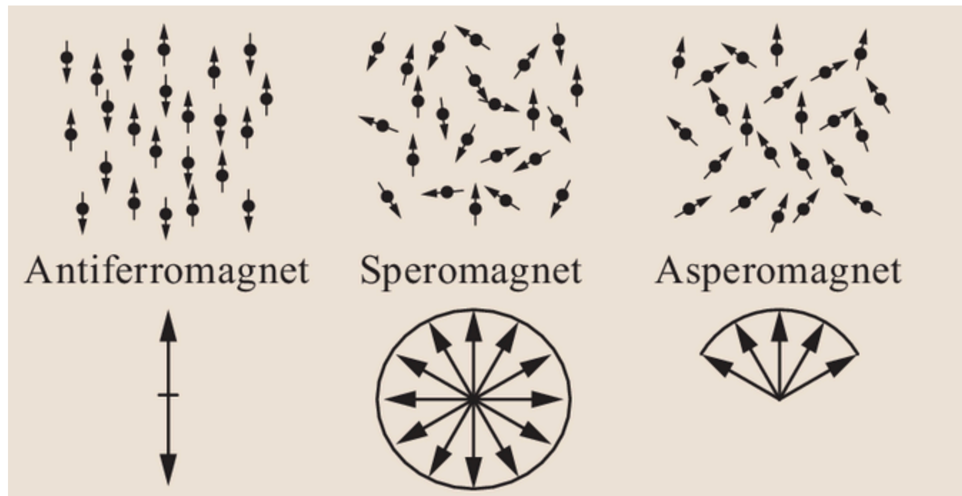
Signatures







Generators, detectors and transmitters of spin currents, spin valves, hard drives.



No variation with a magnetic field at T < Tn

Antiferromagnetism, theoretical debate.

Louis Néel

Lev Landau

- mean field theory
- AF is described as magnetic order on two sublattices.
- Macroscopic picture.

- quantum paramagnet, fluctuating spins in opposition
- no time averaged moment.
- Associated with quantum order

Overview

- Magnetism : a very basic overview
- **Neutron scattering and magnetism**
  - Diffraction
  - Inelastic neutron scattering
  - Polarisation analysis
- Recent examples of magnetic states of matter (Quantum behaviour)
- Overview of some instruments at ESS relevant to magnetism

Neutrons for magnetic and electronic phenomena

1932

Chadwick:Discovery of the neutron

What is the neutron spin?
What is the neutron magnetic moment
How do neutrons scatter from magnetic atoms?

1936/1937

Bloch: Classical dipole-dipole interaction

Spin = 1/2
Magnetic moment = 1.91 μN (~0.001μB)

Phys. Rev. 50. 259. 1936

σ nuclear XS
γ_N neutron moment
γe atomic moment, μ_B
C - Shape of surface

$$\phi_\omega = \sigma_\omega \left| 1 \pm \frac{\gamma_n \gamma_e}{2(\sigma_\omega)^{\frac{1}{2}}} \frac{e^2}{mc^2} \left(\frac{q_z^2}{q^2} - C \right) \right|^2$$

1936/1937

Schwinger: Atomic moments ||or ⊥ to Q

Phys. Rev. 52. 1250. 1937

Calculate scattering probabilities

MAY 15, 1939 PHYSICAL REVIEW VOLUME 55

On the Magnetic Scattering of Neutrons

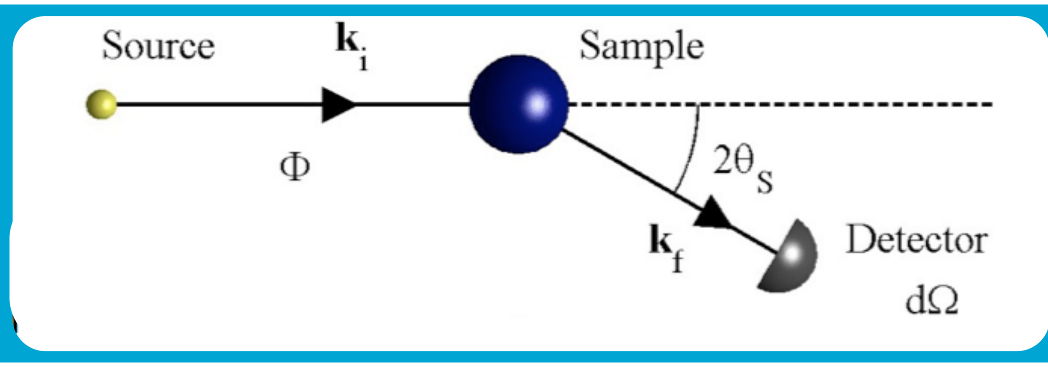
O. HALPERN AND M. H. JOHNSON
New York University, University Heights, New York, New York
(Received December 3, 1938)

In this paper there is contained a full elaboration of two previously published short notes on the subject of magnetic scattering of neutrons together with a comprehensive treatment of certain sides of this problem which have already received some attention from other authors. After presenting the state of the problem in the introduction and discussing in detail our reasons for the choice of an interaction function between neutrons and electrons, and the nonmagnetic interaction between neutrons and nuclei, the various possible cases of coherent and incoherent scattering and depolarization phenomena are treated. Later applications to the theory of ferromagnetic scattering are kept in mind. The general expression for the cross section due to magnetic interaction is obtained and applied to various classes of phenomena (scattering by free, rigidly aligned, and coupled magnetic ions). The influence of the elastic form-factor is treated quantitatively with the aid of a simple model for the current distribution in the ion. Finally a series of performed or suggested experiments is discussed mainly from the point of view whether they will permit theoretical interpretation. Arrangements are described which will allow one to obtain a reliable value for the neutron's magnetic moment and also give insight into the magnetic constitution of the scatterer (ion or crystal) which will exceed the knowledge obtainable from macroscopic magnetic experiments.

Calculate the interaction potential
(Nuclear, magnetic, incoherent contributions)

No equivalent interaction for x-rays (or any other probe)

Neutron Scattering



Intensity α $$\frac{d^2\sigma}{d\Omega dE_f} = \frac{\left(\begin{array}{c}\text{no. neutrons scattered per sec. into solid angle } d\Omega \\ \text{with final energy between } E_f \text{ and } E_f + dE_f\end{array}\right)}{I_0 \times d\Omega \times dE_f}$$

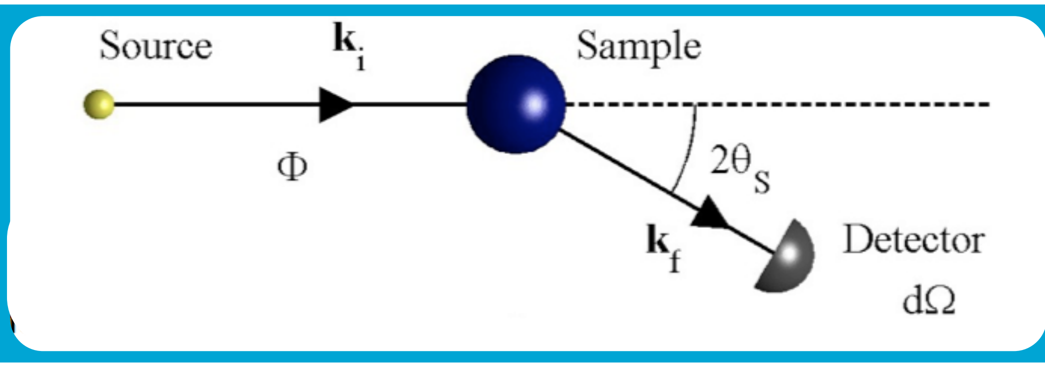
$$\frac{d^2\sigma}{d\Omega dE} = \frac{\sigma}{4\pi}\frac{k_f}{k_i} N S(\mathbf{k},\omega)$$

$$S(Q, \Delta\omega) = \left(\frac{d^2\sigma}{d\Omega dE}\right)_{\lambda_i \to \lambda_f} = \frac{k_f}{k_f}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 |\mathbf{k_f}\lambda_f|V|\mathbf{k_i}\lambda_i|\delta\left(E_{\lambda_i} - E_{\lambda_f} + \hbar\omega\right)$$

Intensity = Experiment

Theory
Separate from Probe
Absolute units

Neutron Scattering



EUROPEAN SPALLATION SOURCE

Intensity α $\frac{d^2\sigma}{d\Omega dE_f} = \frac{\left(\begin{array}{c}\text{no. neutrons scattered per sec. into solid angle } d\Omega \\ \text{with final energy between } E_f \text{ and } E_f + dE_f\end{array}\right)}{I_0 \times d\Omega \times dE_f}$

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\sigma}{4\pi}\frac{k_f}{k_i} N S(\mathbf{k},\omega)$$

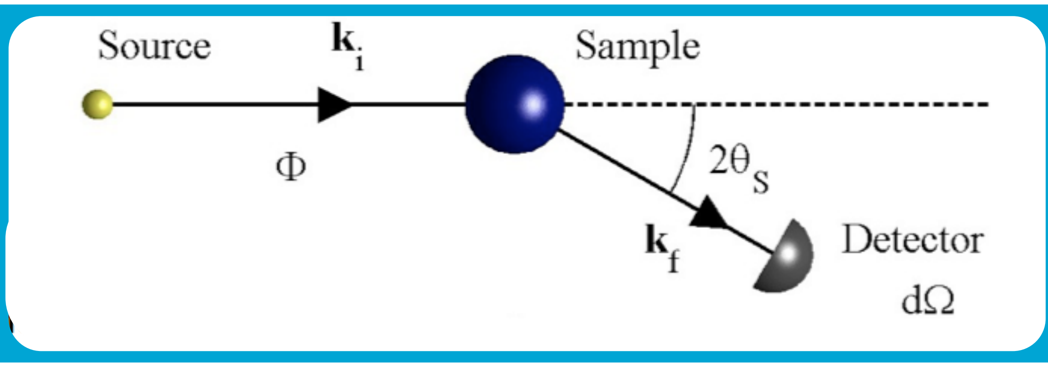
S(Q, Δω) = $\left(\frac{d^2\sigma}{d\Omega dE}\right)_{\lambda_i \to \lambda_f} = \frac{k_f}{k_f}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 |\mathbf{k_f}\lambda_f|V|\mathbf{k_i}\lambda_i|\delta\left(E_{\lambda_i} - E_{\lambda_f} + \hbar\omega\right)$

Intensity = Experiment

Theory
Separate from Probe
Absolute units

Differential neutron cross section :
Sum of all processes in which
(1) State of the scatterer changes from λ to λ’
(2) Wavevector of the neutron changes from k to k’
(3) Spin state of the neutron changes from s to s’
(4) within a solid angle Ω

Magnetic Neutron Scattering

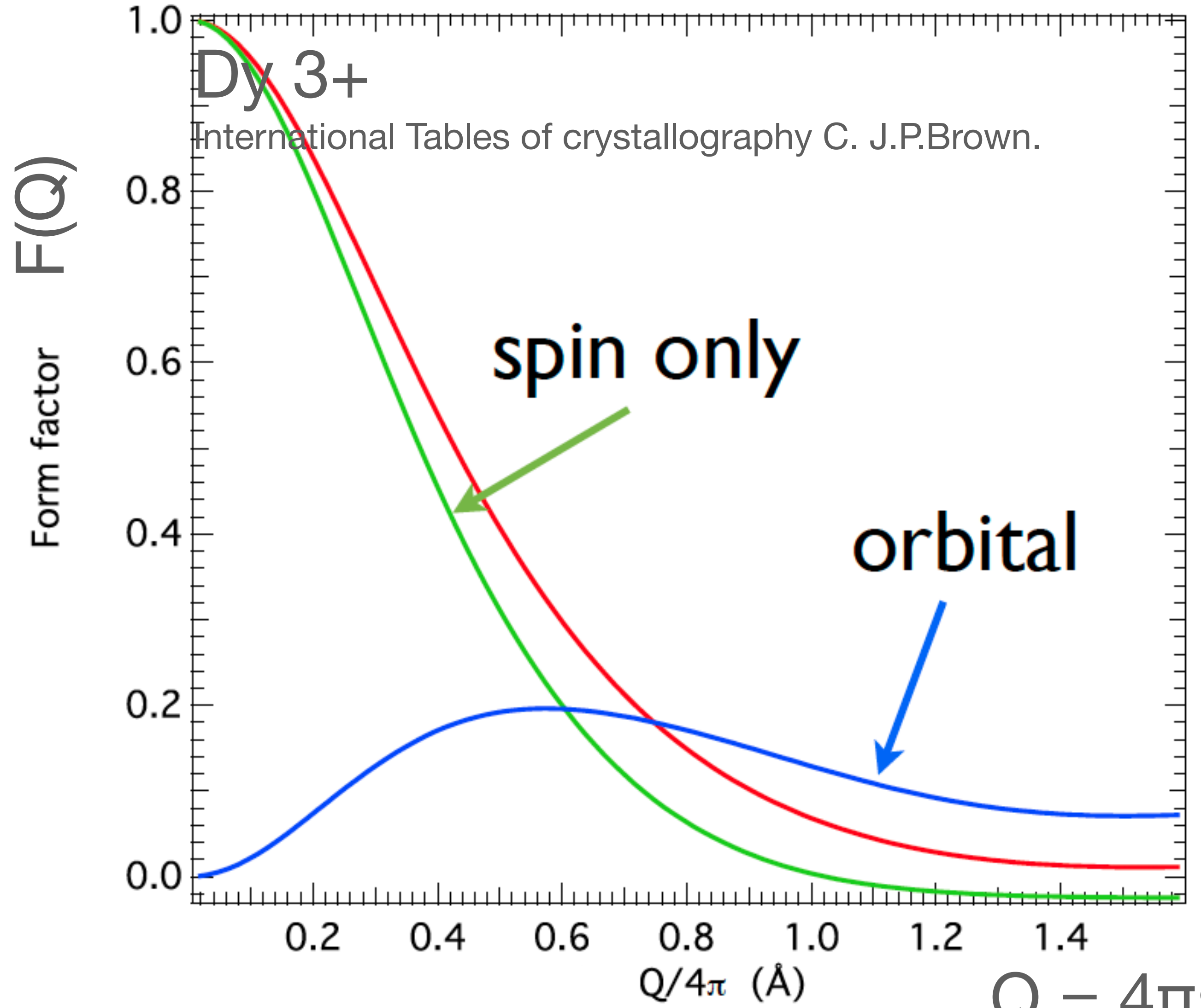


$$\left(\frac{d^2\sigma}{d\Omega dE}\right)_{\lambda_i\to\lambda_f} = \frac{k_f}{k_f}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 |\mathbf{k_f}\lambda_f|V|\mathbf{k_i}\lambda_i|\delta\left(E_{\lambda_i} - E_{\lambda_f} + \hbar\omega\right)$$

$V = \mu.B = \mu(B_S+B_L)$

$V \propto$ 1/2 g F(Q): Only scattering at low Q

$V \propto \delta\alpha\beta$ - $Q\alpha Q_{\beta}$: moments normal to Q contribute



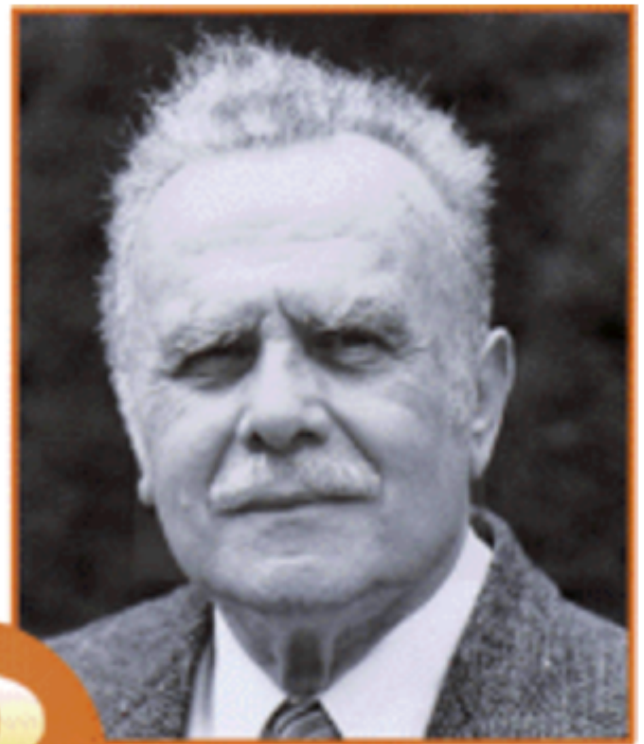
$Q = 4\pi\sin\theta/\lambda$

Neutrons Interacting with matter

EUROPEAN SPALLATION SOURCE

1994 Nobel prize: for pioneering contributions to the development of neutron scattering techniques for studies of condensed matter

Clifford G. Shull, MIT, Camebridge, Massachusetts, USA, receives one half of the 1994 Nobel Prize in Physics for development of the neutron diffraction technique.



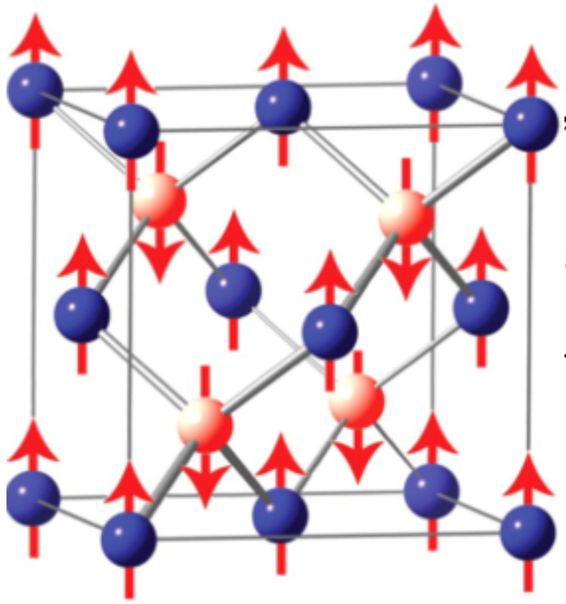
Betram N. Brockhouse, McMater University, Hamilton, Ontario, Canada, receives one half of the 1994 Nobel Prize in Physics for the development of neutron spectroscopy.

Development of neutron diffraction
Where atoms/spin are

Development of neutron spectroscopy
What atoms/spins do

Neutrons Interacting with matter

EUROPEAN SPALLATION SOURCE



Detection of Antiferromagnetism by Neutron Diffraction*

C. G. Shull
: Ridge National Laboratory, Oak Ridge, Tennessee
AND
J. Samuel Smart
nance Laboratory, White Oak, Silver Spring, Maryland
August 29, 1949

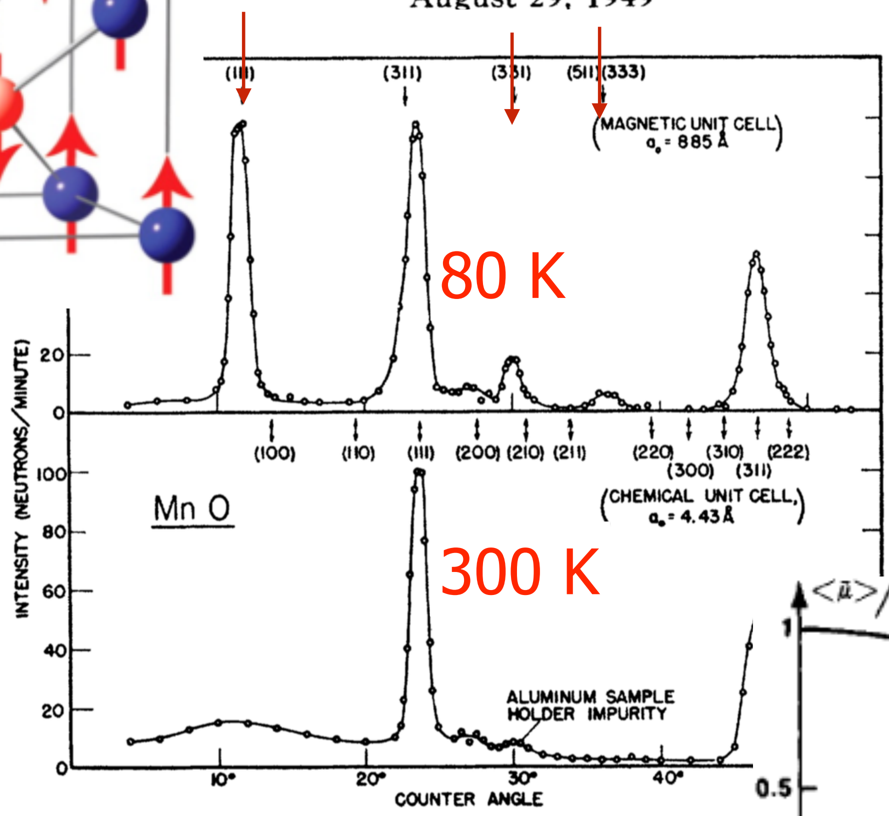
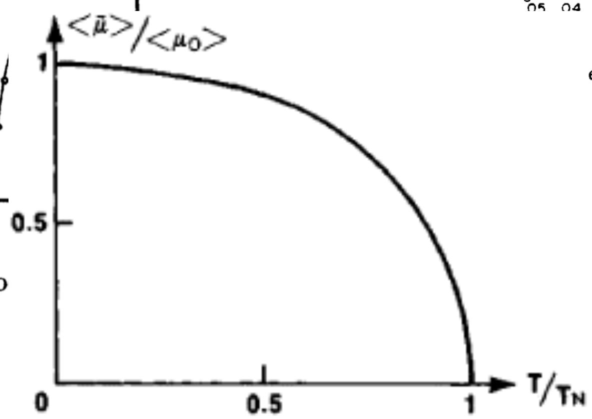


Fig. 1. Neutron diffraction patterns for MnO at ro temperature and at 80°K.



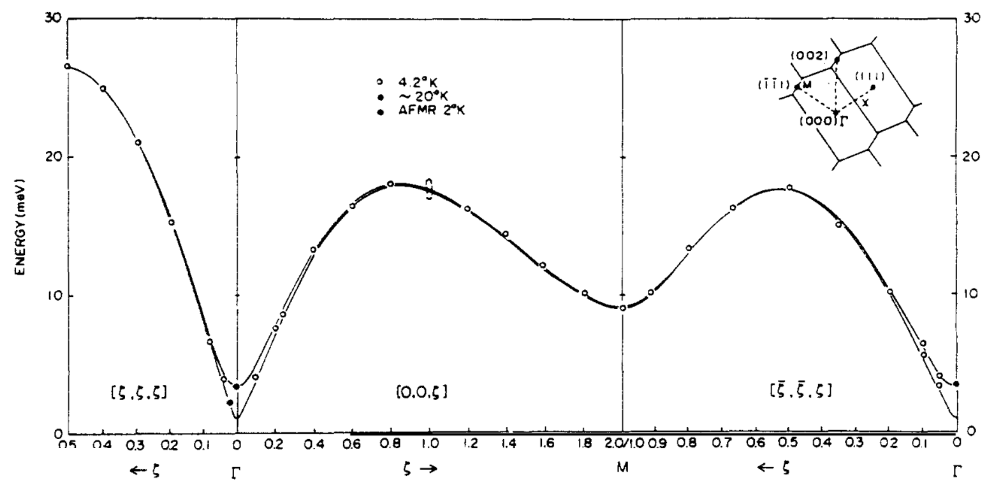
Where atoms are

Solid State Communications, Vol. 11, pp. 391–394, 1972. Pergamon Press. Printed in Great Britain

INELASTIC NEUTRON SCATTERING STUDY OF SPIN WAVES IN MnO

M. Kohgi, Y. Ishikawa and Y. Endoh
Department of Physics, Tohoku University, Sendai 980, Japan

(*Received* 6 *May* 1972 *by* *T. Nagamiya*)



ersion relation $E(q)$ of the spin waves in MnO measured along [111], [001] and [$\bar{1}\bar{1}$1].

