Neutron scattering for magnetism

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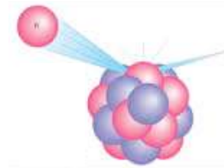
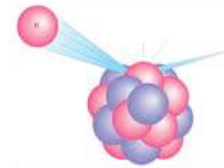
Fédération Française de Diffusion Neutronique



The neutron as a probe of condensed matter
Neutron-matter interaction processes
Diffraction by a crystal: nuclear and magnetic structures
Inelastic neutron scattering: magnetic excitations
Use of Polarized neutrons
Techniques for studying magnetic nano-objects
Complementary muon spectroscopy technique
Conclusion

The neutron as a probe of condensed matter

The neutron as a probe of condensed matter

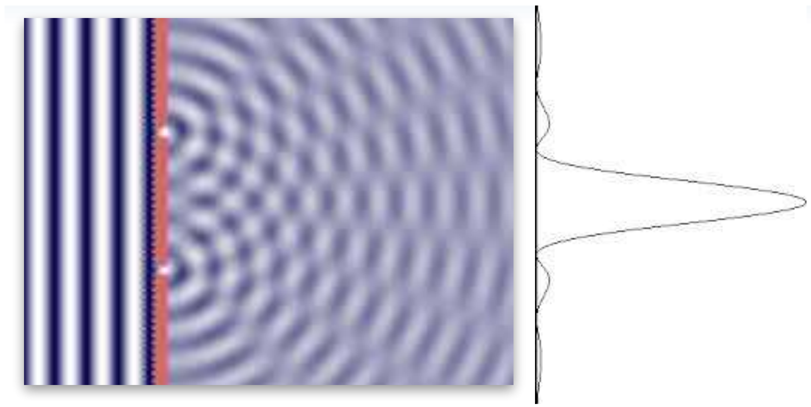


Subatomic particle discovered in 1932 by Chadwik
First neutron scattering experiment in 1946 by Shull

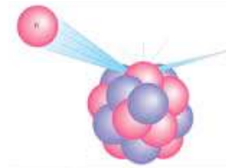
PROPERTIES:

$m_N = 1.675\ 10^{-27}$ kg, $s = 1/2$, $\tau = 888$ s

- Neutron: particle/plane wave with $E = \dfrac{\hbar^2 k^2}{2m_N}$ and $\lambda = 2\pi/k$
- Wavelength of the order of few Å (thermal neutrons) ≈ interatomic distances
  Interference → diffraction condition



The neutron as a probe of condensed matter



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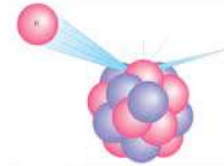
- Neutron: particle/plane wave with $E = \dfrac{\hbar^2 k^2}{2m_N}$ and $\lambda = 2\pi/k$
- Wavelength of the order of few Å ≈ interatomic distances → diffraction condition
- Energies of thermal neutrons ≈ 25 meV
  ≈ energy of excitations in condensed matter
- Neutral: probe volume, nuclear interaction with nuclei

The neutron as a probe of condensed matter



PROPERTIES:

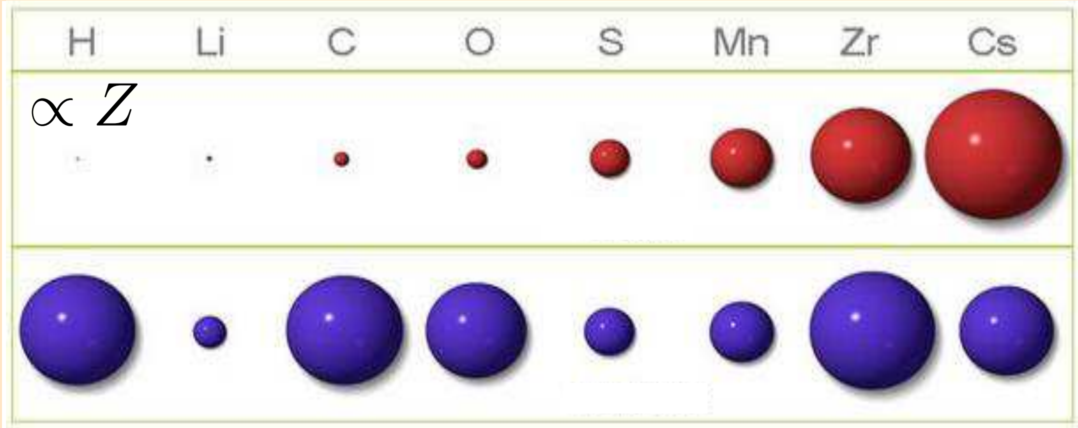
- carries a spin ½: sensitive to the magnetism of unpaired electrons (spin and orbit)
  →Probe magnetic structures and dynamics
  →Possibility to polarize the neutron beam

$$\vec{\mu}_n = -\gamma \mu_N \vec{\sigma}$$

Scattering length

X rays

neutrons



- Better than X rays for light or neighbor elements or isotopes (ex. H, D): complementary
- Neutron needs big samples!