How they move

Overview

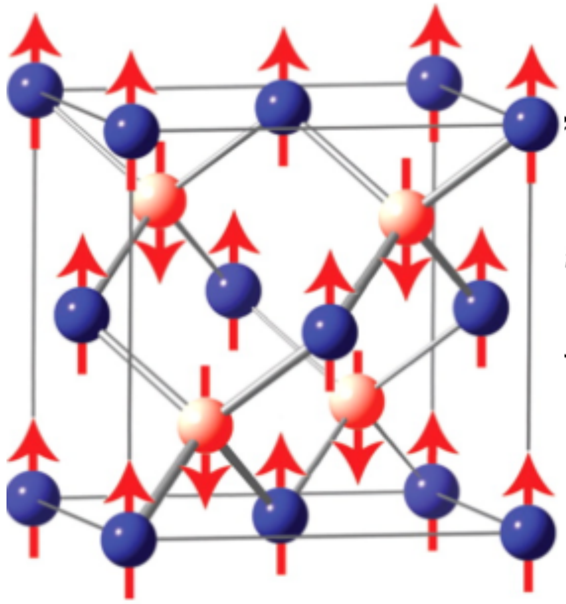
- Magnetism : a very basic overview
- **Neutron scattering and magnetism**
  - **Diffraction**
  - Inelastic neutron scattering
  - Polarisation analysis
- Recent examples of magnetic states of matter (Quantum behaviour)
- Overview of some instruments at ESS relevant to magnetism

Neutrons Interacting with matter

1994 Nobel prize: for pioneering contributions to the development of neutron scattering techniques for studies of condensed matter

Development of neutron diffraction
Where atoms/spin are

Neutrons Interacting with matter



Detection of Antiferromagnetism by Neutron Diffraction*

C. G. SHULL
: Ridge National Laboratory, Oak Ridge, Tennessee

AND

J. SAMUEL SMART
nance Laboratory, White Oak, Silver Spring, Maryland

August 29, 1949

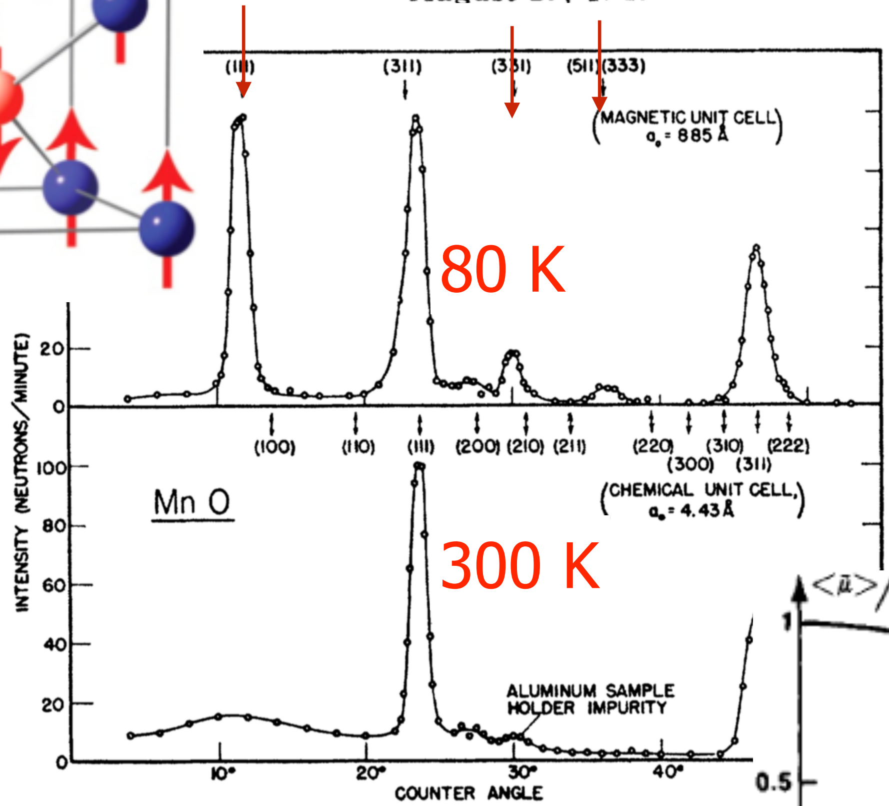
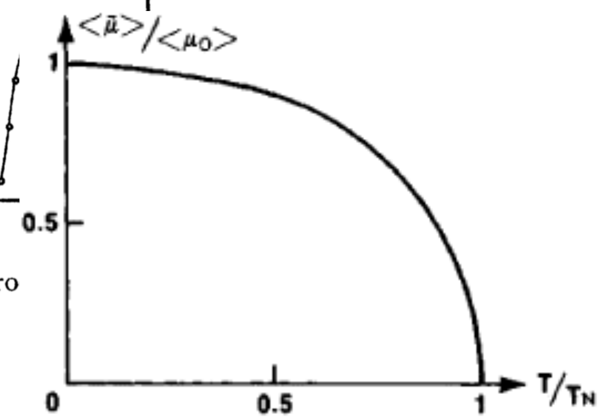


FIG. 1. Neutron diffraction patterns for MnO at ro temperature and at 80°K.

Where atoms are



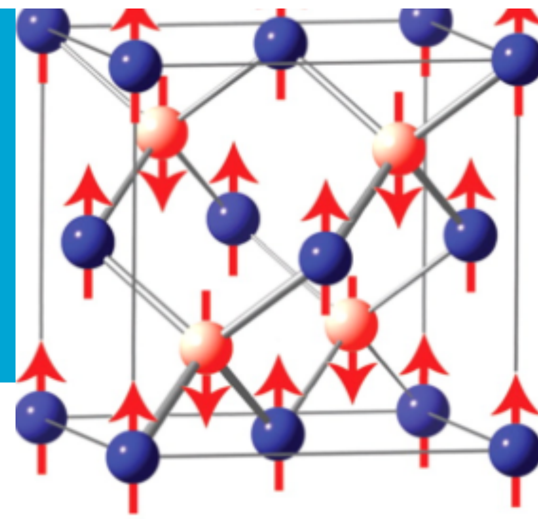
Elastic scattering/Diffraction

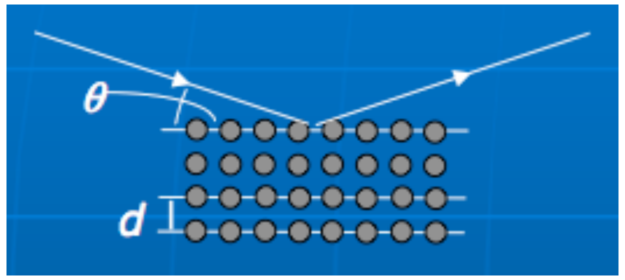
Braggs law: $n\lambda = 2\, d_{hkl} \sin(\theta)$

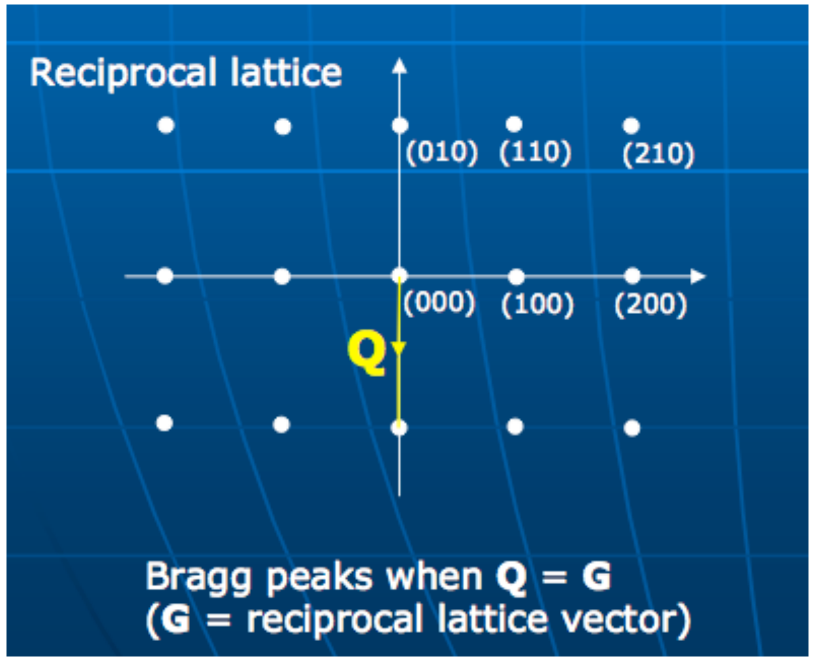
$Q = 2\pi/d_{hkl}$

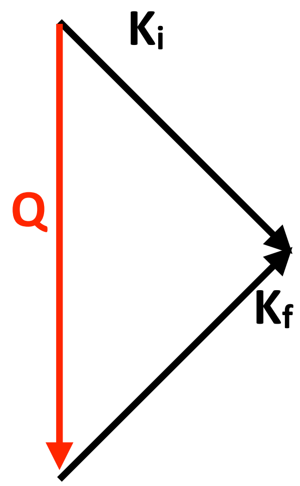
$Q = 4\pi \sqrt{[(h^2+k^2+l^2)/a^2]}$ (cubic)

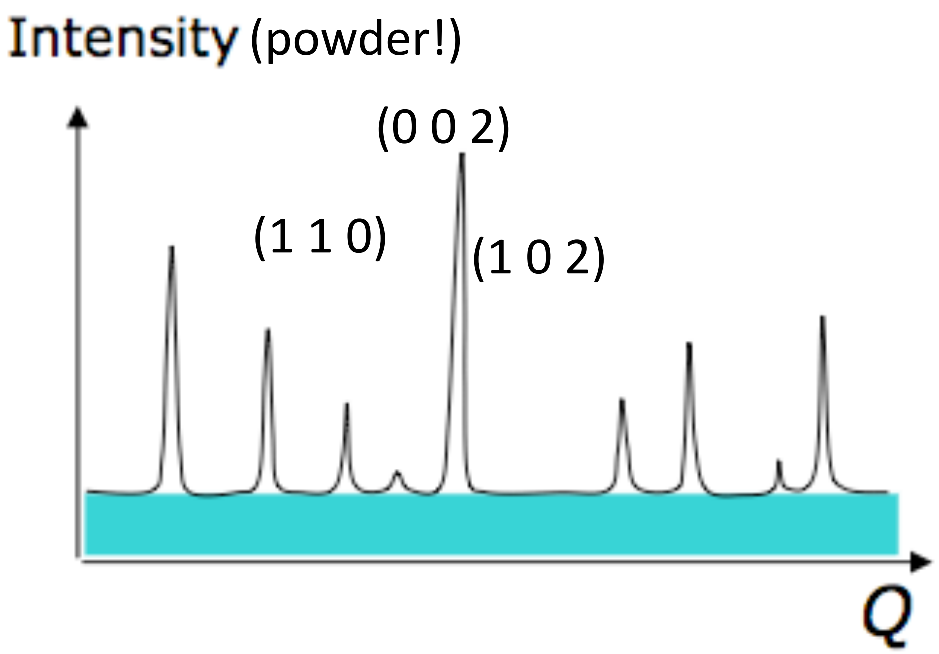
Clifford G. Shull, MIT, Cambridge, Massachusetts, USA, receives one half of the 1994 Nobel Prize in Physics for development of the neutron diffraction technique.











Static behaviour

Only interested in atomic positions (Qx, Qy, Qz)

Static correlations

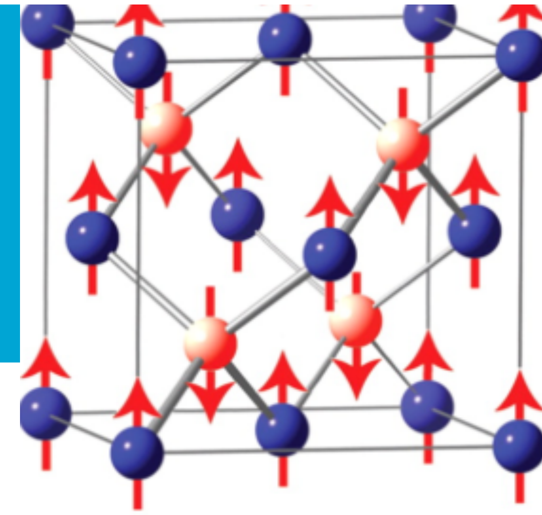
Clifford G. Shull, MIT, Cambridge, Massachusetts, USA, receives one half of the 1994 Nobel Prize in Physics for development of the neutron diffraction technique.

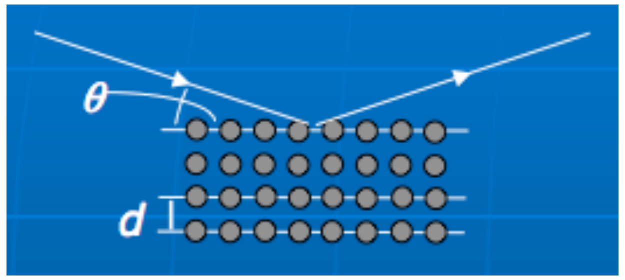
Elastic scattering/Diffraction

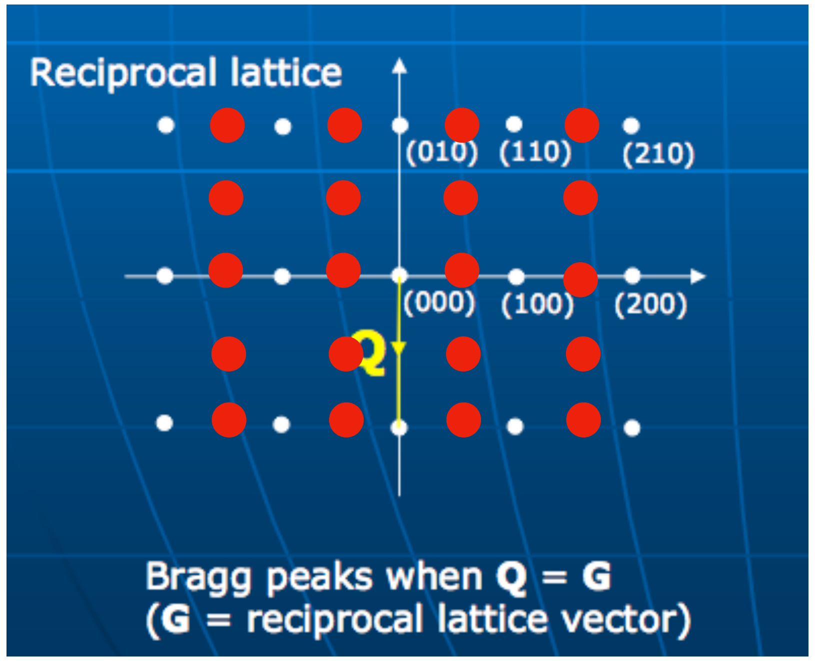
Braggs law: $n\lambda = 2\, d_{hkl} \sin(\theta)$

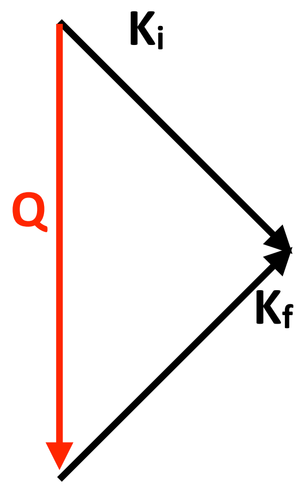
$Q = 2\pi/d_{hkl}$

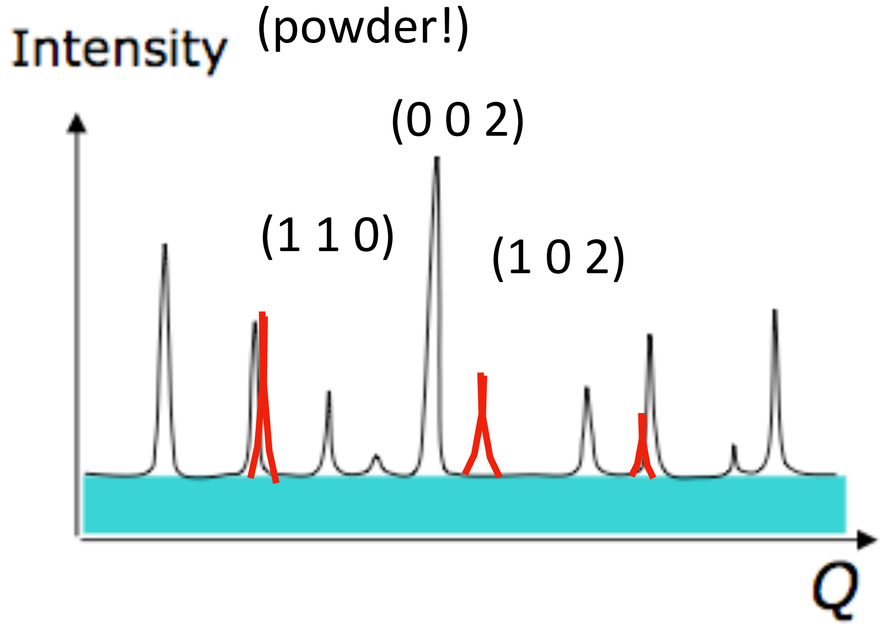
$Q = 4\pi \sqrt{[(h^2+k^2+l^2)/a^2]}$ (cubic)











Static behaviour

Only interested in atomic positions (Qx, Qy, Qz)

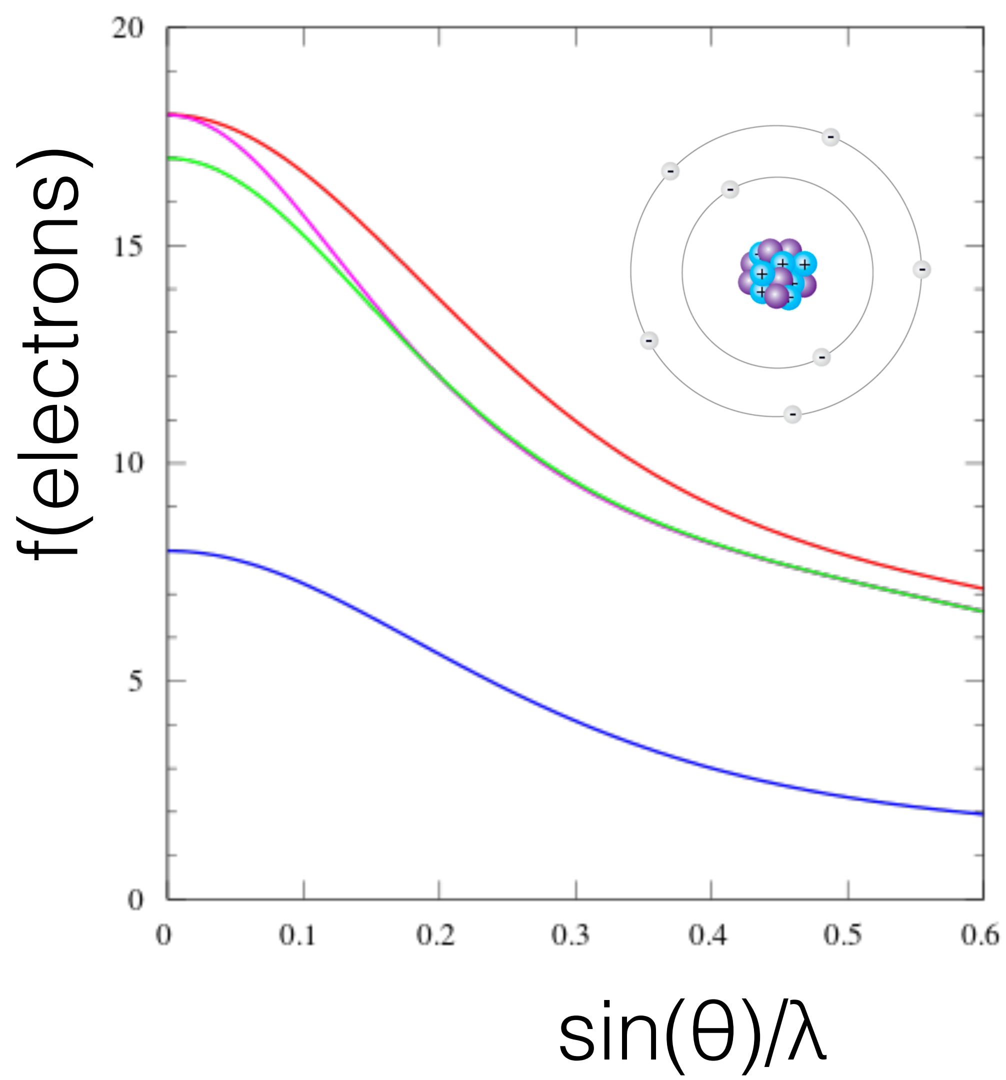
Static correlations

Form factors

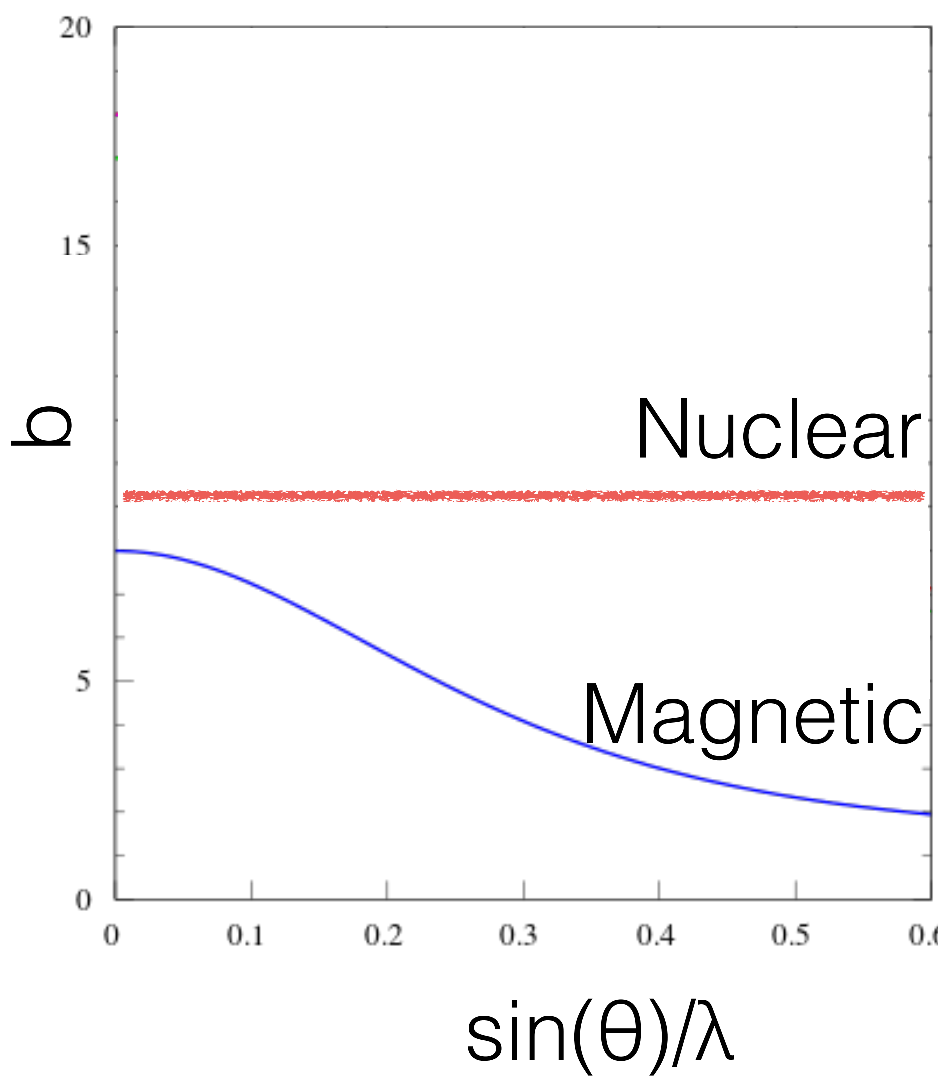
Neutrons interact with

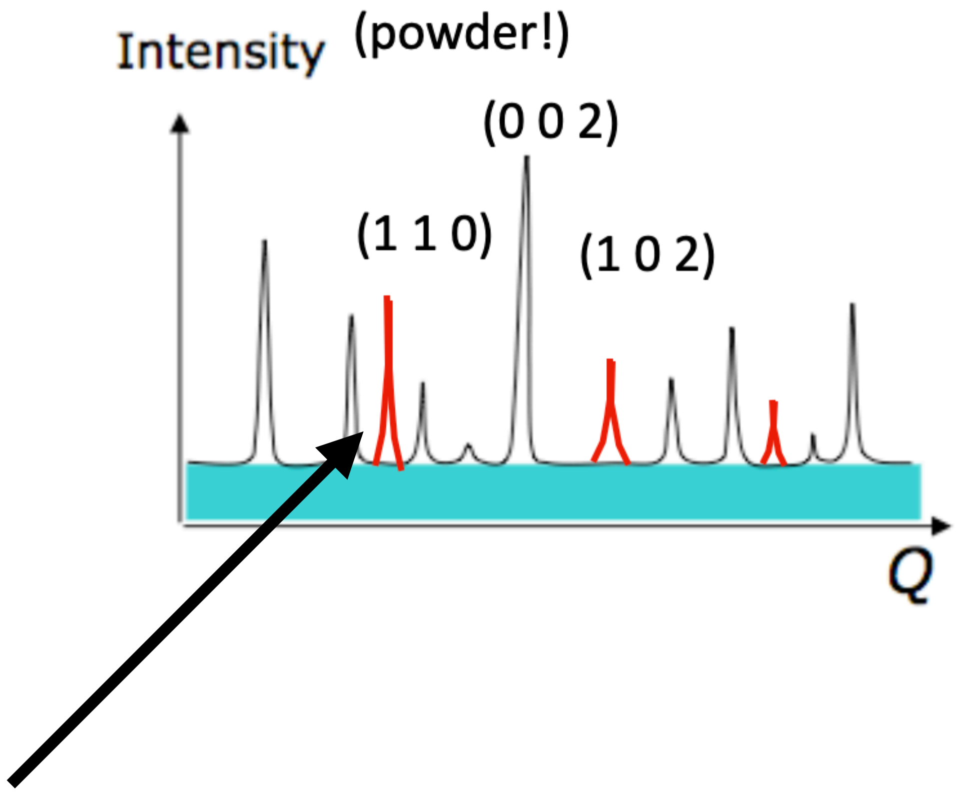
1. Atomic nuclei (strong nuclear force — short-range)
2. Magnetic fields from unpaired electrons (dipole-dipole interaction)