The neutron as a probe of condensed matter

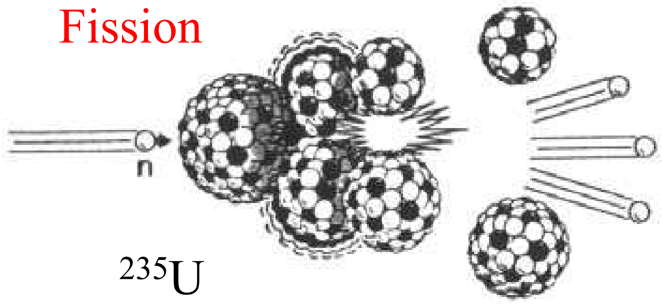
≠ TYPES OF NEUTRON SOURCES FOR RESEARCH:

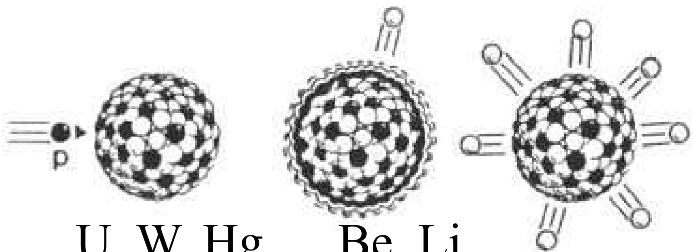
Images ILL and ESS websites

- **Neutron reactor** (continuous flux)
  ex. Institut Laue Langevin in Grenoble

- **Spallation sources** (neutron pulses)
  ex. ISIS UK or ESS future European spallation source (Lund)

- **Compact source** projects (neutron pulses)





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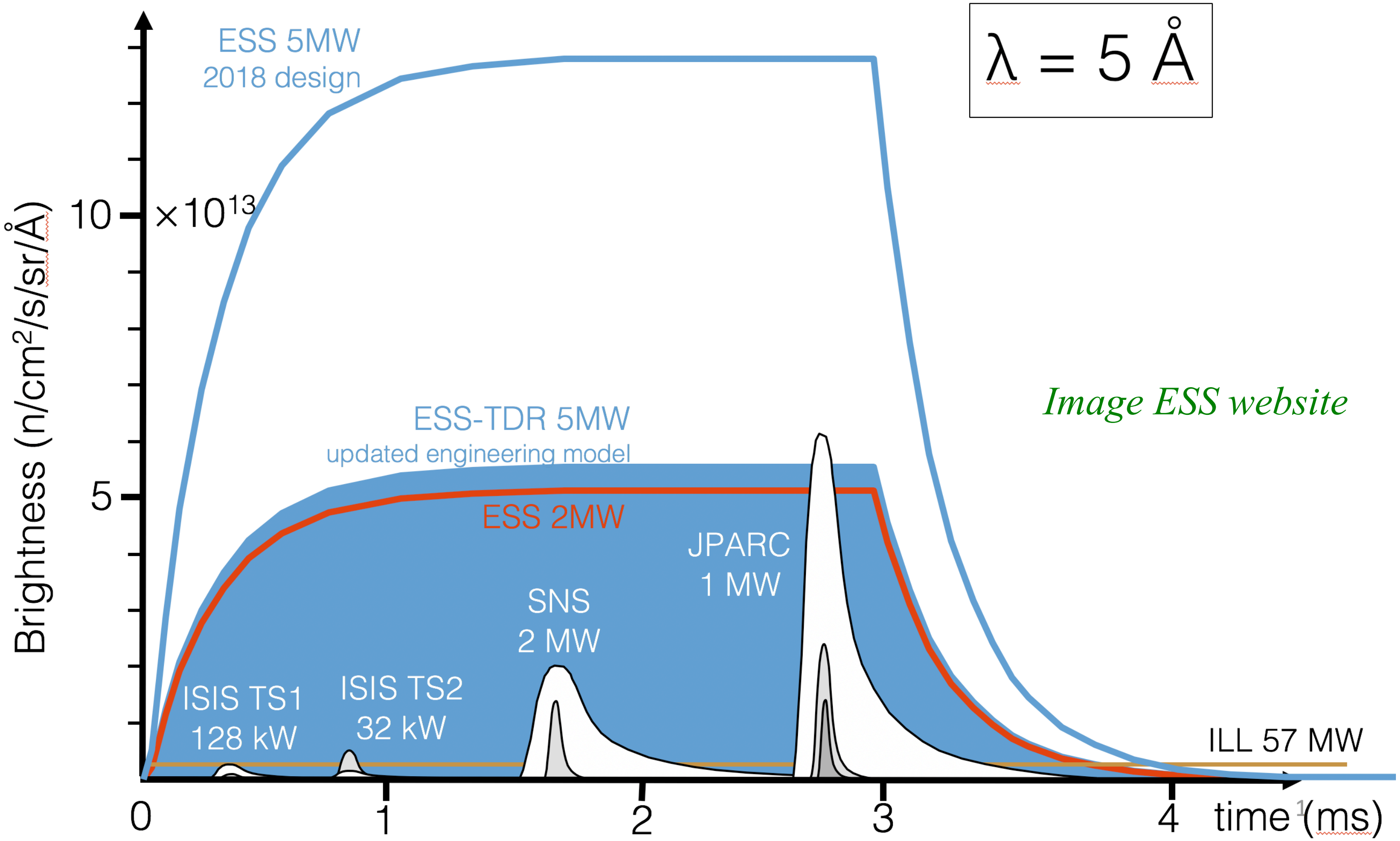
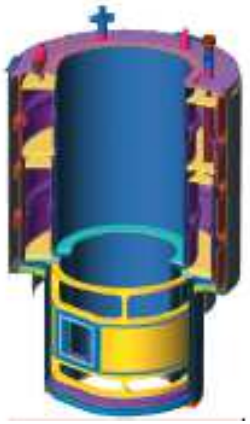


Image ESS website

The neutron as a probe of condensed matter

VARIOUS ENVIRONMENTS :

- Temperature: 30 mK-2000 K
- High magnetic steady fields up to 26 T
  Pulsed fields up to 40 T
- Pressure (gas, Paris-Edinburgh, clamp cells) up to 100 kbar
- Electric field
- CRYOPAD zero field chamber for polarization analysis



Cryopad

D23@ILL
15 T magnet

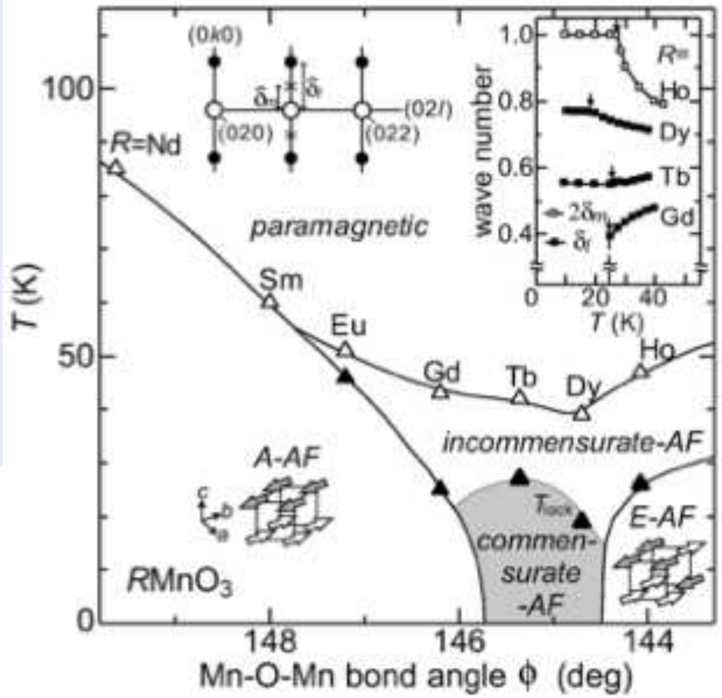
The neutron as a probe of condensed matter