X-rays



Neutrons





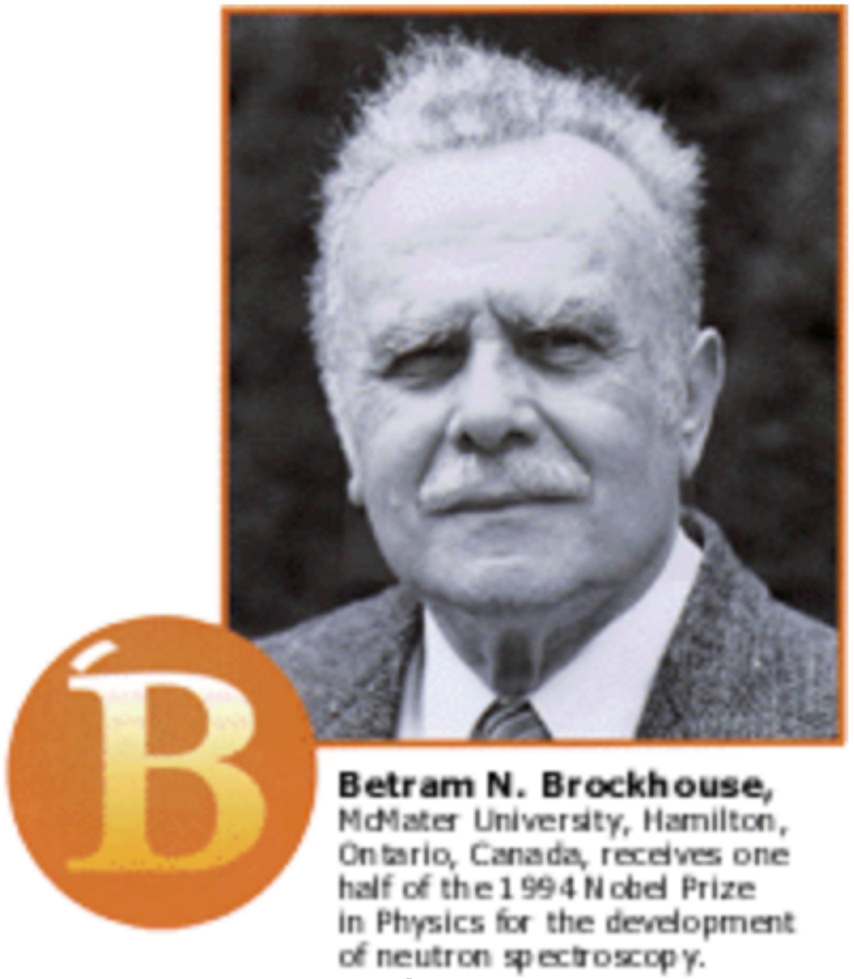
& also paramagnetic scattering

Overview

- Magnetism : a very basic overview
- **Neutron scattering and magnetism**
  - Diffraction
  - **Inelastic neutron scattering**
  - Polarisation analysis
- Recent examples of magnetic states of matter (Quantum behaviour)
- Overview of some instruments at ESS relevant to magnetism

Neutrons Interacting with matter

1994 Nobel prize: for pioneering contributions to the development of neutron scattering techniques for studies of condensed matter



Development of neutron spectroscopy

What atoms/spins do

Neutrons Interacting with matter

Solid State Communications, Vol. 11, pp. 391–394, 1972. Pergamon Press. Printed in Great Britain

INELASTIC NEUTRON SCATTERING STUDY OF SPIN WAVES IN MnO

M. Kohgi, Y. Ishikawa and Y. Endoh

Department of Physics, Tohoku University, Sendai 980, Japan

(*Received* 6 *May* 1972 *by* T. *Nagamiya*)

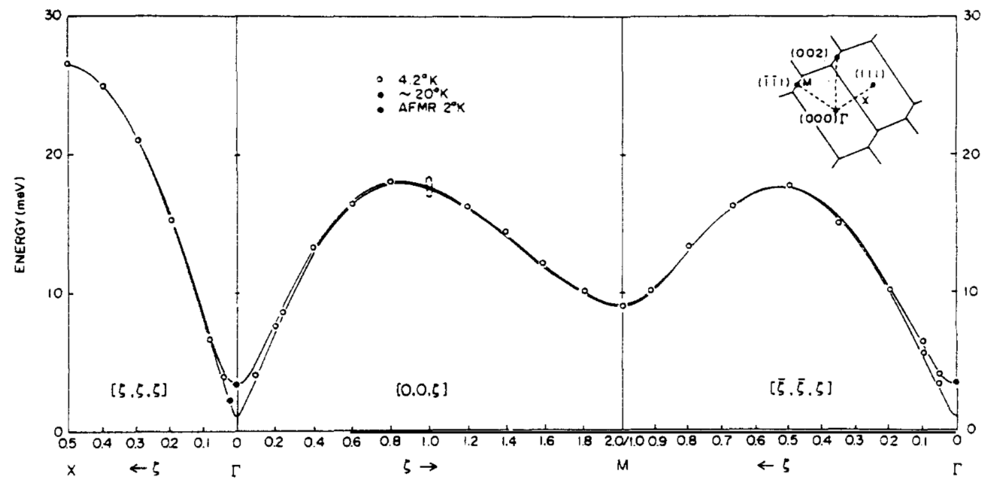
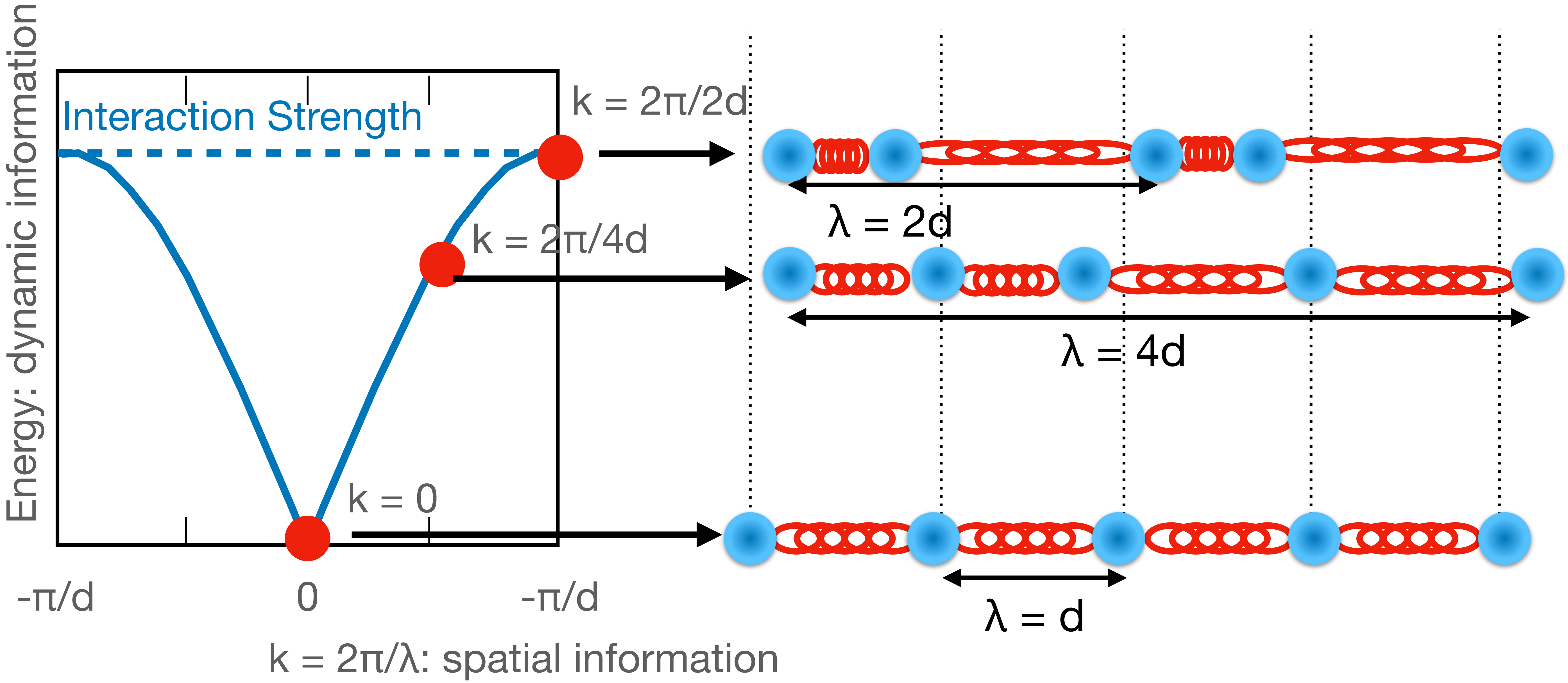


FIG. 1. Dispersion relation $E(q)$ of the spin waves in MnO measured along [111], [001] and $[\bar{1}\bar{1}1]$.

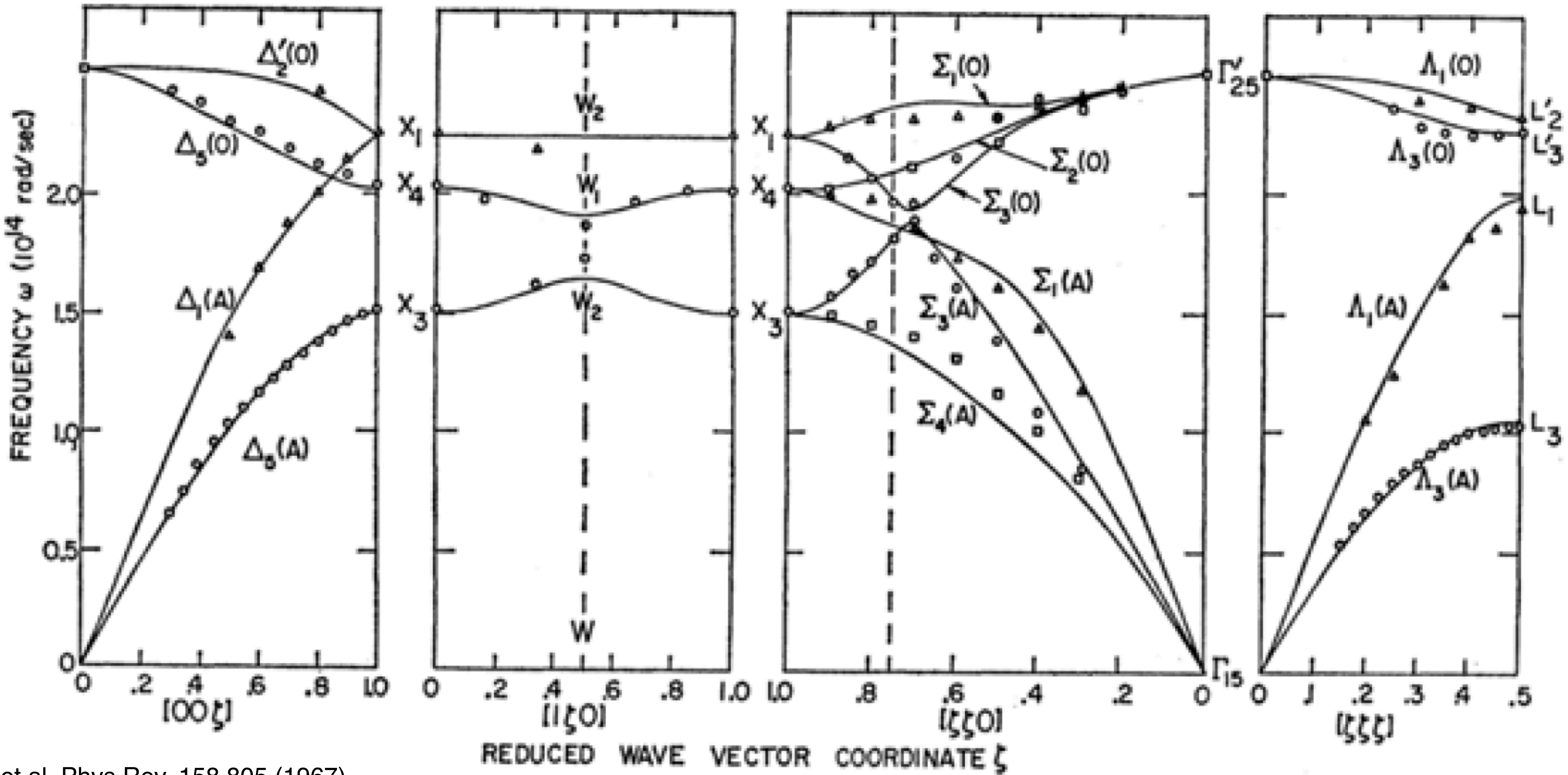
How they move

Inelastic scattering

Collective excitations: phonons (sound)



Inelastic scattering
Collective excitations: phonons
Phonons in diamond

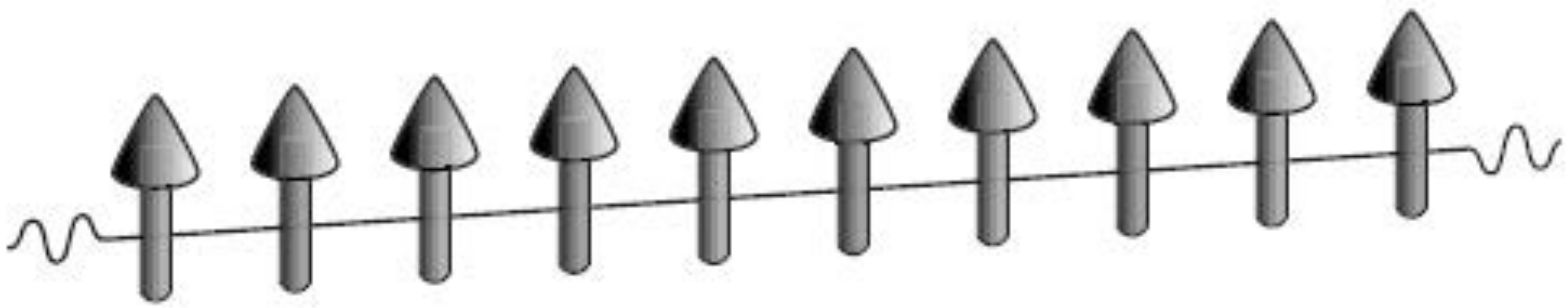


J.L. Warren et al. Phys.Rev. 158 805 (1967)

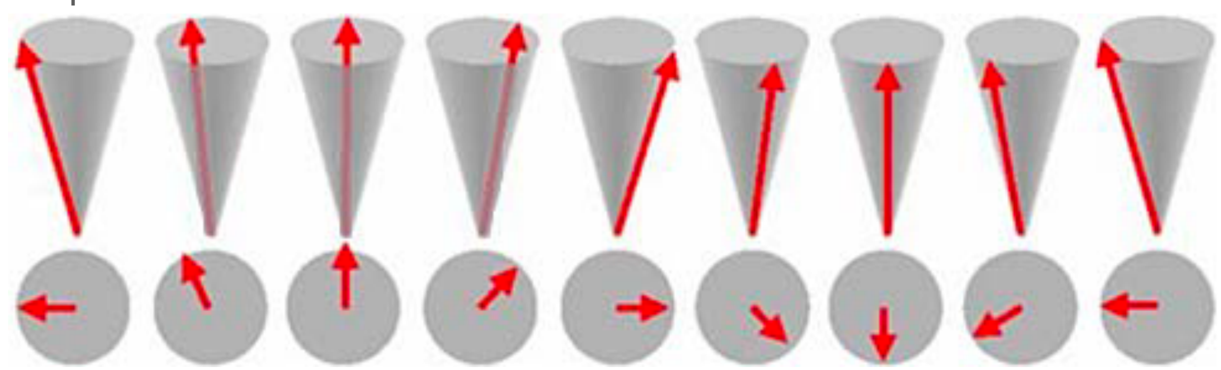
Inelastic scattering

Collective excitations: magnons

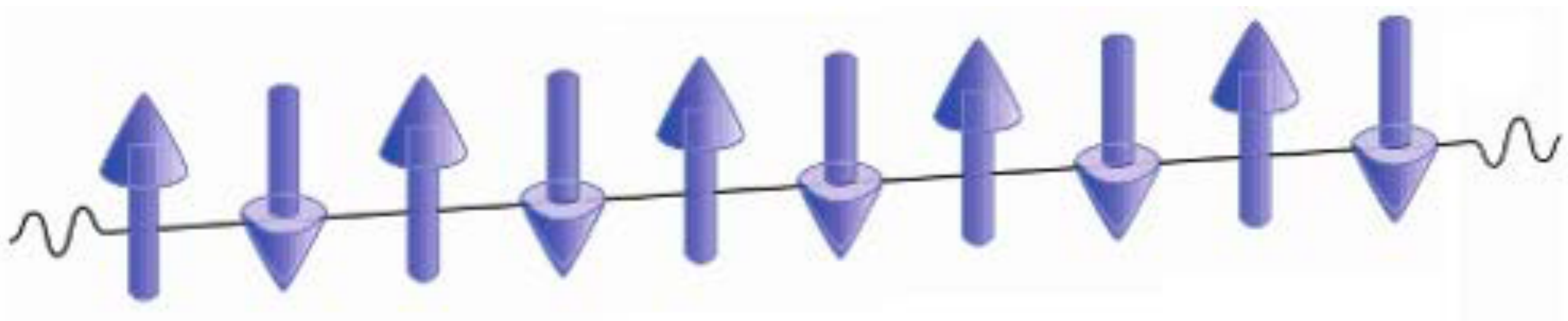
Ferromagnetism



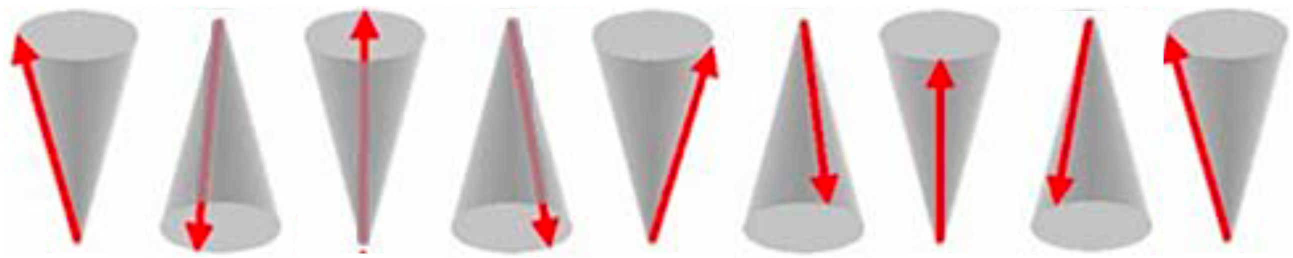
FM Spin wave



Antiferromagnetism



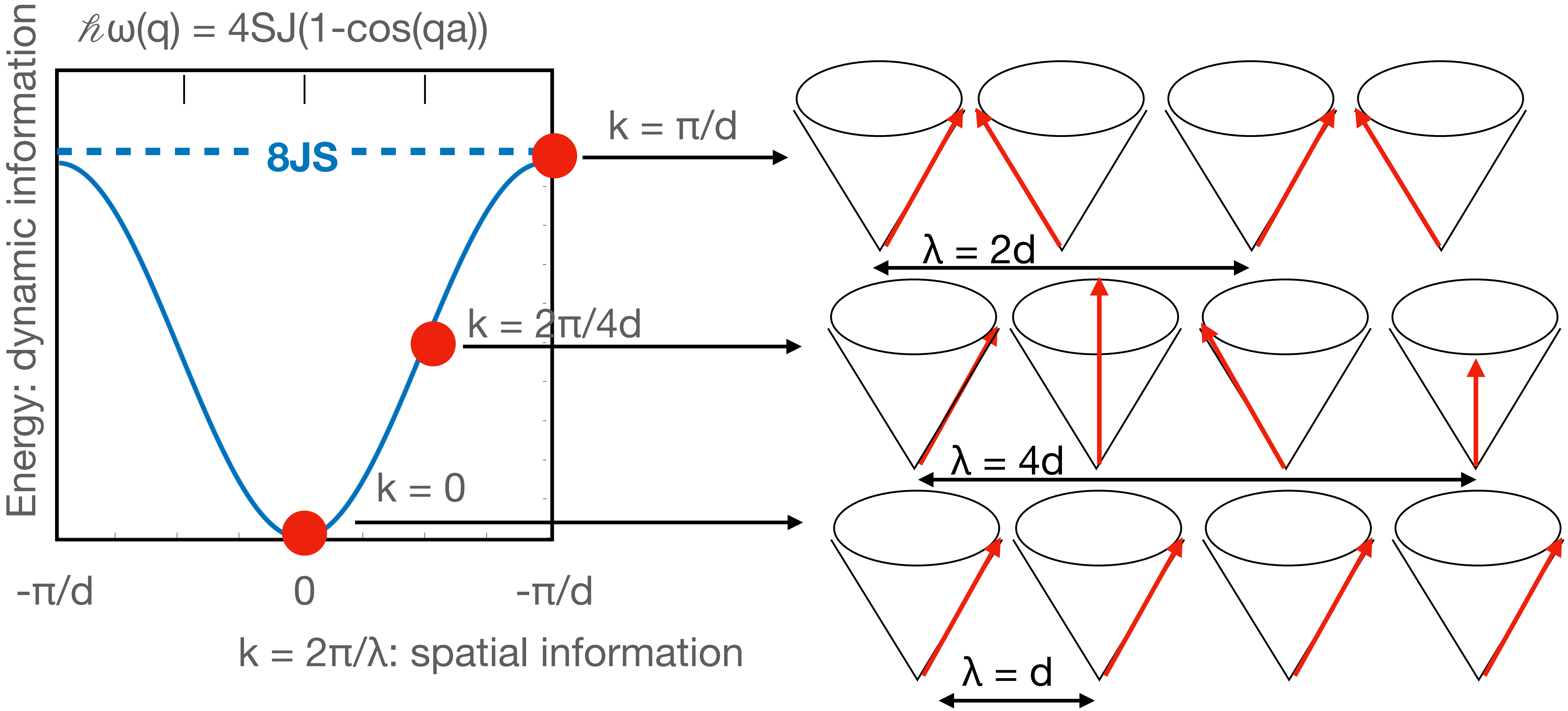
AFM Spin wave



https://www.ill.eu/fileadmin/user_upload/ILL/1_About_ILL/Films_and_animations/Animations/Scientific-animations/Magnons/activity/Magnetic_phases_and_Magnons.html