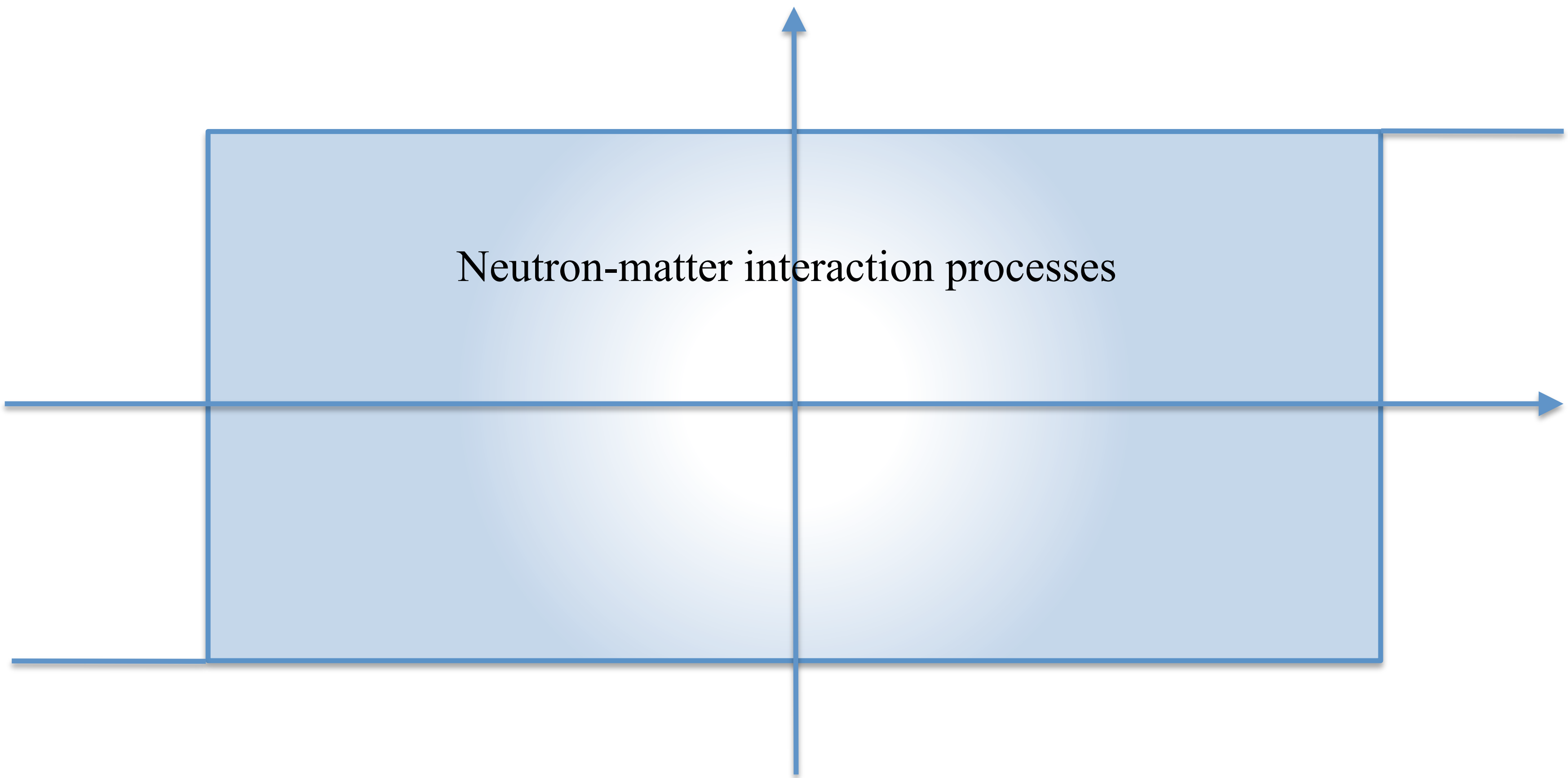
USE OF NEUTRON SCATTERING FOR MAGNETIC STUDIES:

- Most powerful tool to determine complex magnetic structures
  (non-collinear, spirals, sine waves modulated, incommensurate, skyrmion lattice)
- Complex phase diagrams (T, P, H, E) under extreme conditions
- Magnetic excitations and hybrid excitations
- In situ, in operando measurements
- Magnetic domains probe
- Short-range magnetism (ex. spin liquid/glass/ice)
- Magnetic nano-structures/mesoscopic magnetism
- Chirality determination
- Materials with hydrogen

Example: orthorhombic $RMnO_3$

Goto et al. PRL (2005)

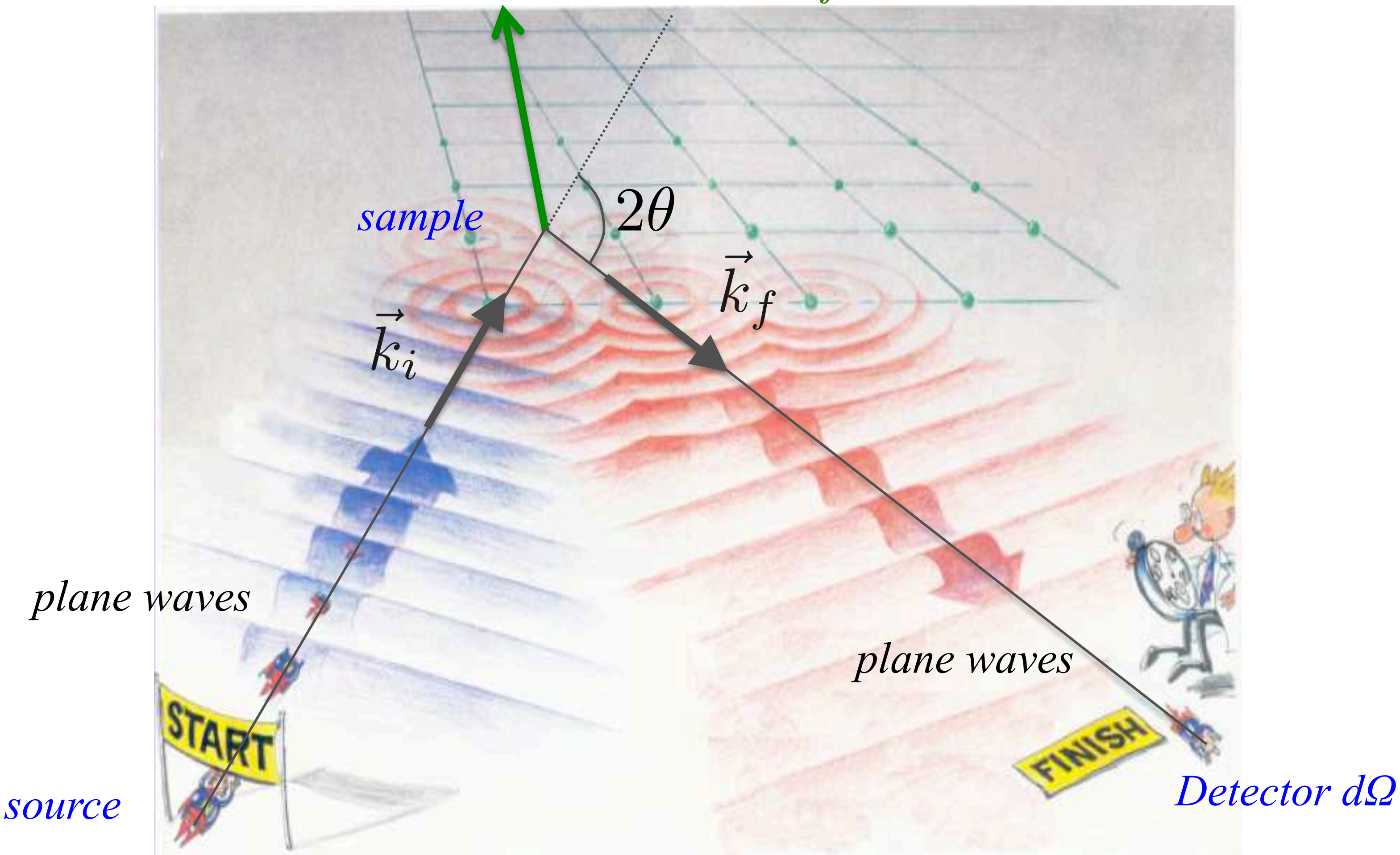



Neutron-matter interaction processes

Neutron-matter interaction processes

SCATTERING PROCESS: INTERFERENCE PHENOMENA

Born approximation $\vec{Q} = \vec{k}_i - \vec{k}_f$ Momentum transfer=scattering vector



Neutron-matter interaction processes

SCATTERING PROCESS: INTERFERENCE PHENOMENA

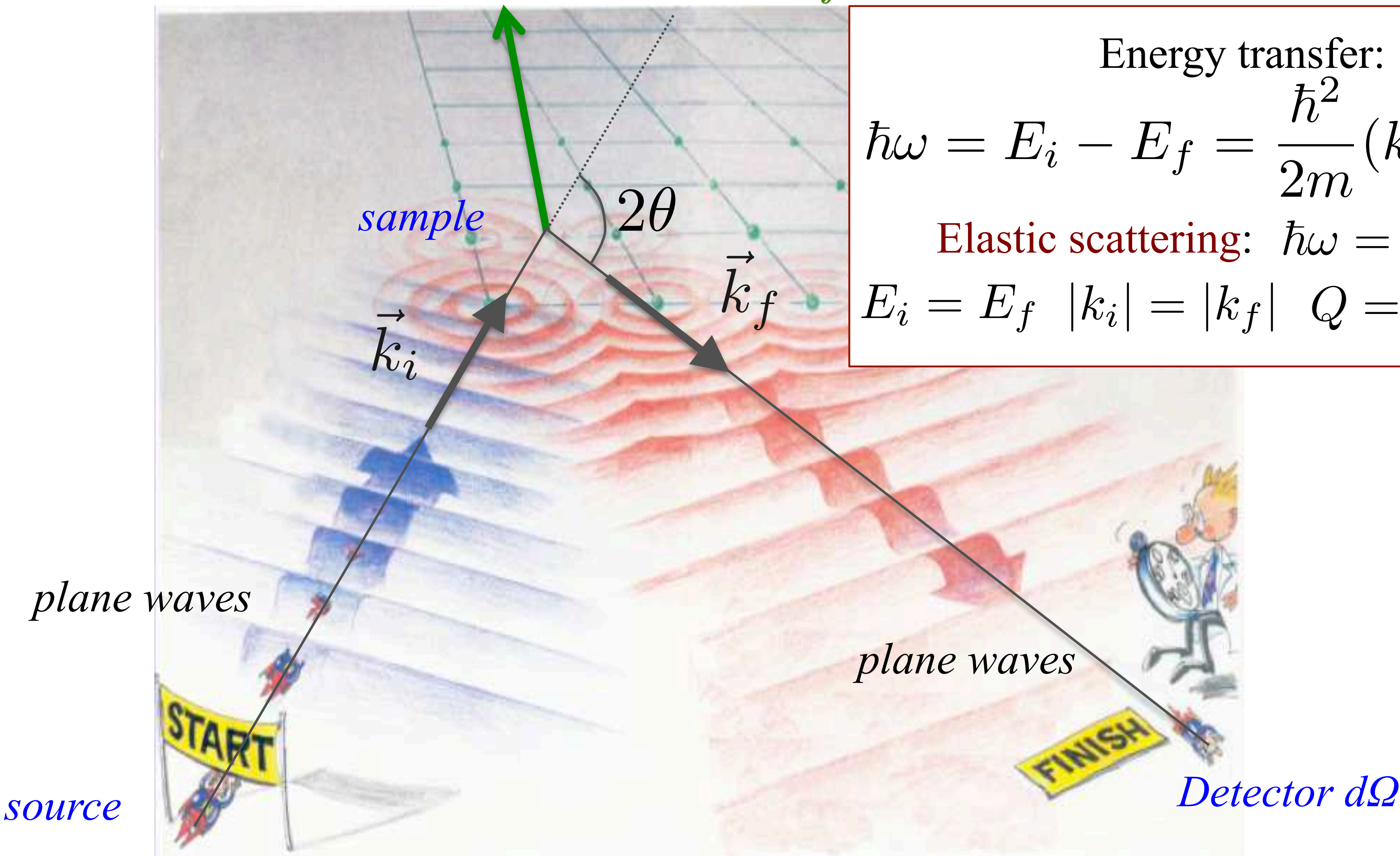
$\vec{Q} = \vec{k}_i - \vec{k}_f$ Scattering vector