Collective excitations: magnons

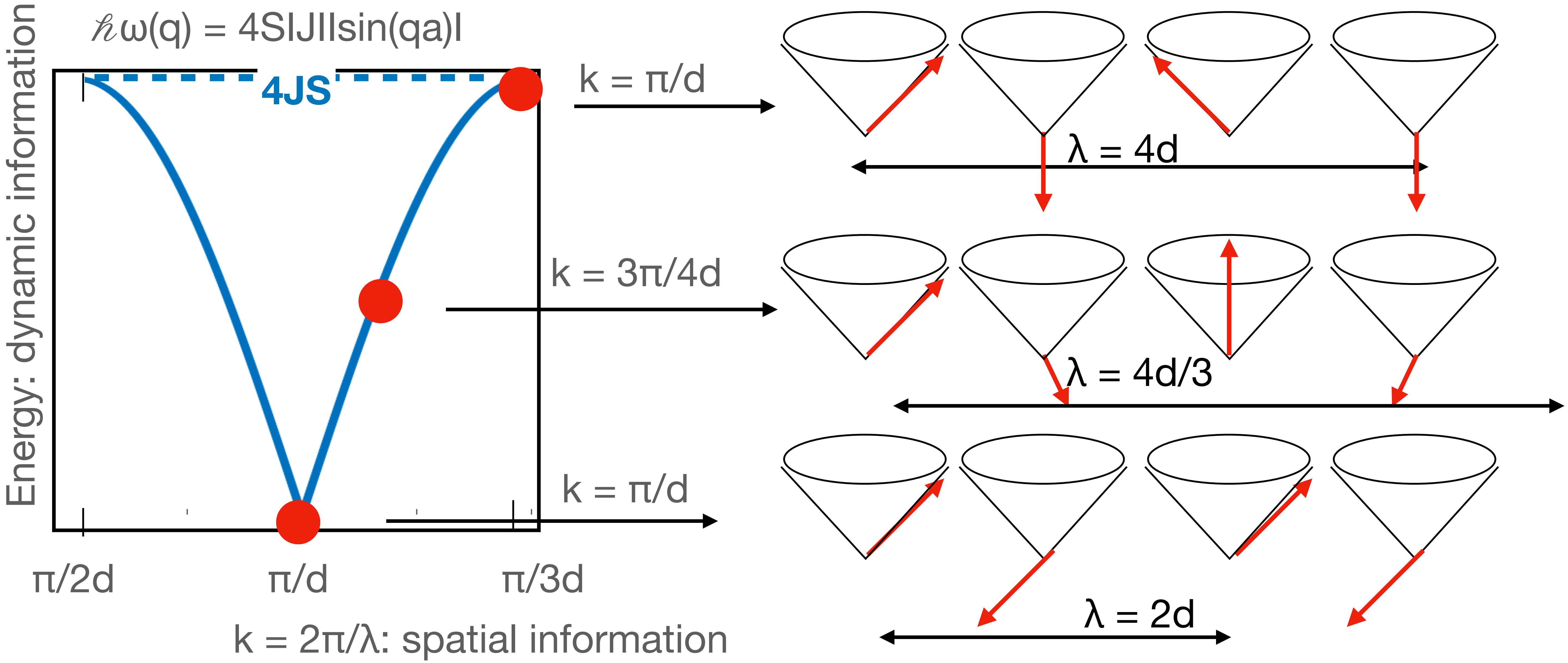
Ferromagnetic spin waves



Collective excitations: magnons

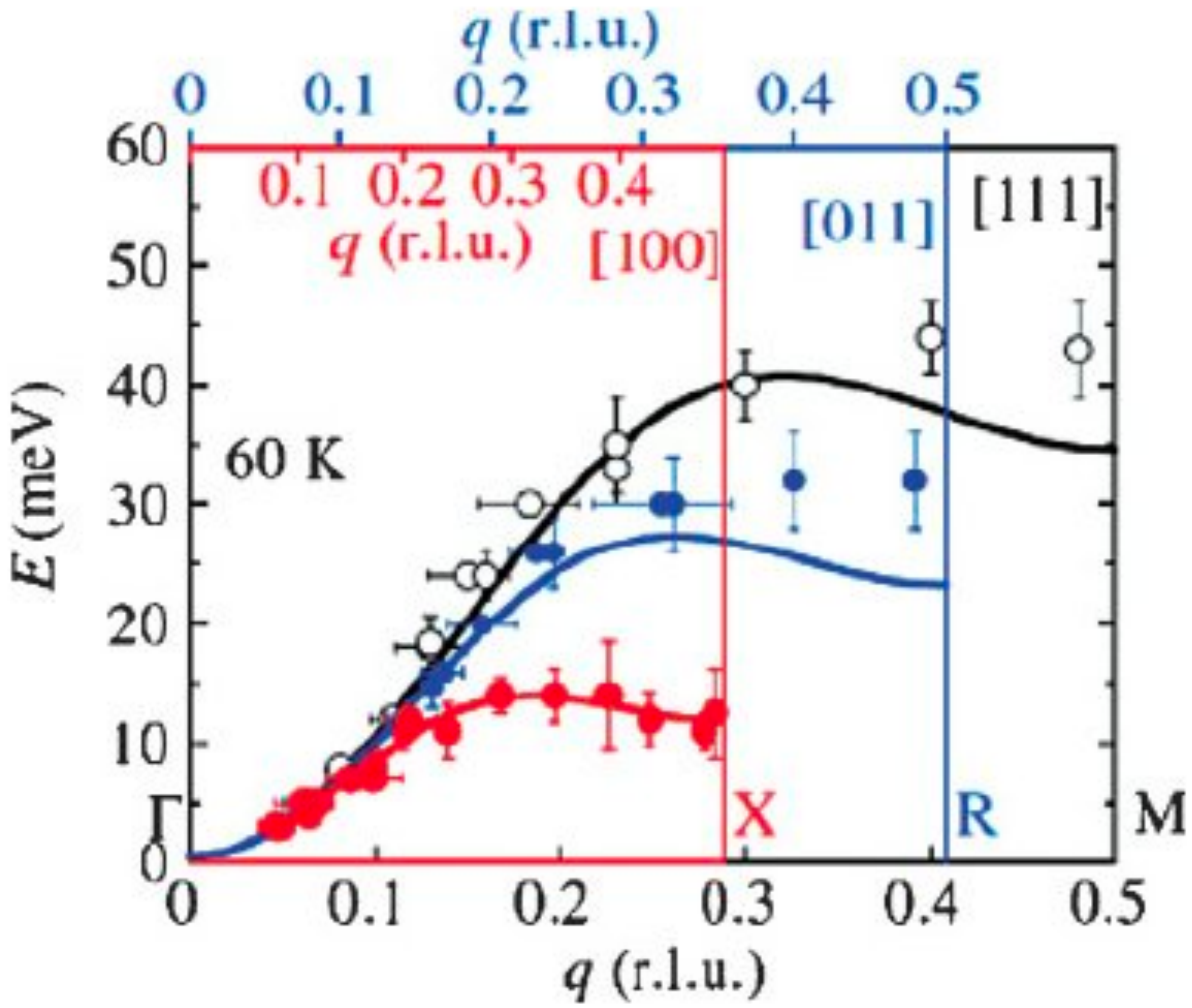
Antiferromagnetic spin waves

$\hbar\omega(q) = 4S|J||\sin(qa)|$



Collective excitations: FM vrs AFM

$Sm_{0.55}Sr_{0.45}MnO_3$



Y. Endoh, H. Hiraka, Y. Tomioka, Y. Tokura, N. Nagaosa and T. Fujiwara: Phys. Rev. Lett. 94 (2005) 17206.

INELASTIC NEUTRON SCATTERING STUDY OF SPIN WAVES IN MnO

M. Kohgi, Y. Ishikawa and Y. Endoh

Department of Physics, Tohoku University, Sendai 980, Japan

(*Received* 6 *May* 1972 *by T. Nagamiya*)

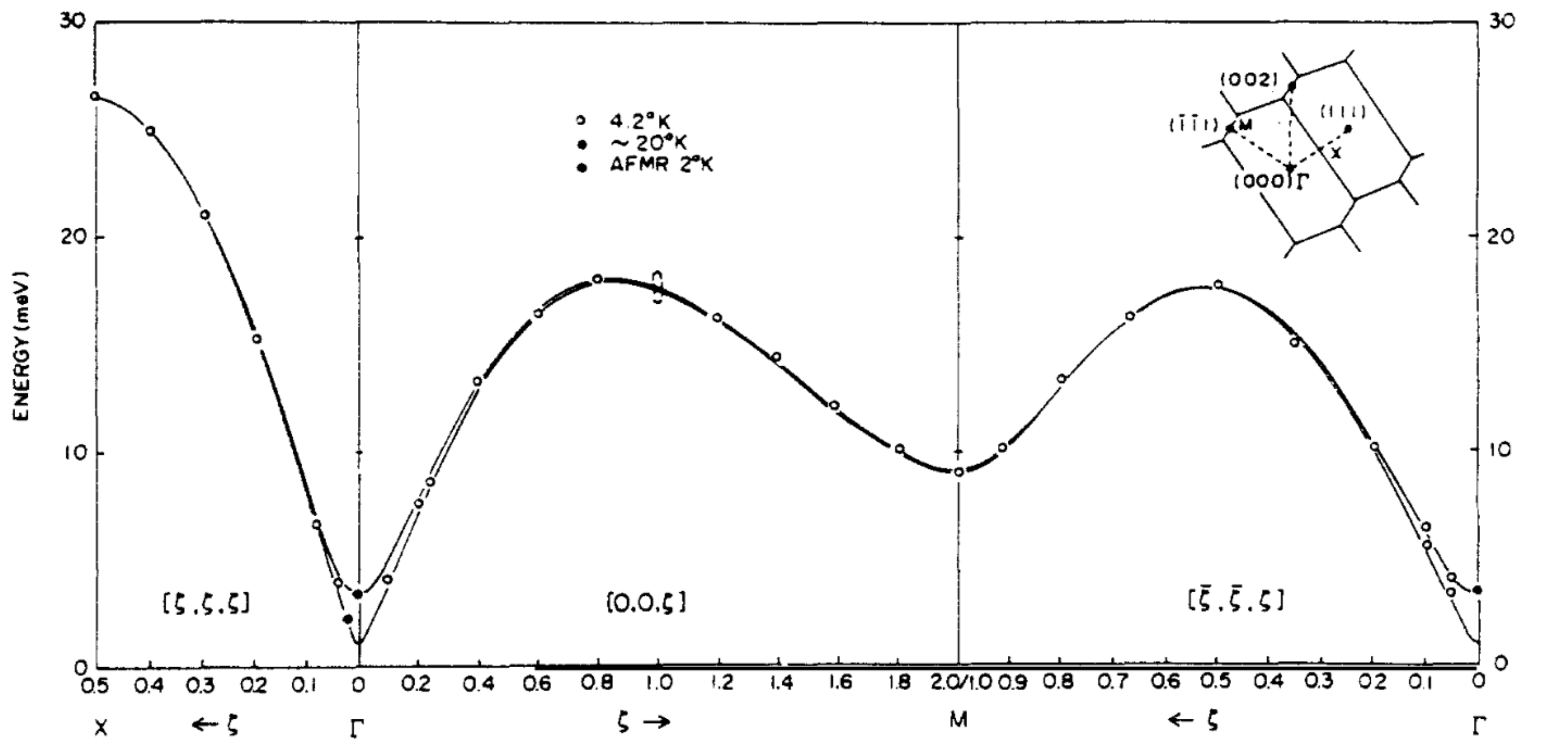
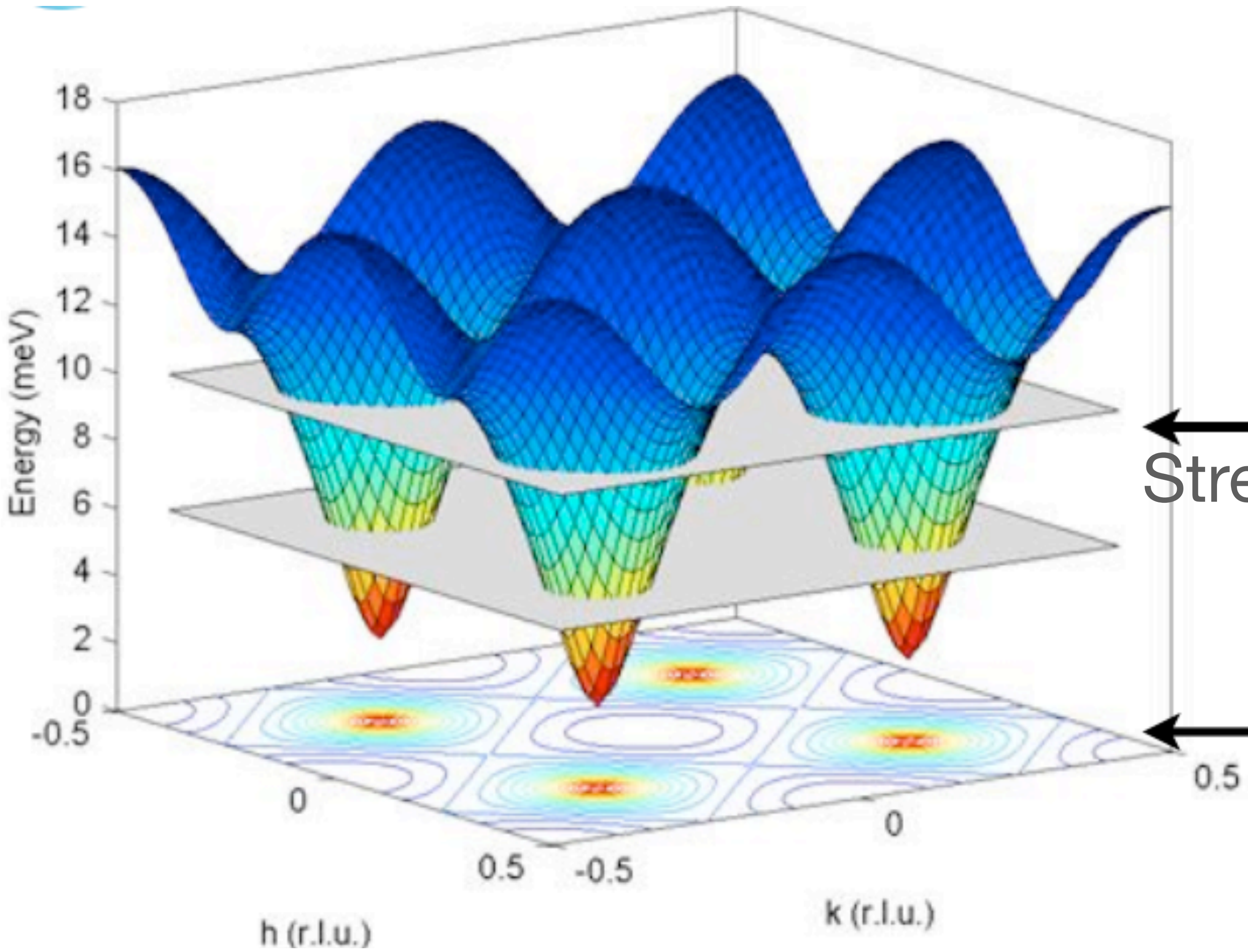


FIG. 1. Dispersion relation $E(q)$ of the spin waves in MnO measured along [111], [001] and [$\bar{1}\bar{1}$1].

S(**Q**,ω) on a single crystal = 4 D space (Qx,Qy,Qz, E)



Inelastic ($\Delta\omega \neq 0$)

Strength of the exchange interactions / anisotropy

Elastic ($\Delta\omega = 0$)

Information on the spin structure

Overview

- Magnetism : a very basic overview
- **Neutron scattering and magnetism**
  - Diffraction
  - Inelastic neutron scattering
  - **Polarisation analysis**
- Recent examples of magnetic states of matter (Quantum behaviour)
- Overview of some instruments at ESS relevant to magnetism

History of neutron polarisation analysis

Phys. Rev. 181, 920 (1969)

PHYSICAL REVIEW VOLUME 181, NUMBER 2 10 MAY 1969

Polarization Analysis of Thermal-Neutron Scattering*

R. M. Moon, T. Riste,† and W. C. Koehler
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
(Received 30 December 1968)

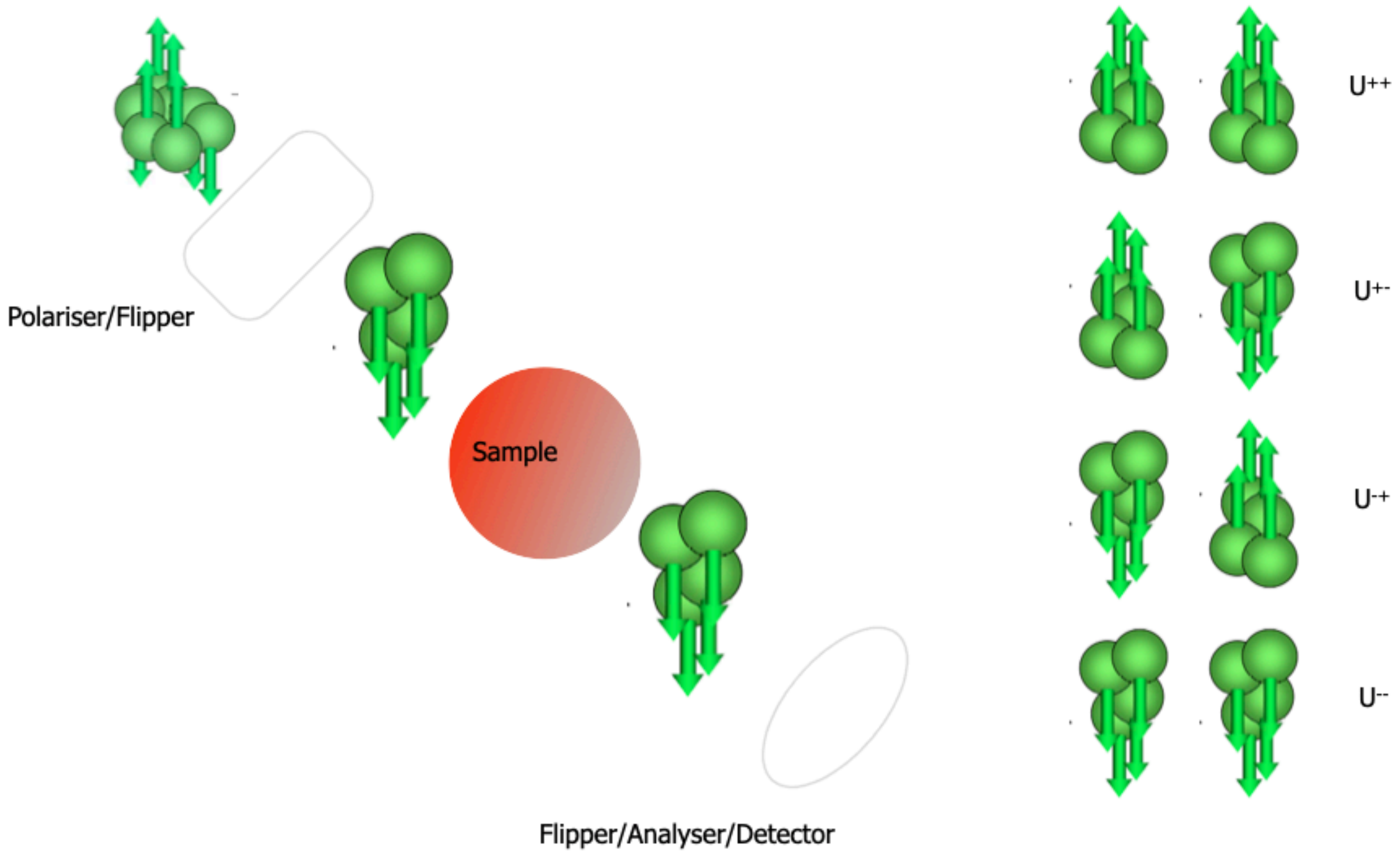
A triple-axis neutron spectrometer with polarization-sensitive crystals on both the first and third axes is described. The calculation of polarized-neutron scattering cross sections is presented in a form particularly suited to apply to this instrument. Experimental results on nuclear incoherent scattering, paramagnetic scattering, Bragg scattering, and spin-wave scattering are presented to illustrate the possible applications of neutron-polarization analysis.

neutron spin is either spin up $|\uparrow\rangle = \begin{pmatrix}1\\0\end{pmatrix}$ or spin down $|\downarrow\rangle = \begin{pmatrix}0\\1\end{pmatrix}$

1st comprehensive experimental work (Bragg scattering, Incoherent scattering, spin wave scattering)
Illustration of Polarisation Analysis

History of neutron polarisation analysis

neutron spin is either spin up $|\uparrow\rangle = \begin{pmatrix}1\\0\end{pmatrix}$ or spin down $|\downarrow\rangle = \begin{pmatrix}0\\1\end{pmatrix}$



History of neutron polarisation analysis

Moon, Riste Koehler, Phys. Rev. 181 (1969) 920.