Energy transfer:

$$\hbar\omega = E_i - E_f = \frac{\hbar^2}{2m}(k_i^2 - k_f^2)$$

Elastic scattering: $\hbar\omega = 0$

$$E_i = E_f \quad |k_i| = |k_f| \quad Q = 2\sin\theta/\lambda$$



Neutron-matter interaction processes

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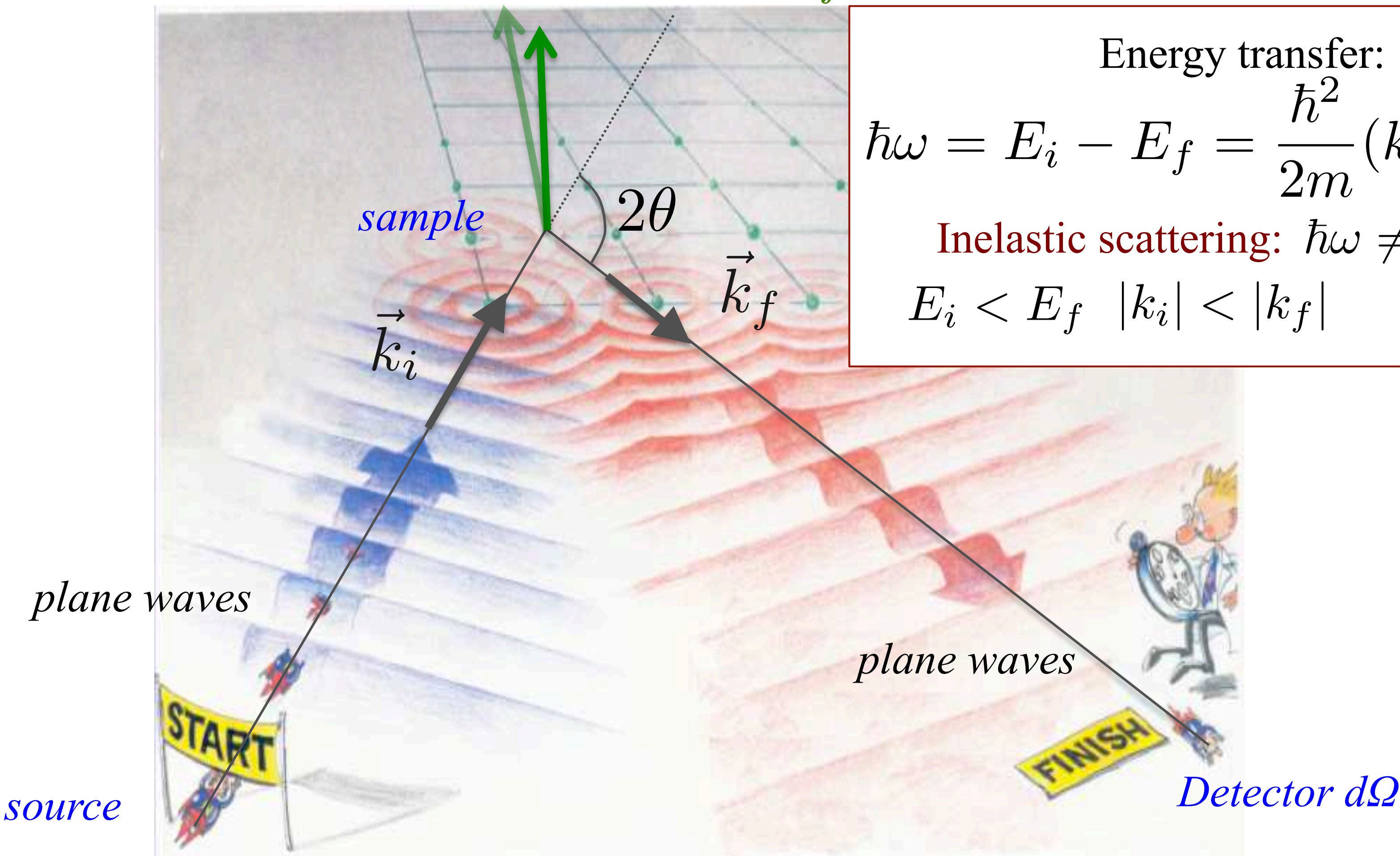


Energy transfer:

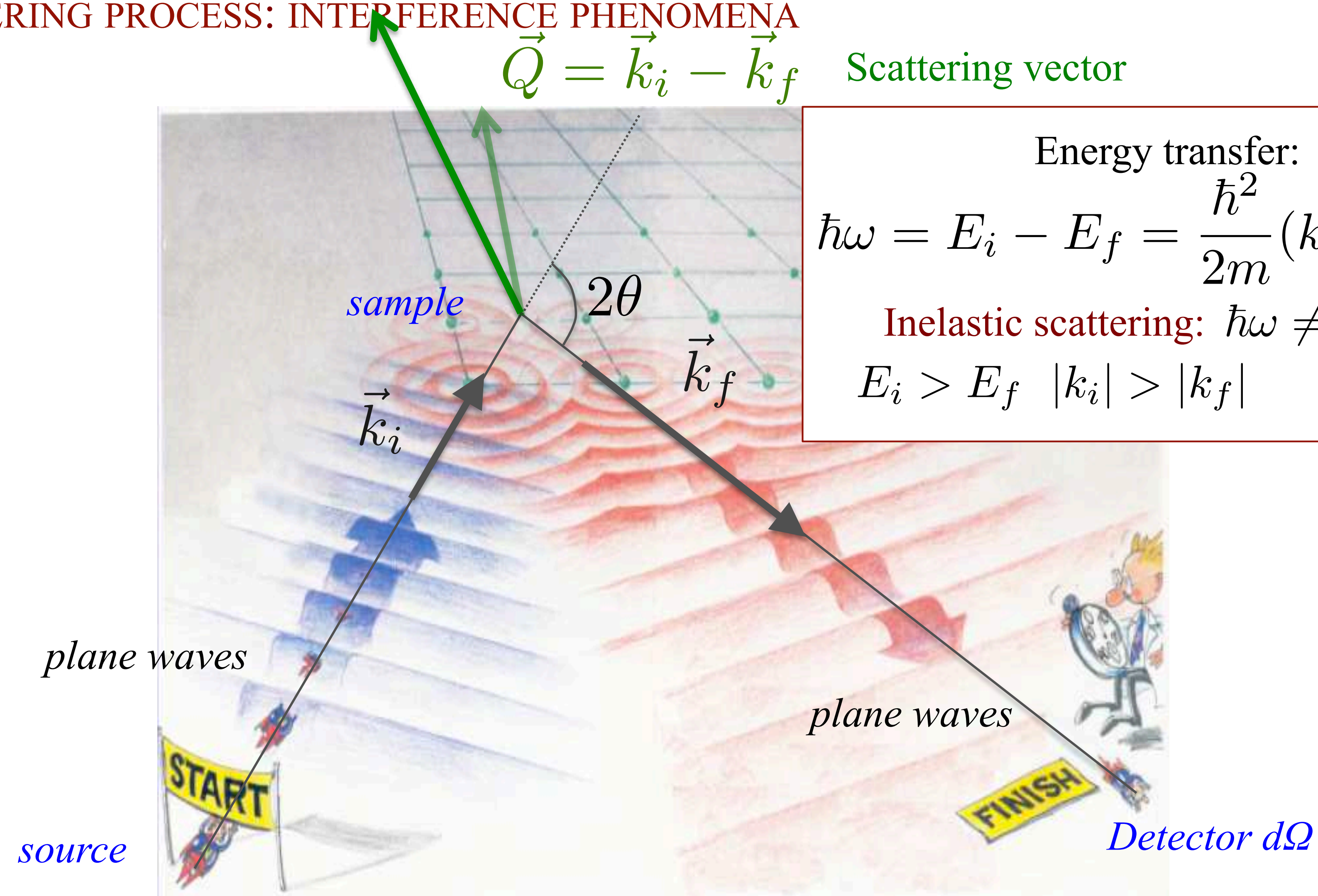
$$\hbar\omega = E_i - E_f = \frac{\hbar^2}{2m}(k_i^2 - k_f^2)$$