MnF_2 = simple antiferromagnet

Measurement of formfactor above Tn

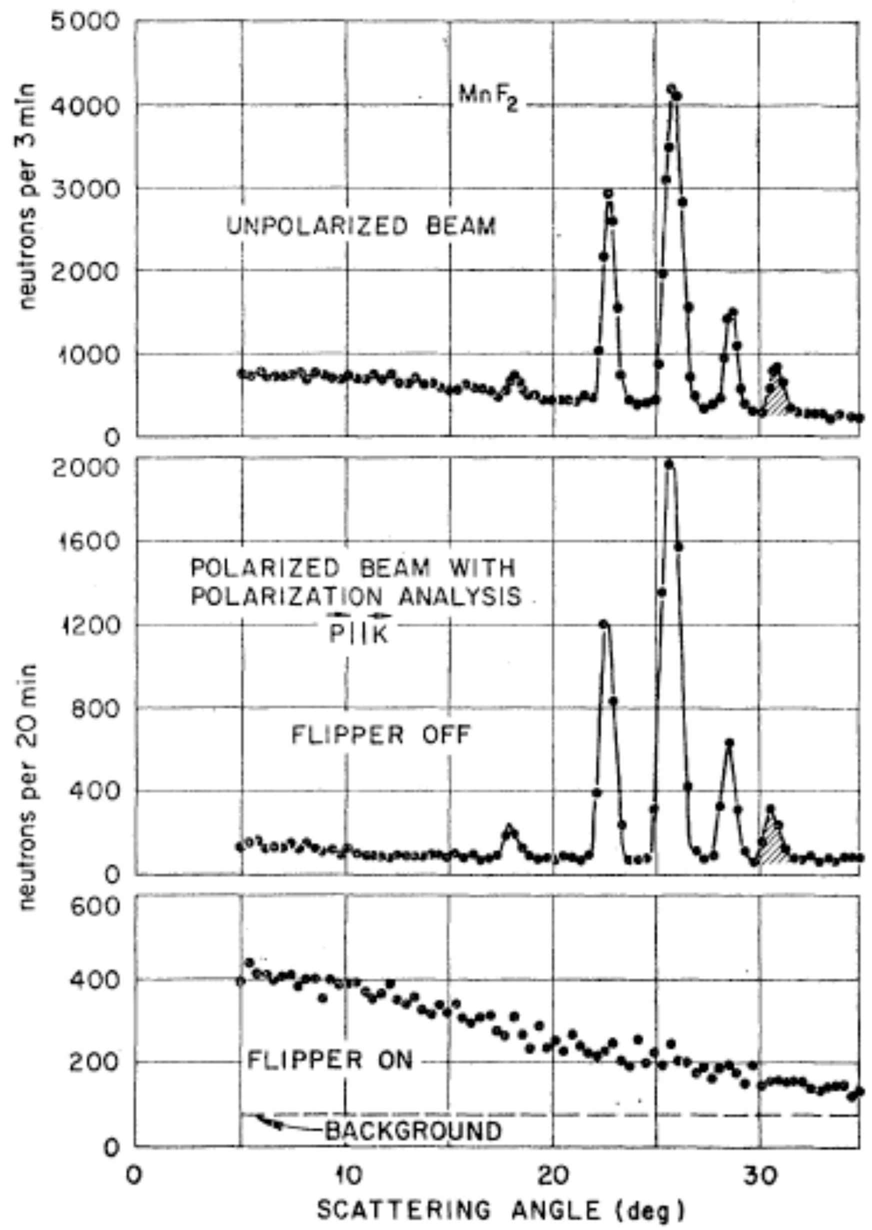


FIG. 5. MnF_2 powder pattern—separation of paramagnetic scattering through polarization analysis. No analyzer was used in the unpolarized-beam experiment. Note the loss of intensity in the polarization analysis experiment.

Unpolarised off

Flipper off

Flipper on

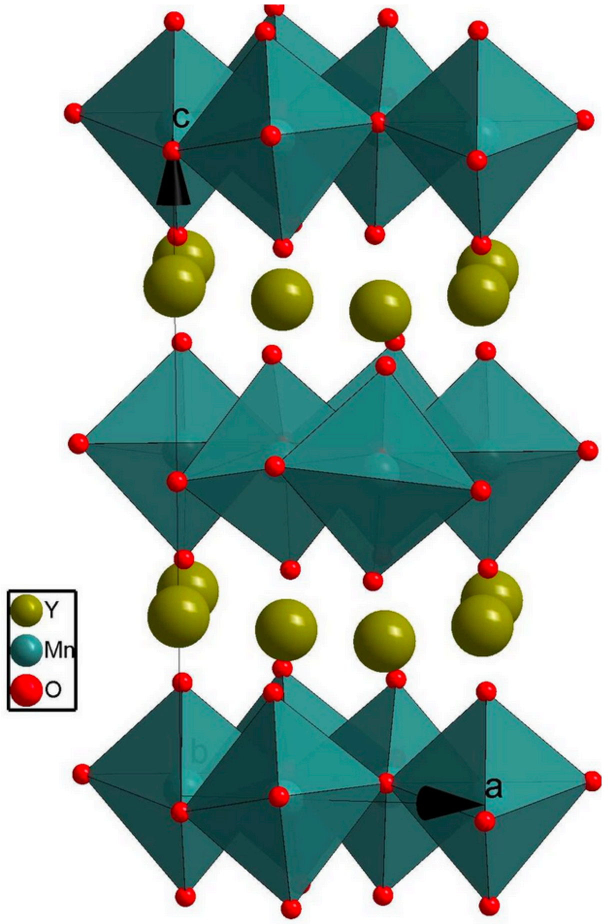
Overview

- Magnetism : a very basic overview
- **Neutron scattering and magnetism**
  - Diffraction
  - Inelastic neutron scattering
  - Polarisation analysis
- Recent examples of magnetic states of matter (Quantum behaviour)
- Overview of some instruments at ESS relevant to magnetism

2 examples:
Elucidate typical neutron scattering signals today

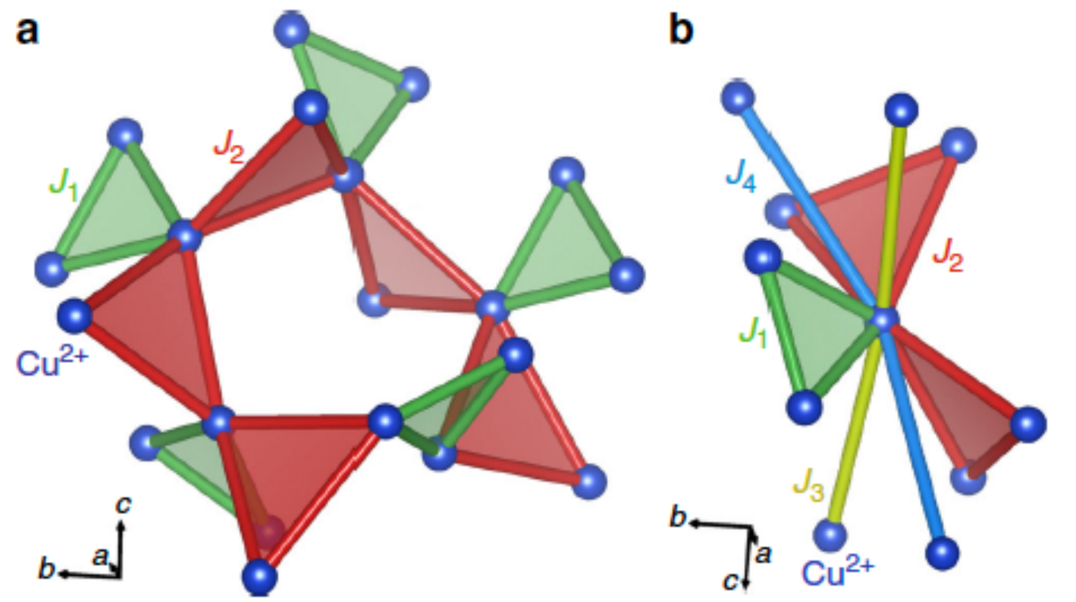
EUROPEAN SPALLATION SOURCE

$YMnO_3$ Multiferroicity
Ferrromagnetic & Ferroelectricity
Interplay between magnon & phonons



Phys. Rev. B 97, 134304 (2018)

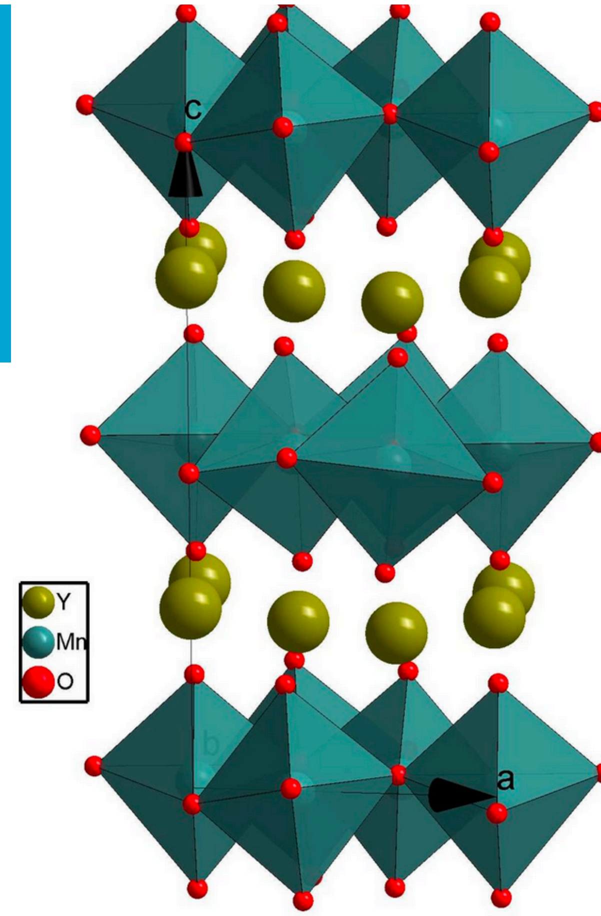
3D quantum spin liquid in PbCuTe2O6



NATURE COMMUNICATIONS | https://doi.org/10.1038/s41467-020-15594-1

$YMnO_3$ Multiferroicity

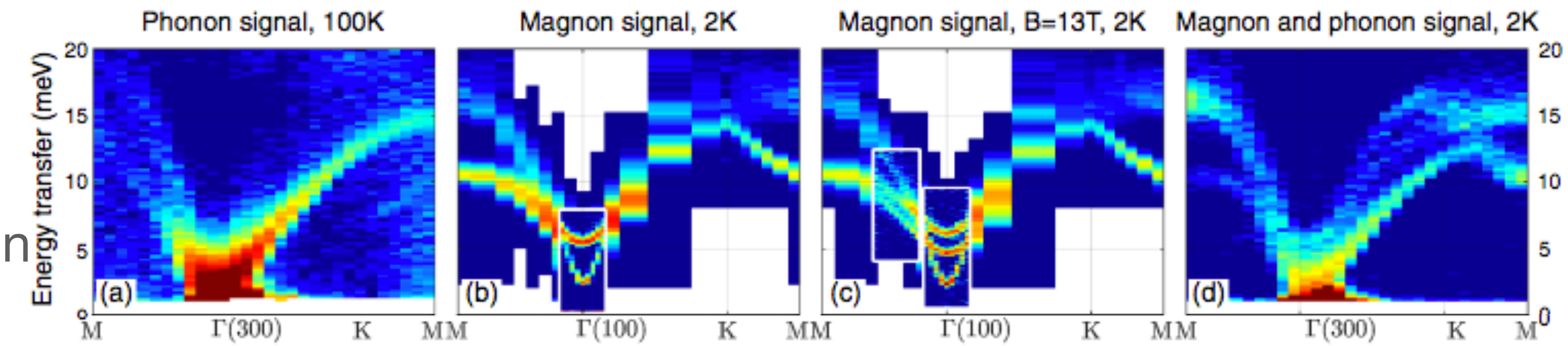
ΔE = 0 - 30 meV



Magnetic ground state and magnon-phonon interaction in multiferroic *h*-$YMnO_3$

S. L. Holm,[1,*] A. Kreisel,[1,2] T. K. Schäffer,[1] A. Bakke,[1] M. Bertelsen,[1] U. B. Hansen,[1] M. Retuerto,[1,3] J. Larsen,[4]
D. Prabhakaran,[5] P. P. Deen,[6] Z. Yamani,[7] J. O. Birk,[1,8] U. Stuhr,[8] Ch. Niedermayer,[8]
A. L. Fennell,[8] B. M. Andersen,[1] and K. Lefmann[1]

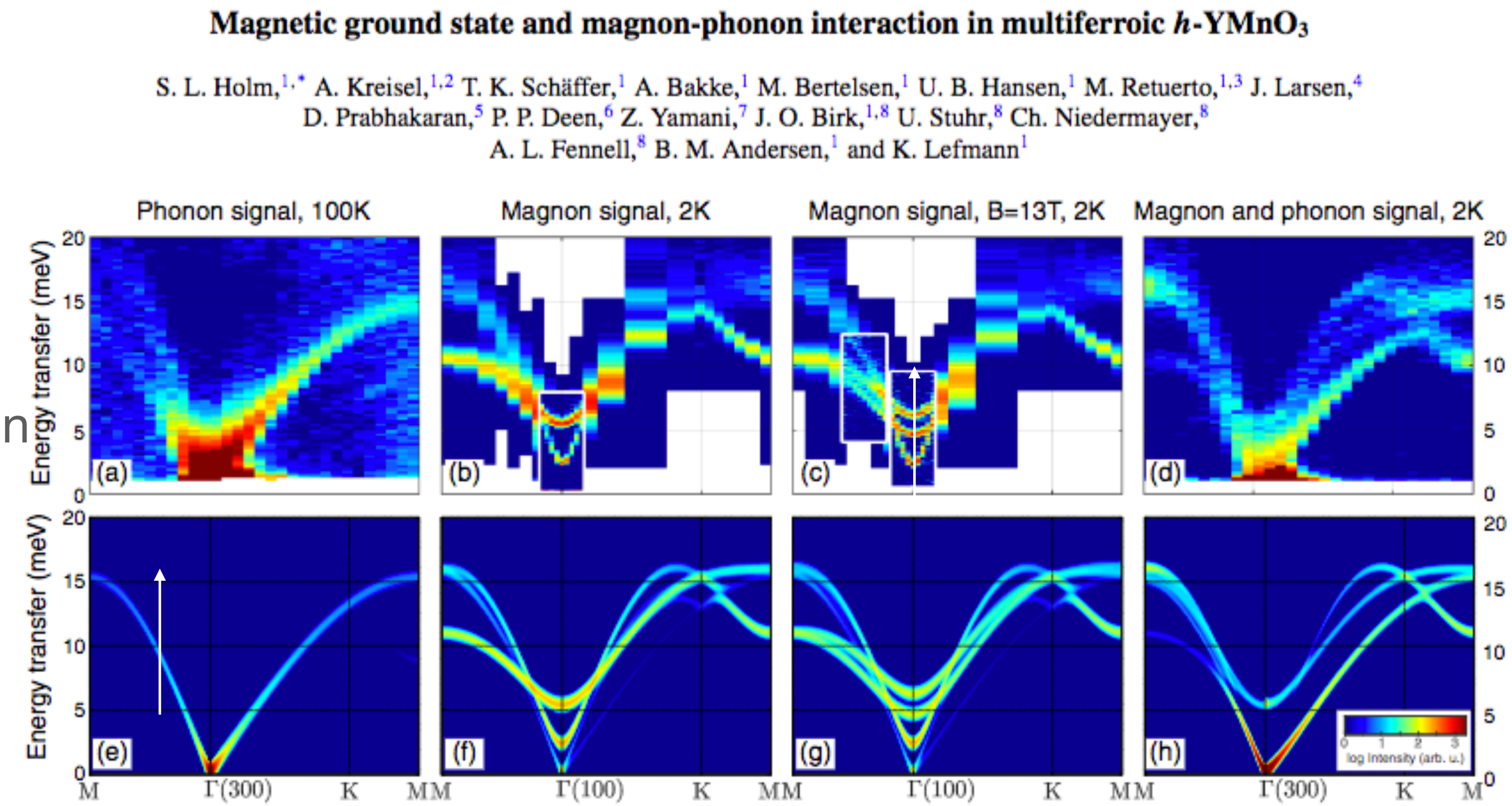
J1, J2
Anisotropy
M/P Hybridisation

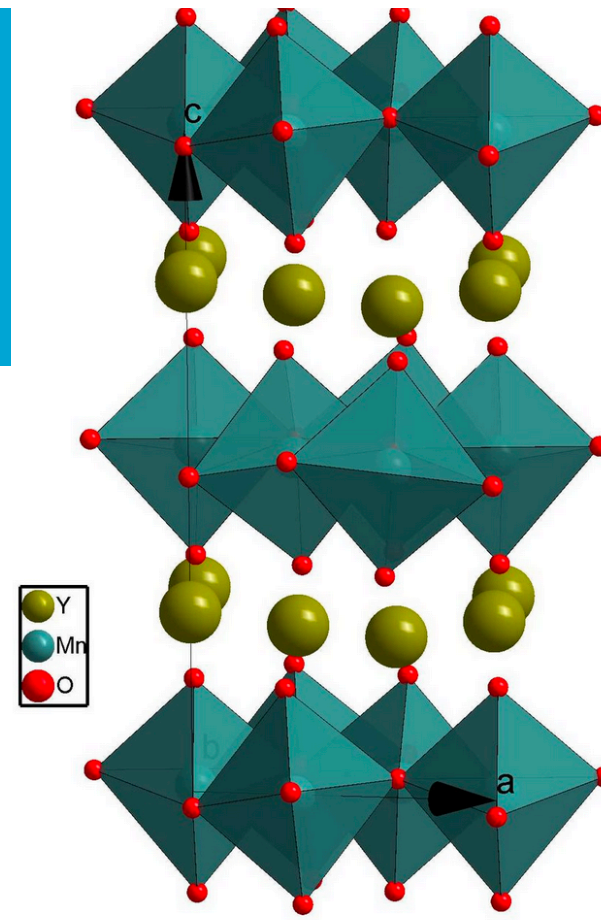


Phys. Rev. B 97, 134304 (2018)

$YMnO_3$ Multiferroicity
ΔE = 0 - 30 meV

J1, J2
Anisotropy
M/P Hybridisation





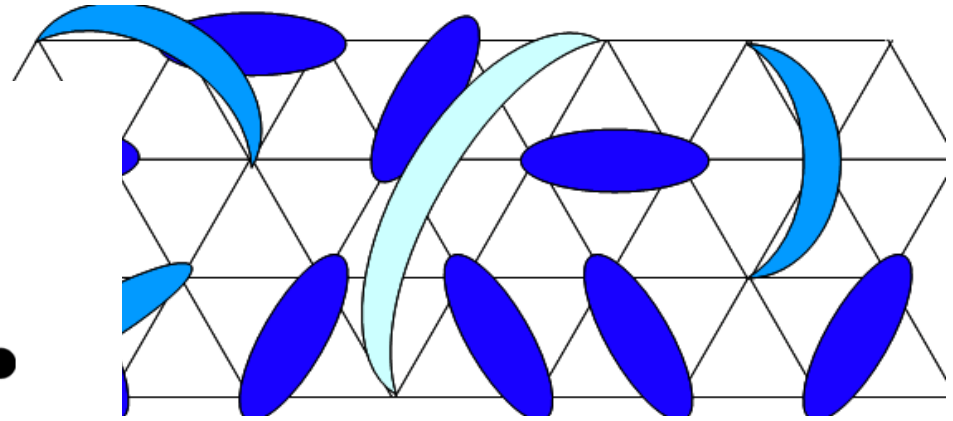
Need to study broad region of S(Q, omega)
High energy resolution
Signal to noise > 10^4
Interplay between phonons and magnon

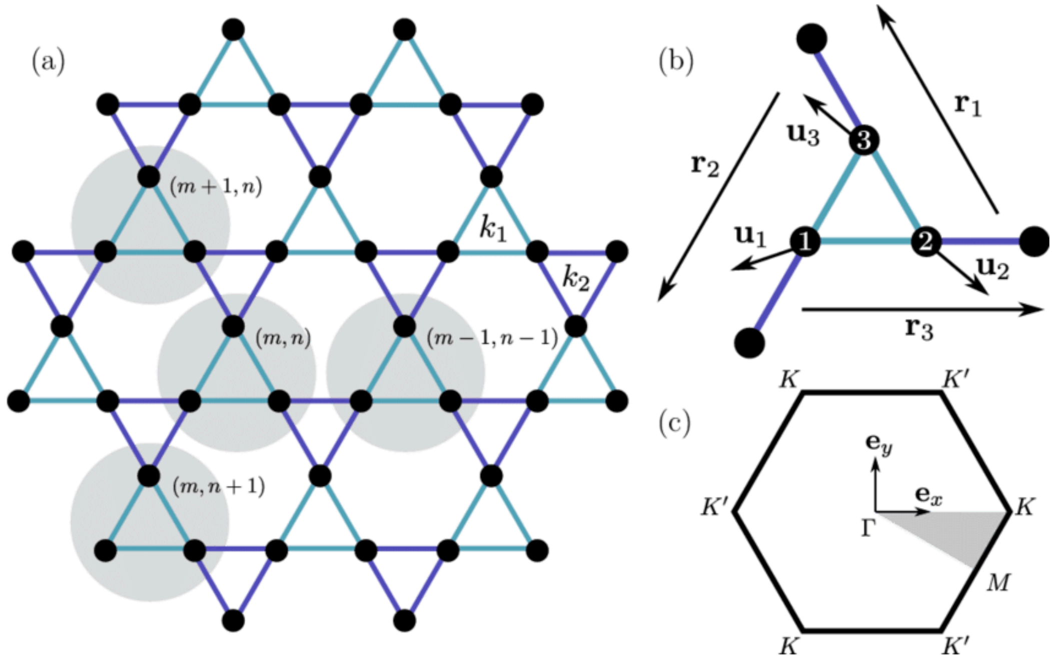
Phys. Rev. B 97, 134304 (2018)

Quantum Spin liquids

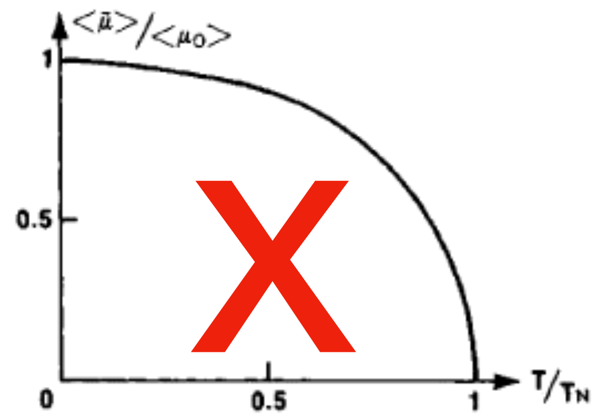
Understand, drive and manipulate quantum effects

- Coherence, entanglement, superposition, quantum transport.
- Quantum computing





A Regular Kagome Lattice - Kagome Lattice Brillouin Zone

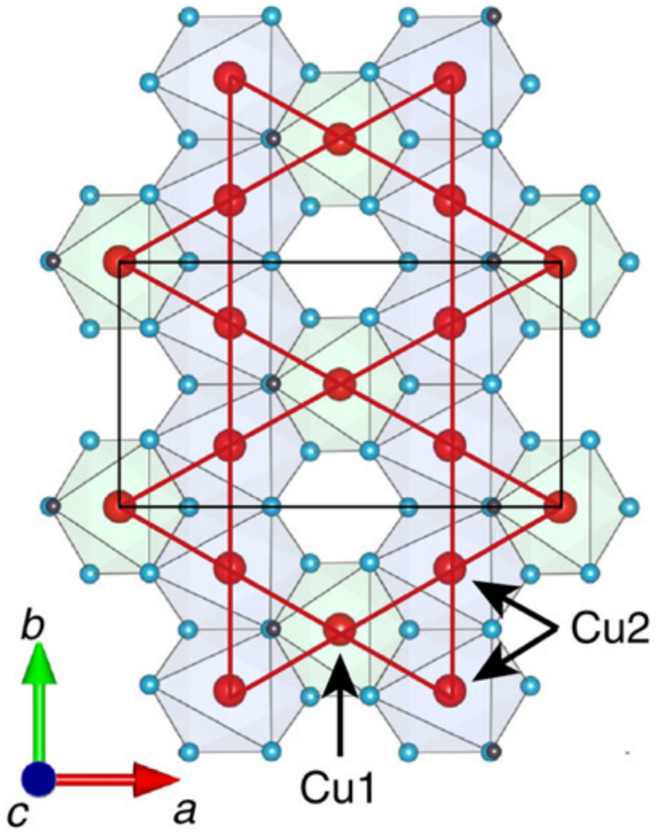


Signatures:

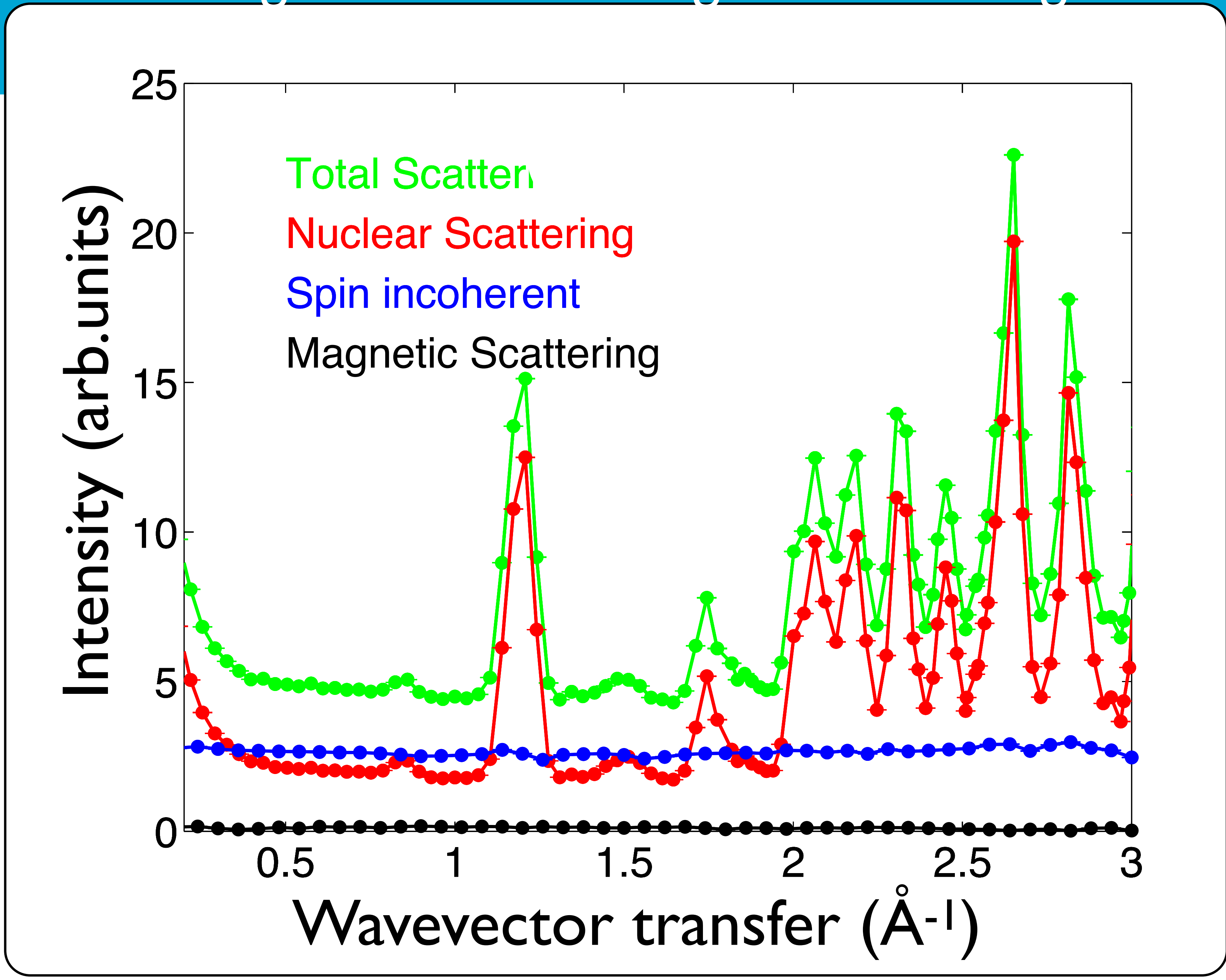
- A lack of broken symmetry
- No magnetic order (S = 1/2)
- Fractionalised excitations

Volborthite ($Cu_3V_2O_7(OH)_2 \cdot 2H_2O$) (powder)

S = 1/2 Kagome Heisenberg antiferromagnet



Phys. Rev. B 84, 172401

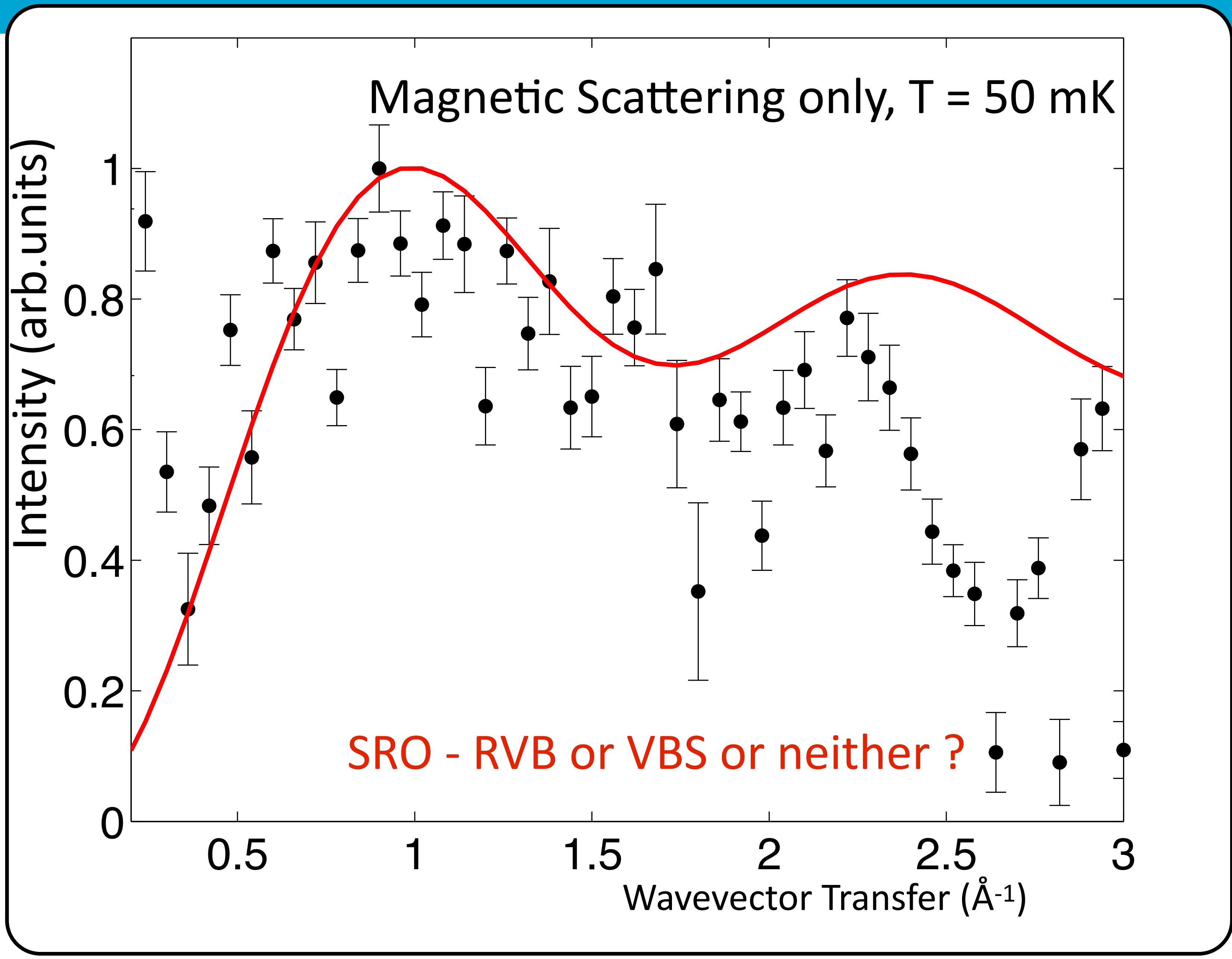


XYZ neutron polarisation analysis, D7 @ ILL, T = 50 mK

Spin liquid?

A lack of broken symmetry

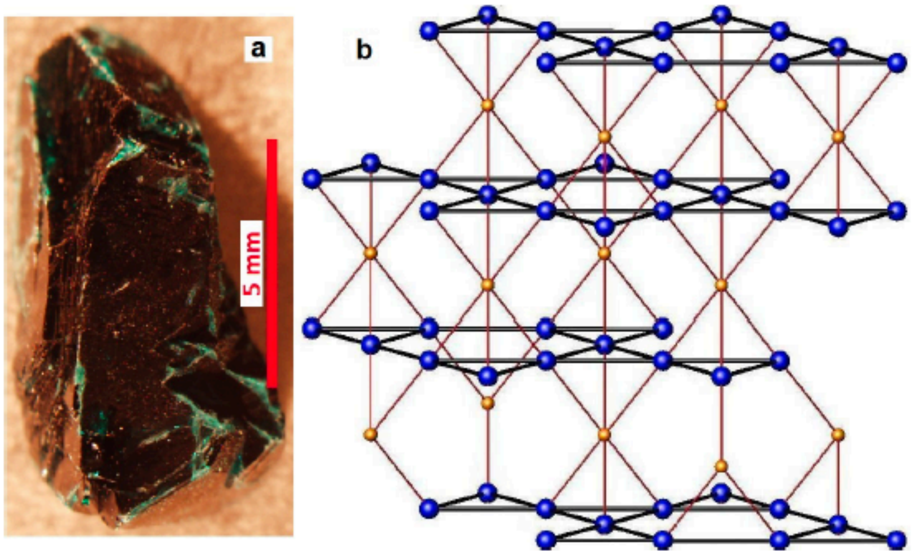
No magnetic order (S = 1/2)

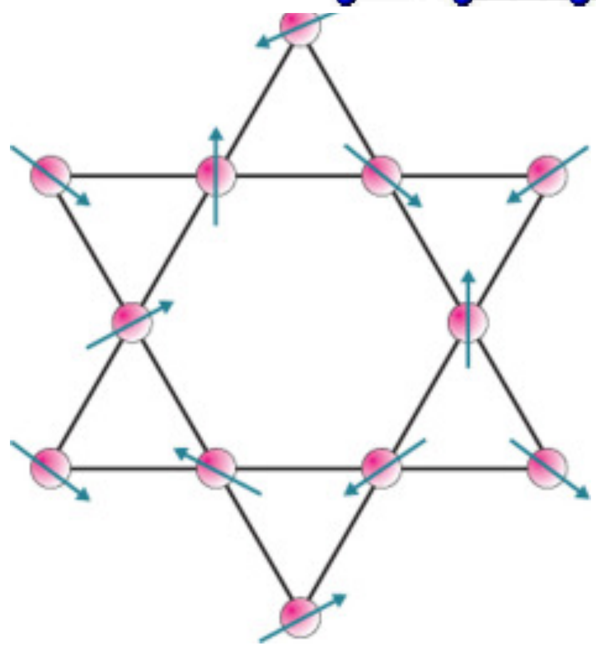


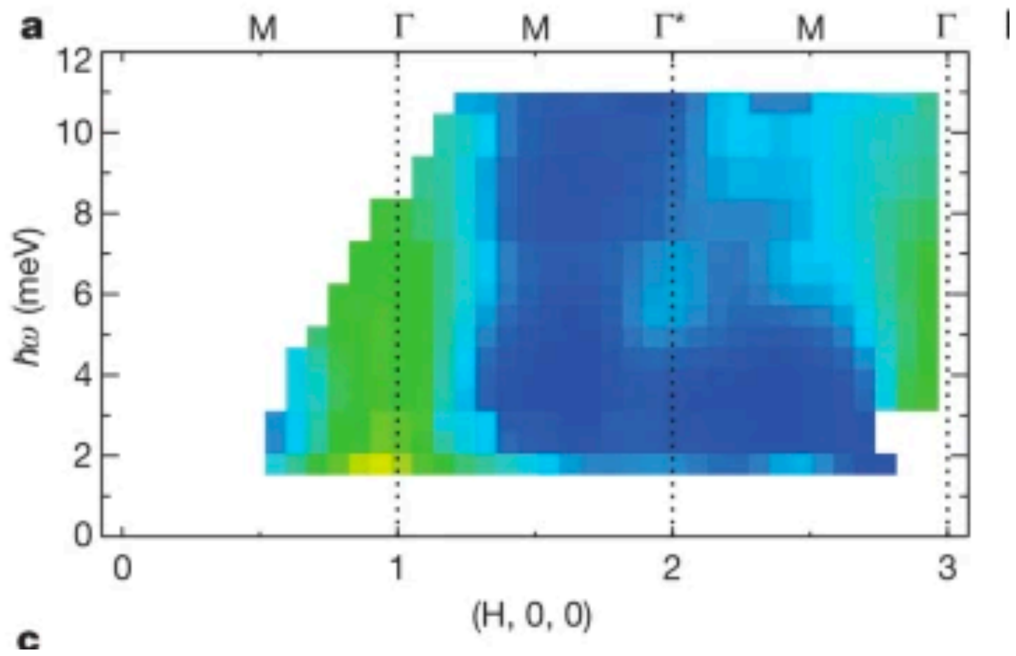
Determination not possible without Polarisation Analysis

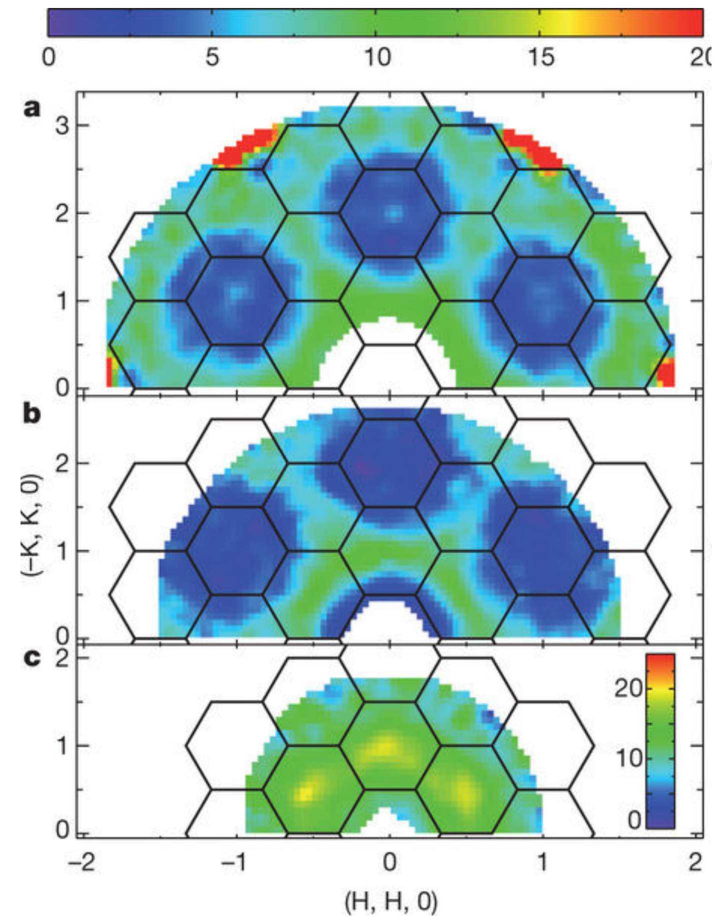
A 2D spin liquid state in Herbertsmithite $ZnCu_3(OD)_6Cl_2$

Nature **492**, 406 ,*Nature Phys.***12**, 942









Inelastic neutron scattering measured along symmetry directions and at high symmetry locations. T = 1.6 K.
NB: No well defined excitations & correlations.
Fractionalised excitations.

A 3D spin liquid state in PbCuTe2O6

EUROPEAN SPALLATION SOURCE

nature COMMUNICATIONS

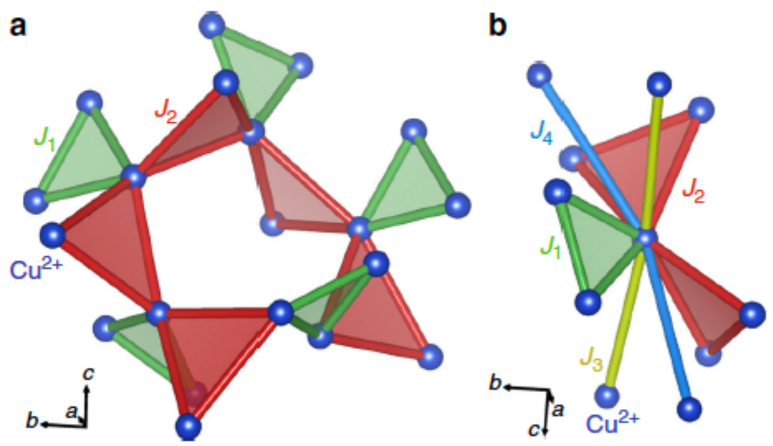
ARTICLE

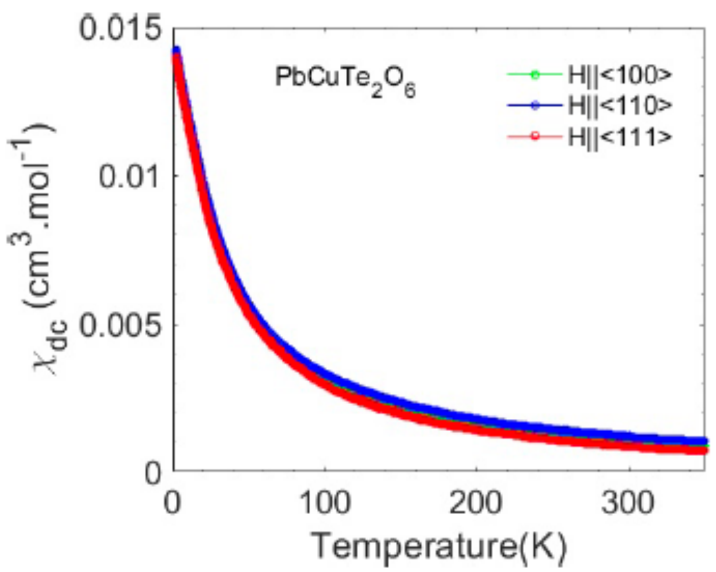
https://doi.org/10.1038/s41467-020-15594-1 OPEN

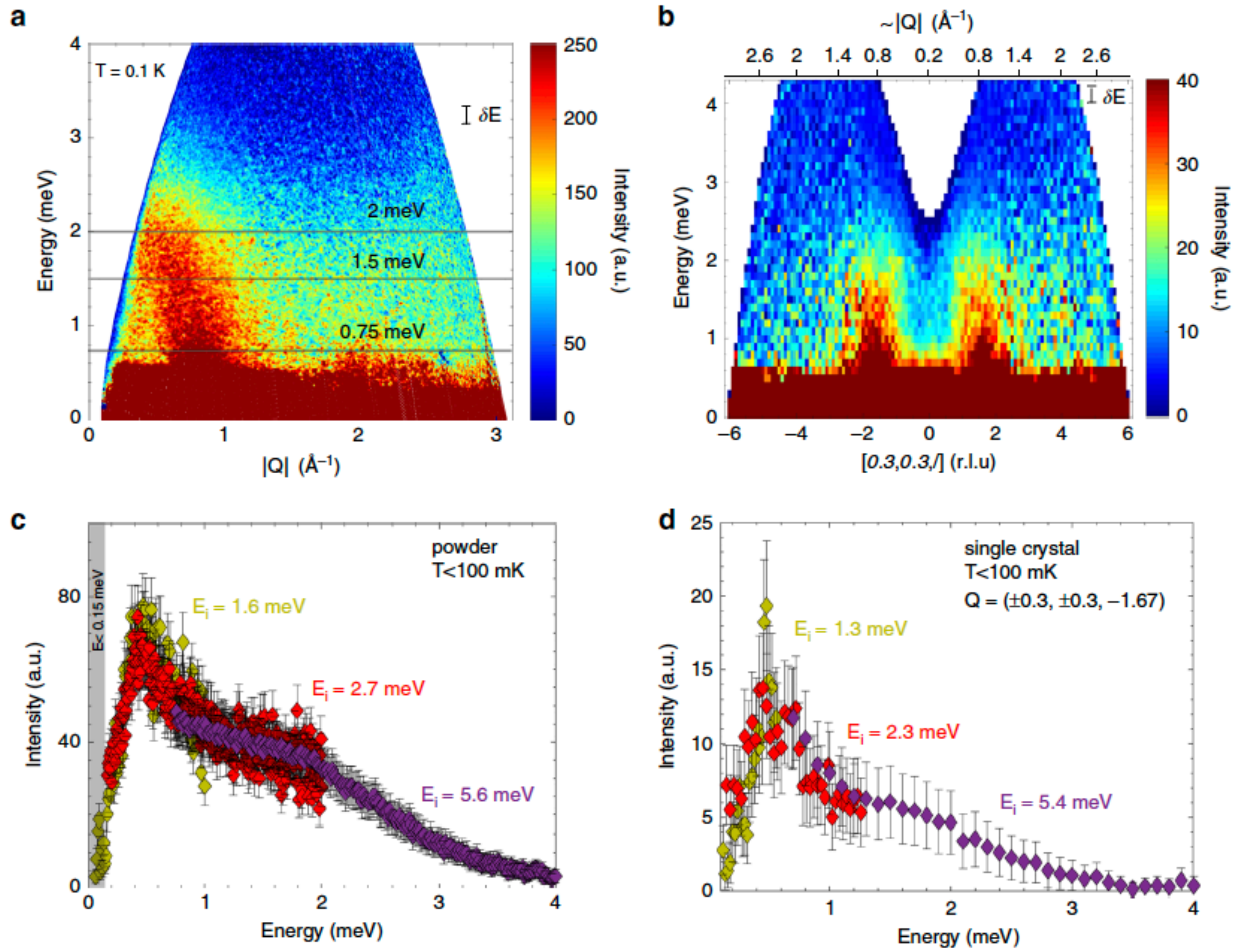
Check for updates

Evidence for a three-dimensional quantum spin liquid in $PbCuTe_2O_6$

Shravani Chillal[1✉], Yasir Iqbal[2], Harald O. Jeschke[3], Jose A. Rodriguez-Rivera[4,5], Robert Bewley[6], Pascal Manuel[6], Dmitry Khalyavin[6], Paul Steffens[7], Ronny Thomale[8], A. T. M. Nazmul Islam[1], Johannes Reuther[1,9] & Bella Lake[1,10]





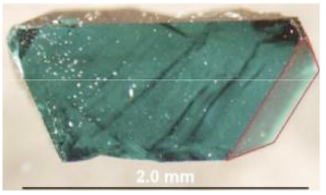


Overview

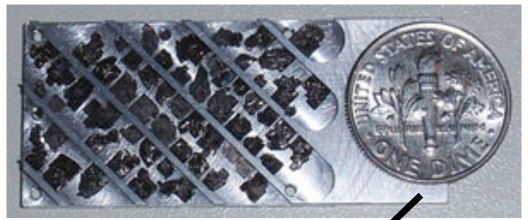
- Magnetism : a very basic overview
- **Neutron scattering and magnetism**
  - Diffraction
  - Inelastic neutron scattering
  - Polarisation analysis
- Recent examples of magnetic states of matter (Quantum behaviour)
- Overview of some instruments at ESS relevant to magnetism

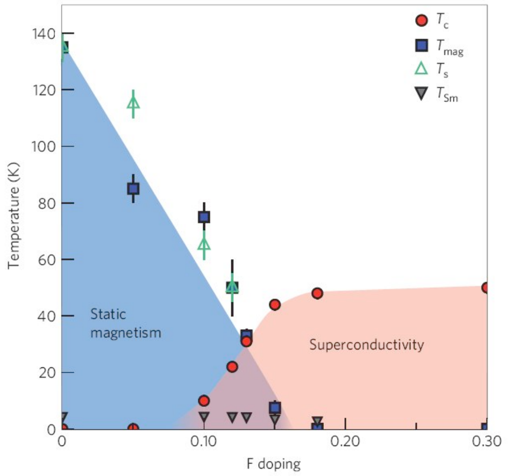
Neutron scattering at ESS

Current: Fe-arsenide single crystals



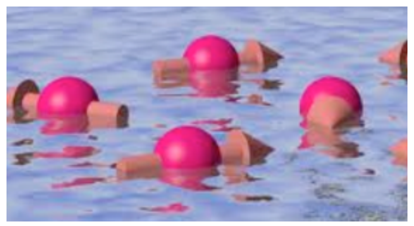
2.0 mm



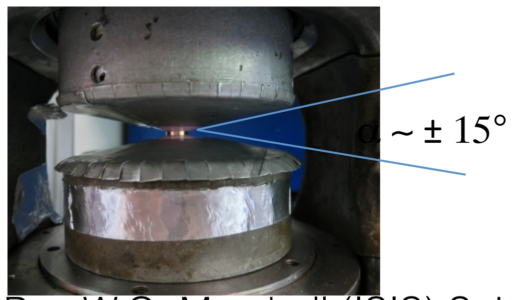


AJ Drew et al., Nature Materials 8 (2009) 310

Small single crystals: high quality, few imperfections.
High pressure synthesis:global behaviour.
Study many stochiometries
Study high absorption isotopes.
Magnetic multilayers.

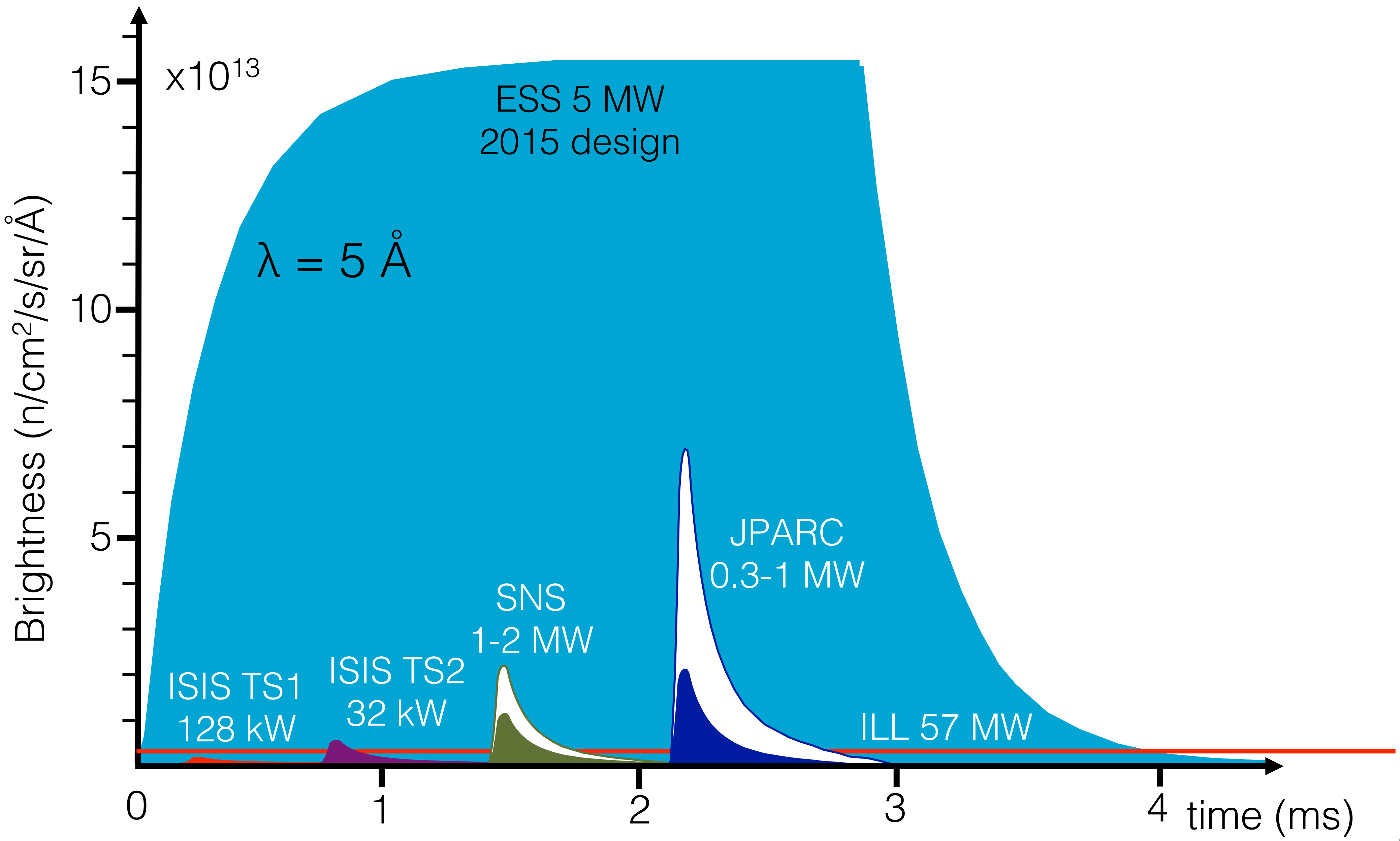


Strongly correlated physics:
High pressure, high magnetic field and low temperature simultaneously. Out of equilibrium physics

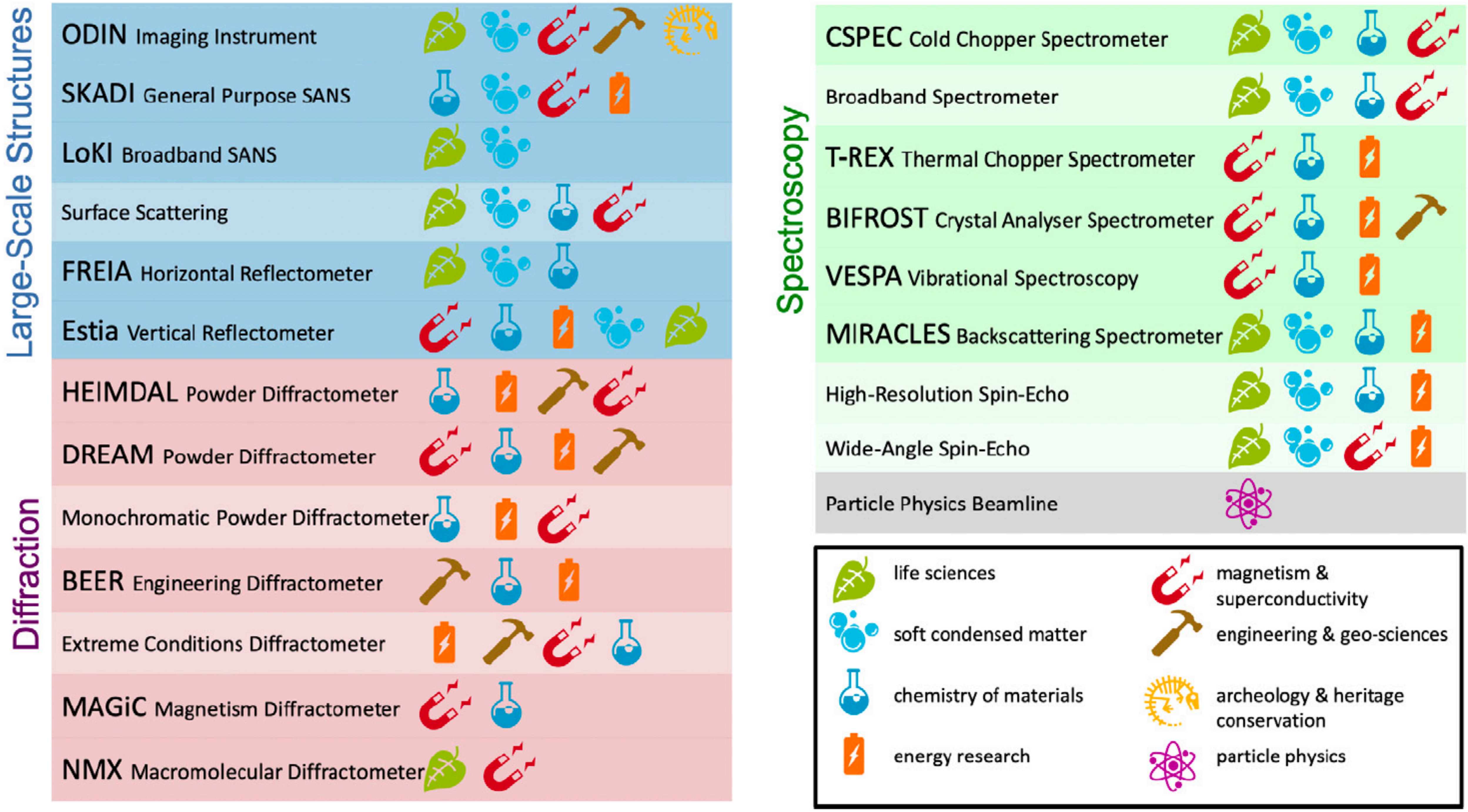


7 GPa : W.G. Marshall (ISIS) S. Klotz, unpublished R. Iizuka et al, High Press. Res. (2013)

Long pulse versus short pulse of ESS

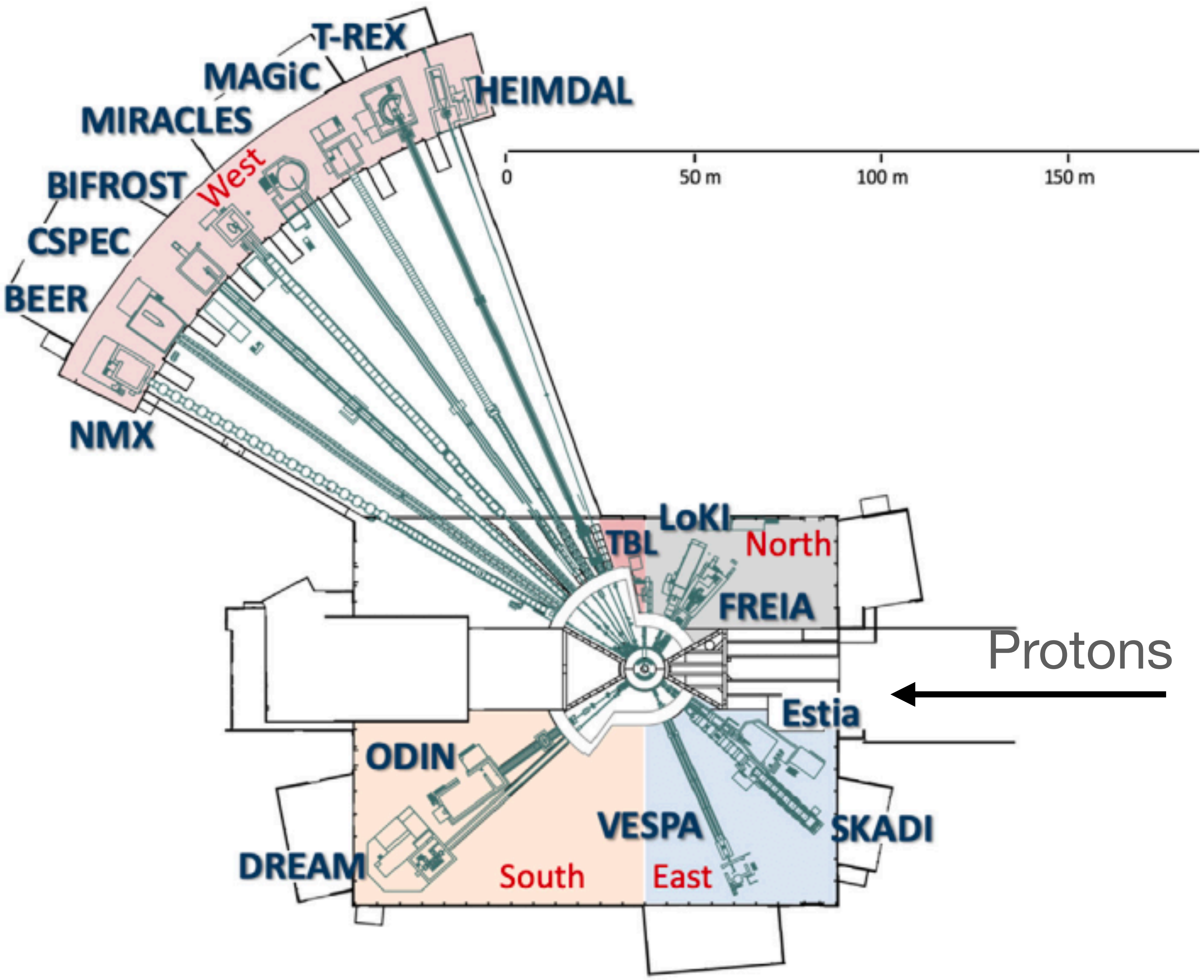


ESS Instrument suite (Phase 1) 2023-2025



ESS Instrument suite (Phase 1)
Novel magnetic states

EUROPEAN SPALLATION SOURCE

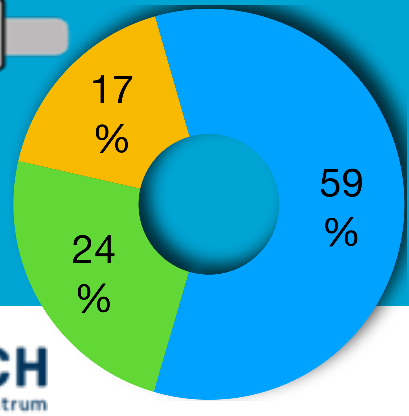


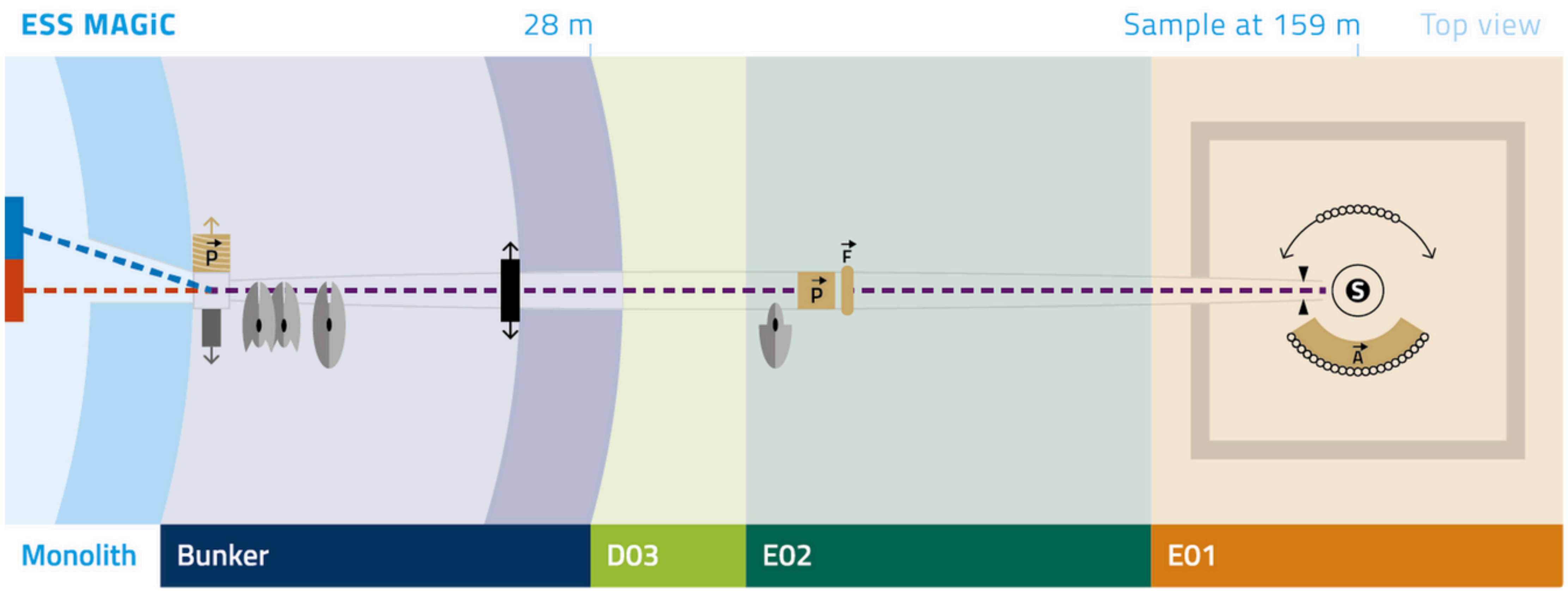
Spectroscopy

- CSPEC Cold Chopper Spectrometer
- Broadband Spectrometer
- T-REX Thermal Chopper Spectrometer
- BIFROST Crystal Analyser Spectrometer
- VESPA Vibrational Spectroscopy
- MIRACLES Backscattering Spectrometer

MAGiC Magnetism Diffractometer

MAGIC: Diffraction



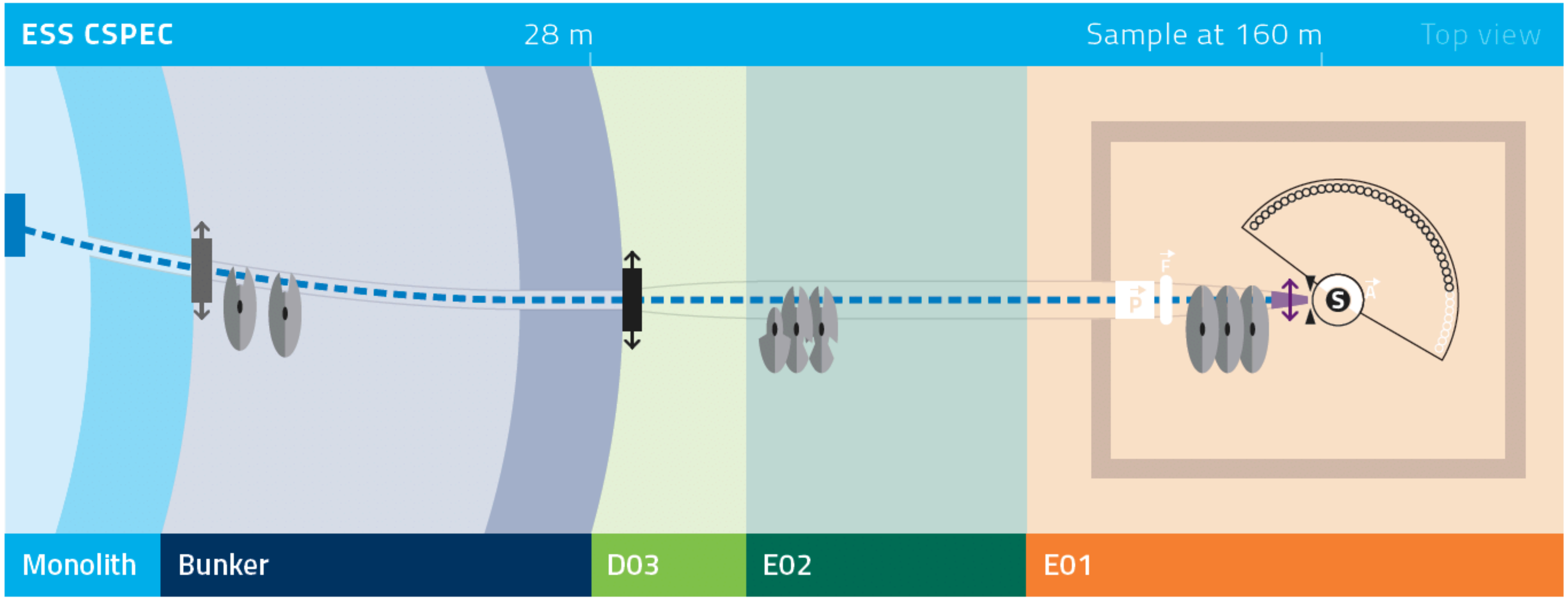


- High flux: up to $4x10^9$ n/s/cm^2
- Polarised over $0.6<\lambda<6$ Å (>97%)
- Polarisation analysis for $\lambda>2$ Å
- Flexible longitudinal and transverse resolutions
- Focusing capabilities: study of sub-mm^3 samples

CSPEC

The Cold Chopper spectrometer of the ESS

Increased flux with reduced noise

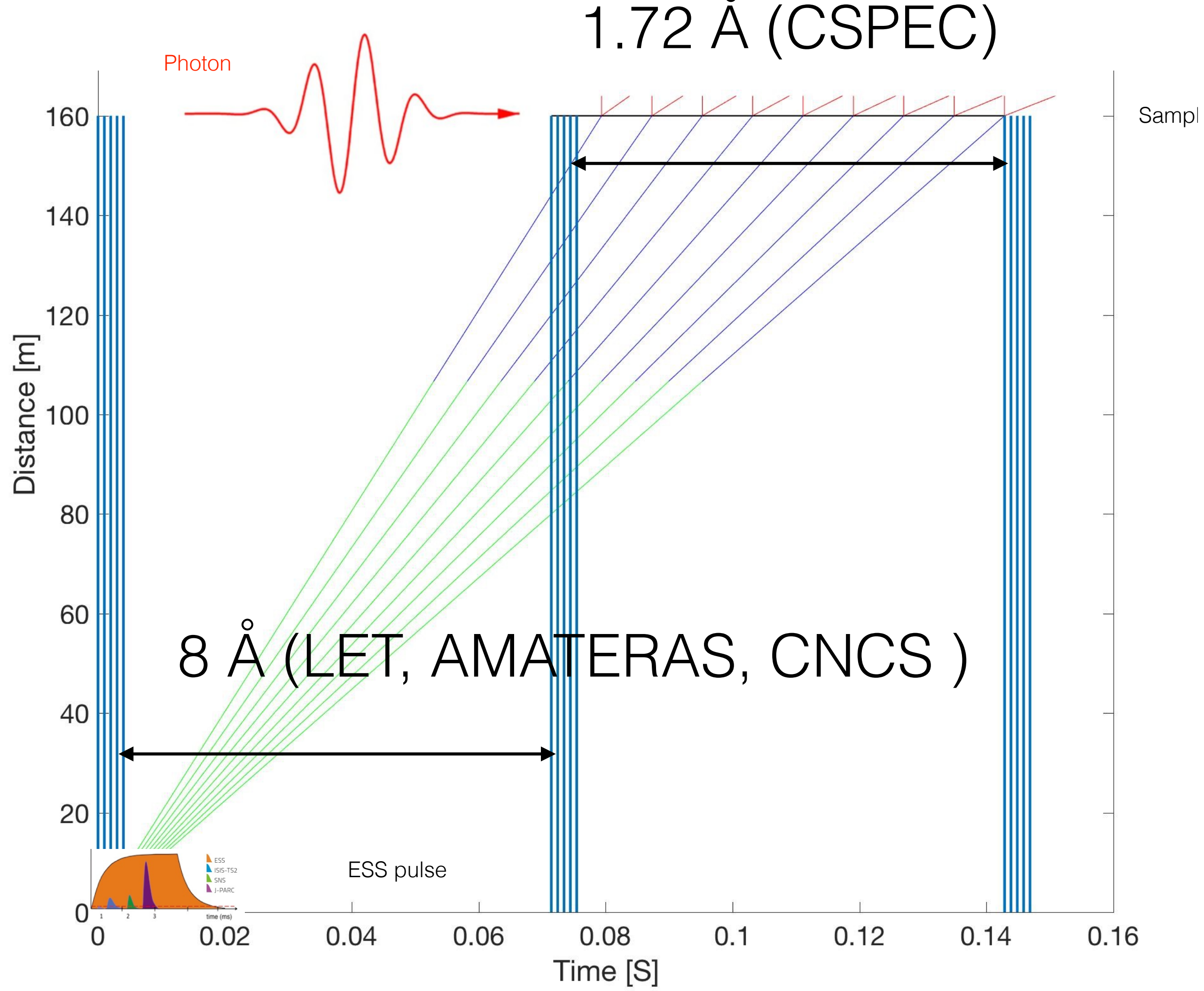


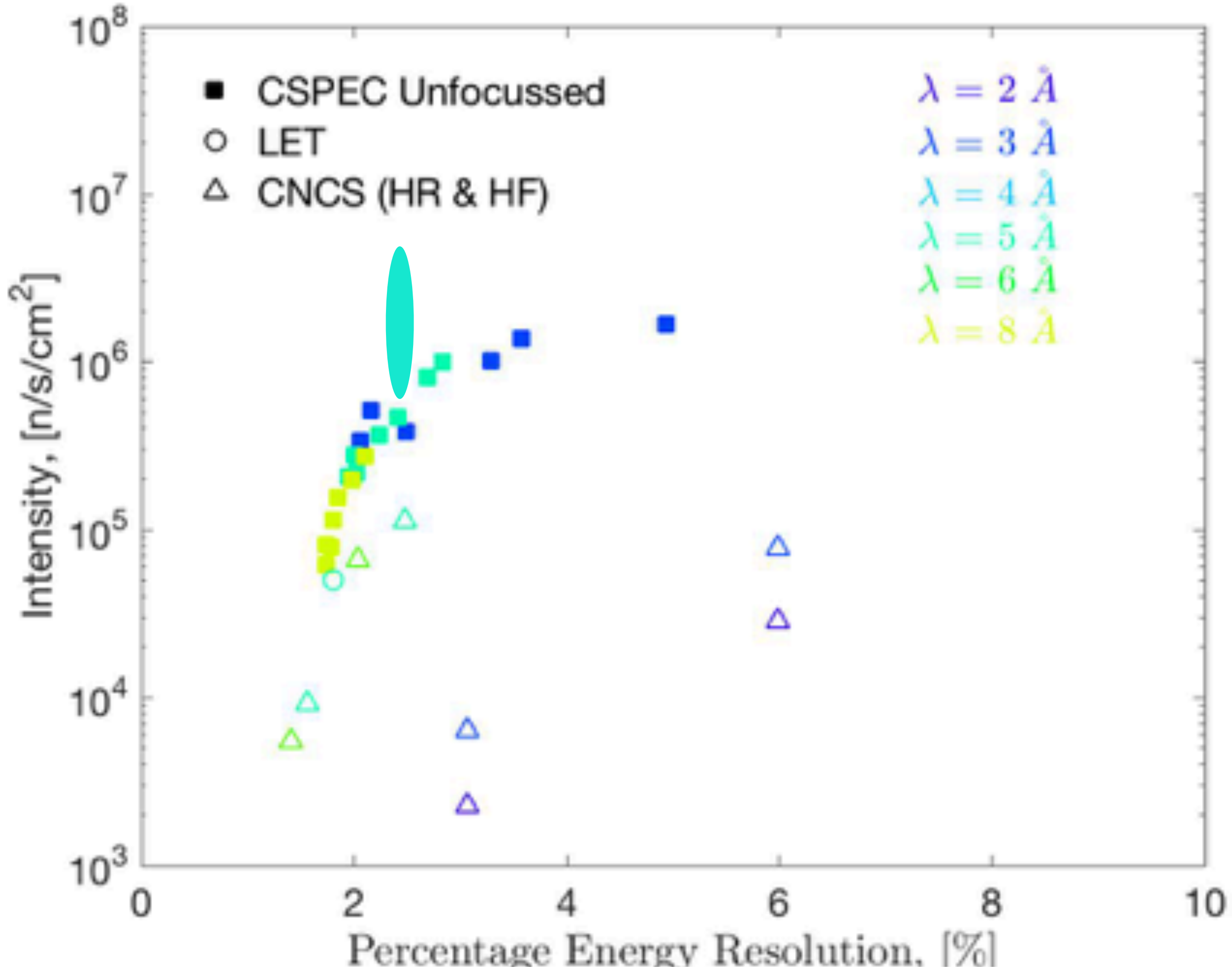
Ei = 2 - 20 Å
Instrument length = 160 m
Bandwidth = 1.72 Å
Energy resolution = 1 - 5 % of Ei
Sample size 1x 1 cm^2 & 4 x 2 cm^2
Polarisation analysis

CSPEC

The Cold Chopper spectrometer of the ESS

(1) 160 m = more flux.
In-situ/kinetic phenomena. 1 min resolution.



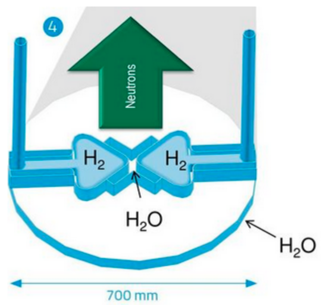


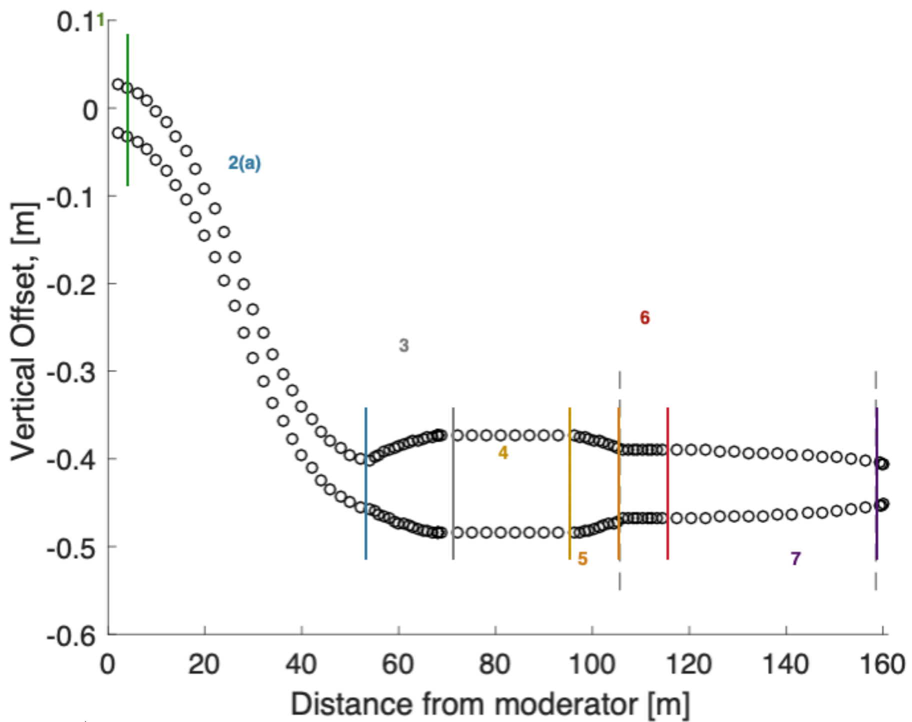
3 Å = E $_{min}$ = 5.67, E $_{max}$ = 16.9 meV
6 Å = E $_{min}$ = 1.76, E $_{max}$ = 3.02 meV
8 Å = E $_{min}$ = 1.06, E $_{max}$ = 1.58 meV - Add pulses when possible - gain in flux

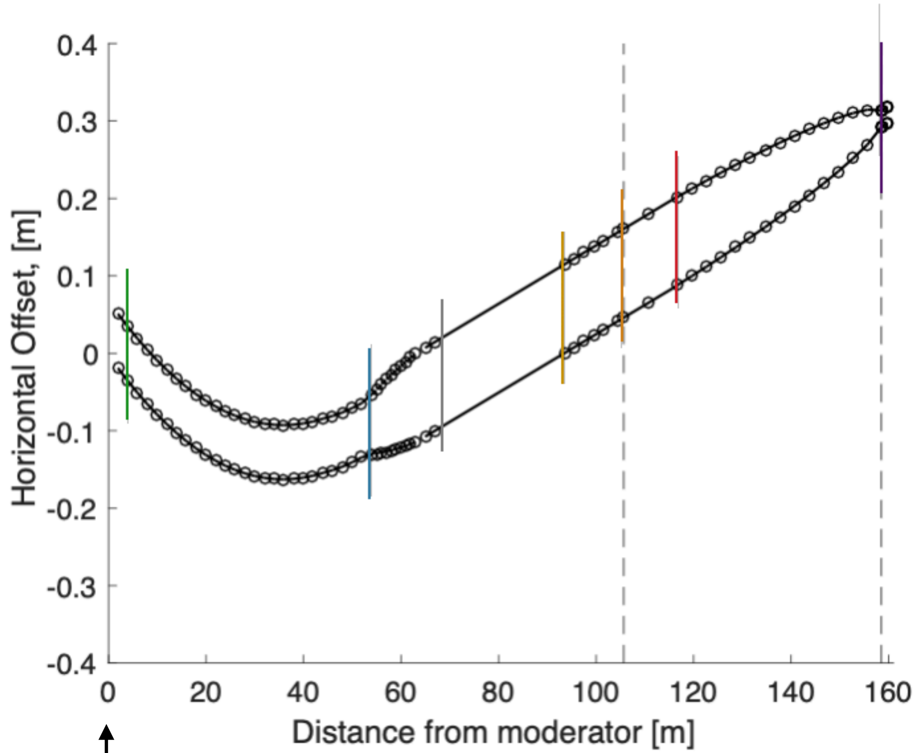
CSPEC

The Cold Chopper spectrometer of the ESS

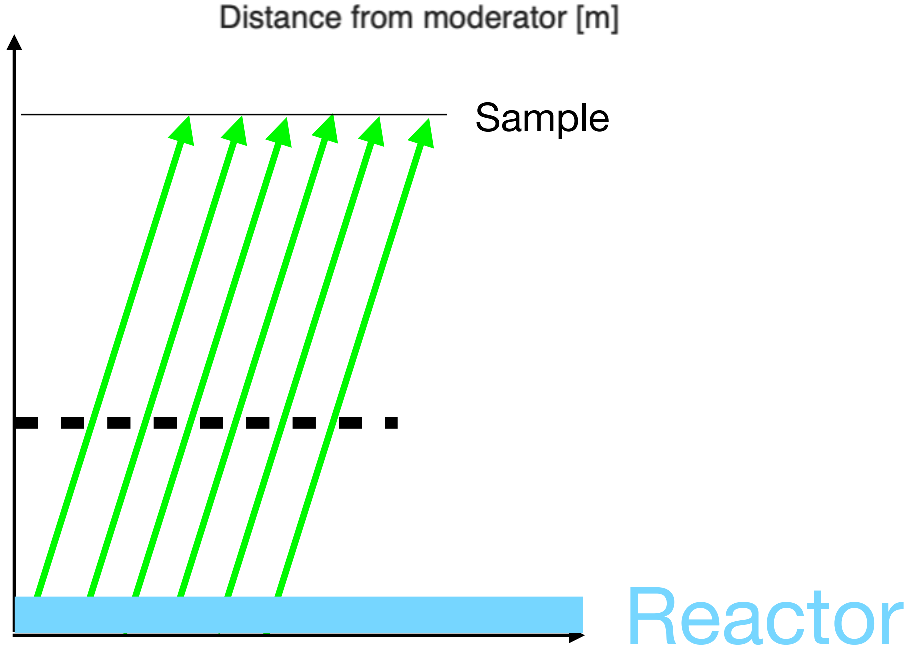
(2) 160 m & cold neutrons & spallation source = less noise. S/N 10^5.

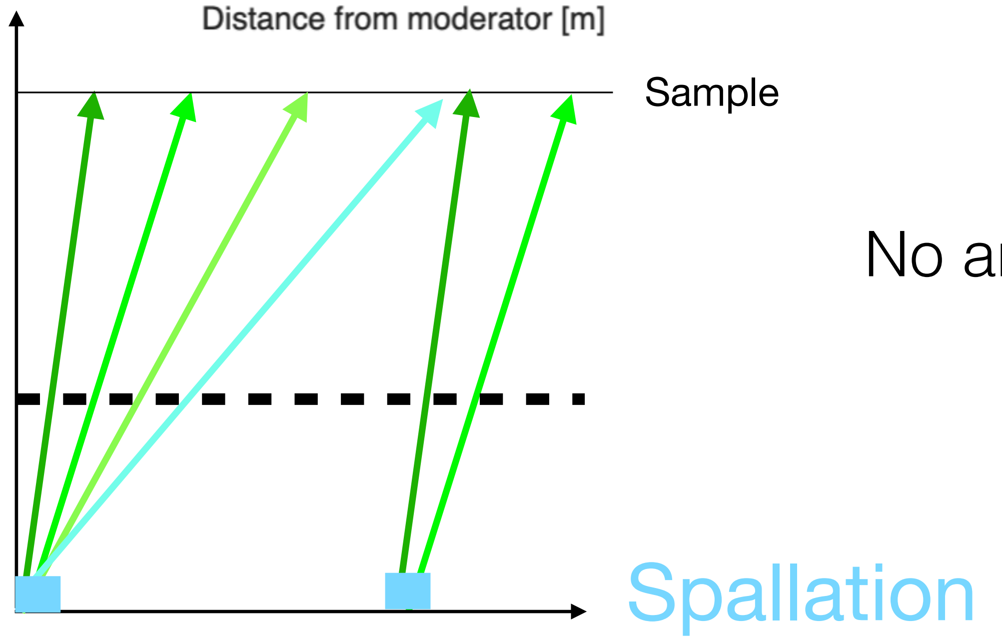






Cold neutrons: S-Bender





No ambient background

TECHNISCHE UNIVERSITÄT MÜNCHEN

rphée Laboratoire Léon Brillouin

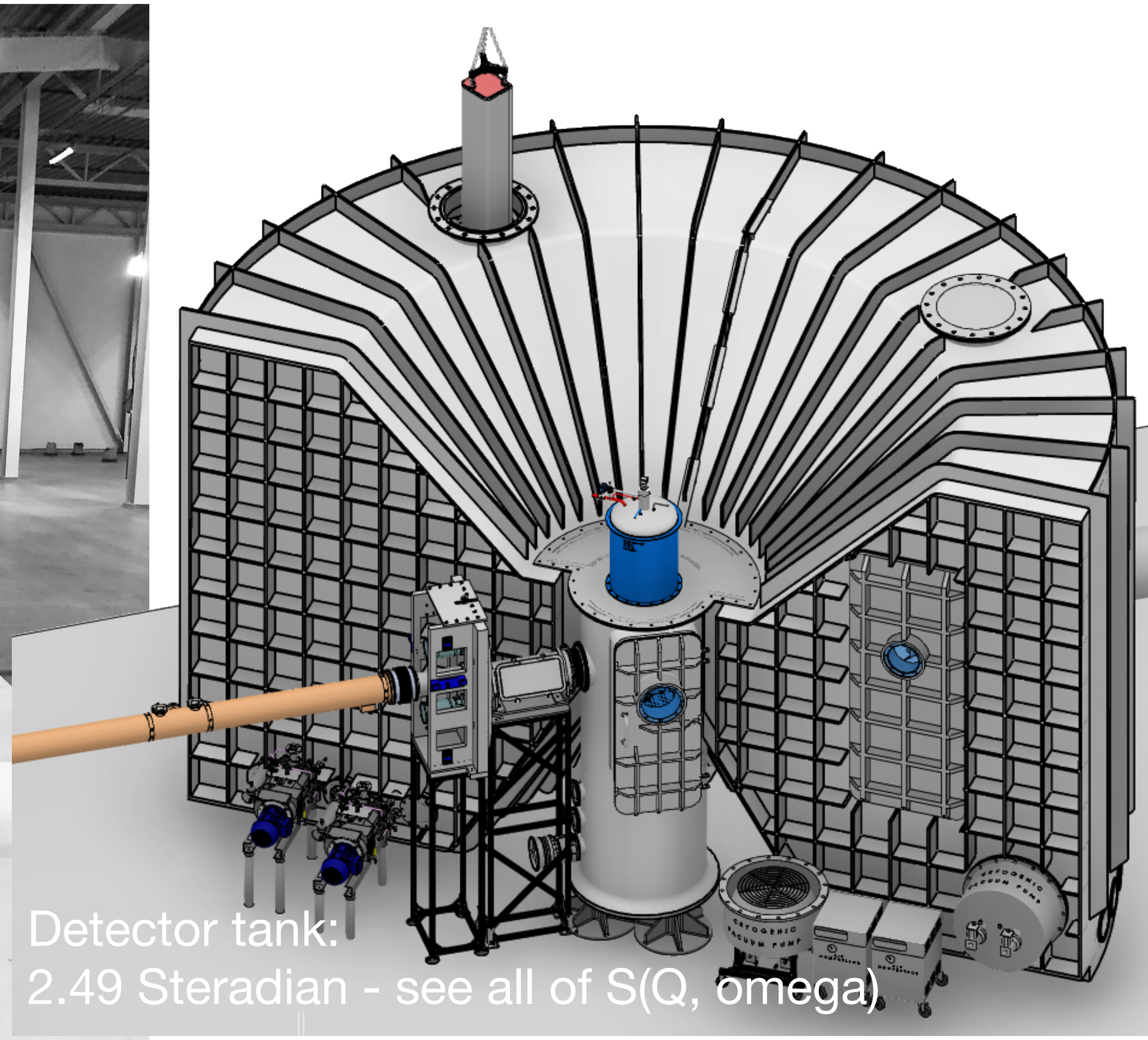
CSPEC

The Cold Chopper spectrometer of the ESS

EUROPEAN SPALLATION SOURCE

Guide production : TUM

Guide shielding

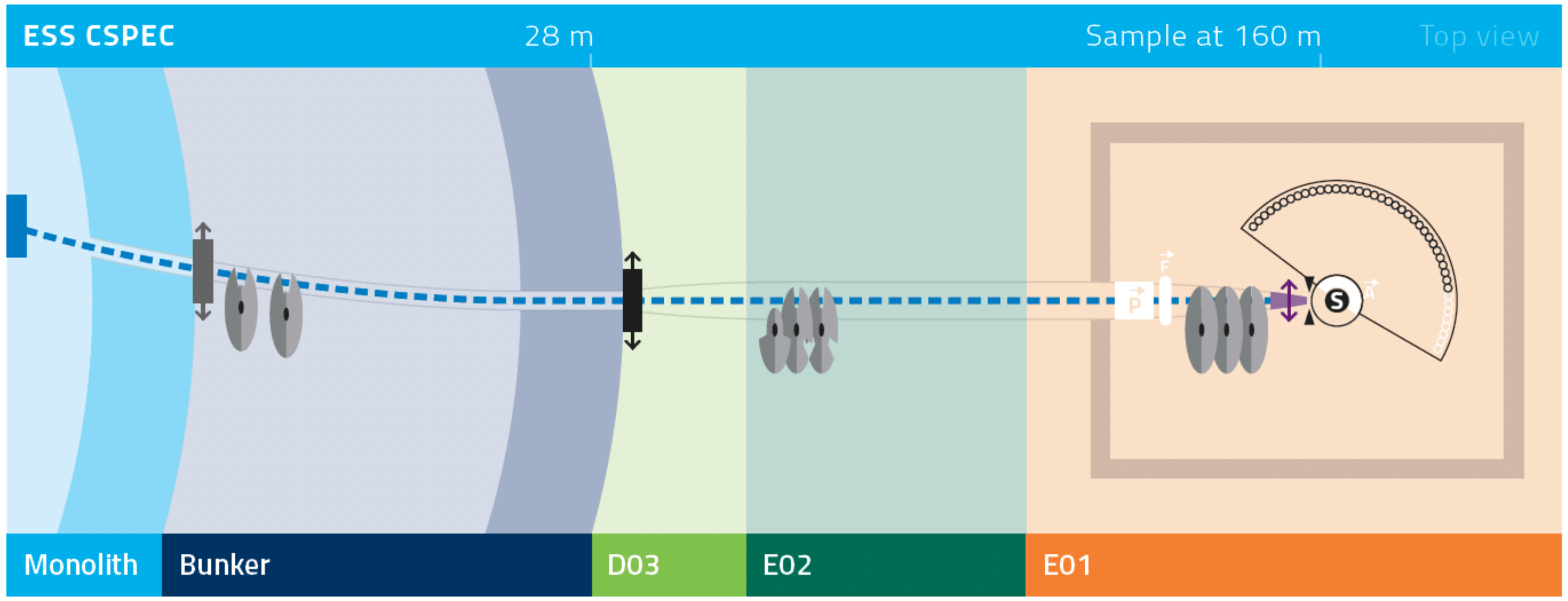


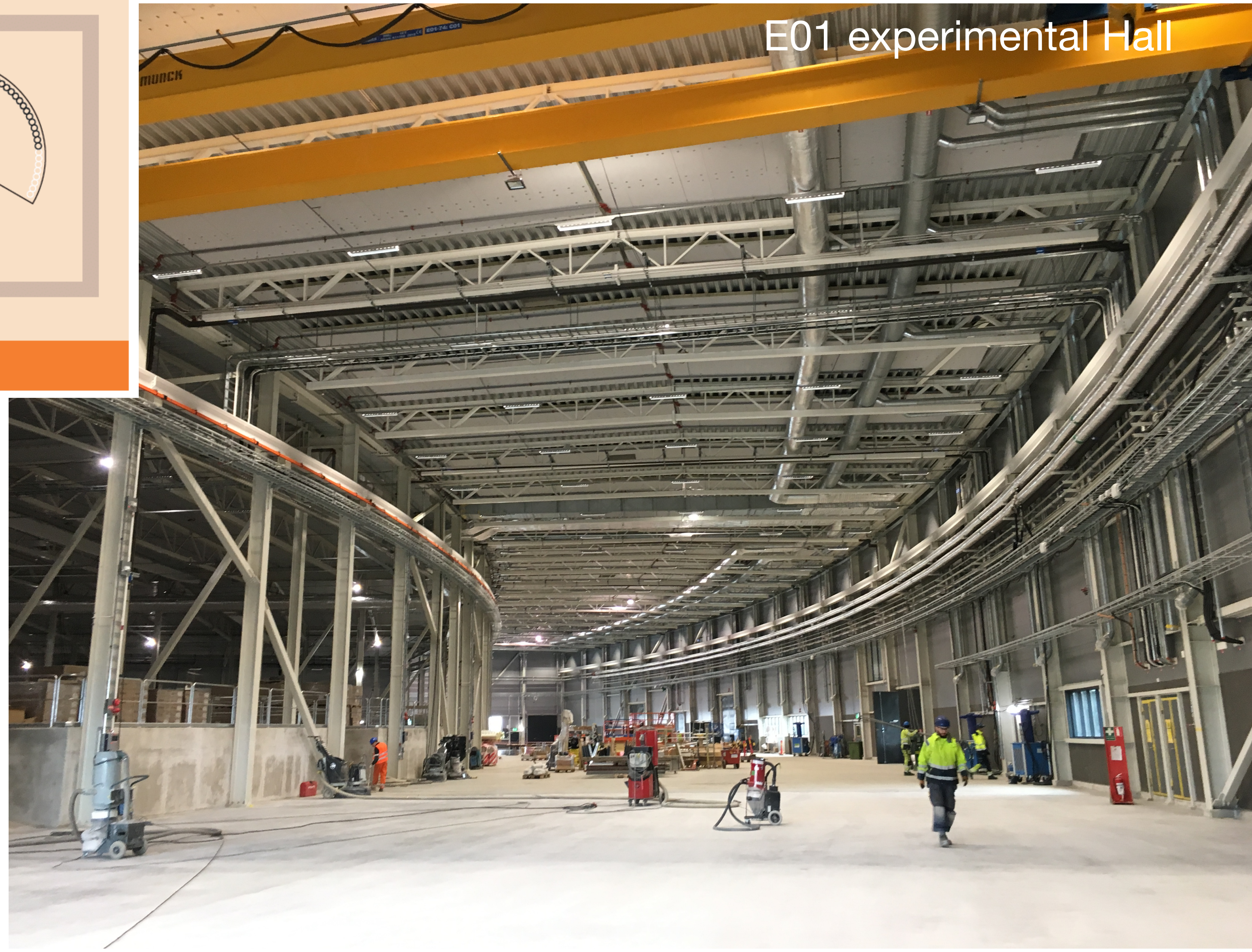
Detector tank:
2.49 Steradian - see all of S(Q, omega)

Ready December 2022.

CSPEC

The Cold Chopper spectrometer of the ESS





https://europeanspallationsource.se/instruments/cspec

EUROPEAN
SPALLATION
SOURCE

ESS December 2020

We welcome you all soon.