Inelastic scattering: $\hbar\omega \neq 0$

$E_i < E_f \quad |k_i| < |k_f|$

Neutron-matter interaction processes

SCATTERING PROCESS: INTERFERENCE PHENOMENA



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Inelastic scattering: $\hbar\omega \neq 0$

$E_i > E_f \quad |k_i| > |k_f|$

SCATTERING PROCESS: INTERFERENCE PHENOMENA

The cross-sections (in barns 10^{-24} cm^2) = quantities measured during a scattering experiment:

Total cross-section σ : number of neutrons scattered per second /flux of incident neutrons

Differential cross section $\frac{d\sigma}{d\Omega}$: per solid angle element

Partial differential cross section $\frac{d^2\sigma}{d\Omega dE}$: per energy element

Neutron-matter interaction processes

FERMI'S GOLDEN RULE

Partial differential cross section

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}(\frac{m_N}{2\pi\hbar^2})^2 \sum_{\lambda,\sigma_i} \sum_{\lambda',\sigma_f} p_\lambda p_{\sigma_i} |\langle k_f \sigma_f \lambda_f | V | k_i \sigma_i \lambda_i \rangle|^2 \delta(\hbar\omega + E - E')$$

Energy conservation

Initial and final wave vector and spin state of the neutrons

Initial and final state of the sample

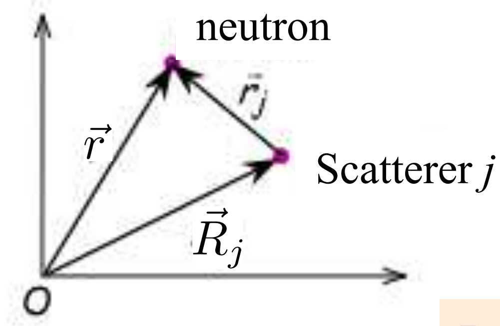
Interaction potential = Sum of nuclear and magnetic scattering

Neutron-matter interaction processes

Interaction potential = Sum of nuclear and magnetic scattering

<u>Nuclear interaction potential</u>

- very short range
- isotropic



<u>Magnetic interaction potential</u>

- Longer range (e$^-$ cloud)
- Anisotropic

$$V(\vec{r}) = \left(\frac{2\pi\hbar^2}{m_N}\right)\sum_i b_i \delta(\vec{r} - \vec{R}_i)$$

b Scattering length
depends on isotope and nuclear spin

Dipolar interaction of the neutron magnetic moments μ_n with magnetic field from unpaired e$^-$

$$V(\vec{r}) = -\vec{\mu}_n . \vec{B}(\vec{r})$$

$$\vec{B}(\vec{r}) = \frac{\mu_0}{4\pi}\sum_i \left[\mathrm{rot}\left(\frac{\vec{\mu}_{ei} \times (\vec{r} - \vec{R}_i)}{|\vec{r} - \vec{R}_i|^3}\right) - \frac{2\mu_B}{\hbar}\frac{\vec{p}_i \times (\vec{r} - \vec{R}_i)}{|\vec{r} - \vec{R}_i|^3}\right]$$

Spin contribution *Orbital contribution*

Neutron-matter interaction processes

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}(\frac{m_N}{2\pi\hbar^2})^2 \sum_{\lambda,\sigma_i}\sum_{\lambda',\sigma_f} p_\lambda p_{\sigma_i} |\langle k_f\sigma_f\lambda_f|V|k_i\sigma_i\lambda_i\rangle|^2 \delta(\hbar\omega + E - E')$$

Some algebra (hyp. no spin polarization)

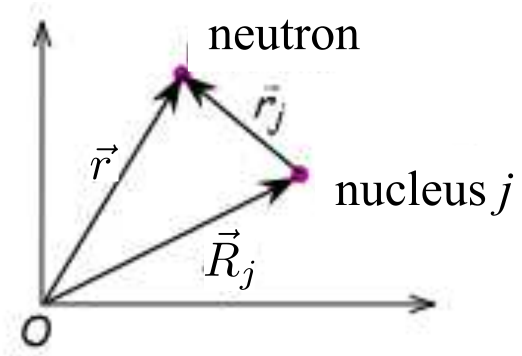
$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'}\int_{-\infty}^{+\infty}\langle A_j^*(0)A_{j'}(t)e^{-i\vec{Q}\vec{R}_{j'}(0)}e^{i\vec{Q}\vec{R}_j(t)}\rangle e^{-i\omega t}dt$$

with $A_j(t)$ the scattering amplitude

Scattering experiment
→FT of interaction potential

Neutron-matter interaction processes

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'}\int_{-\infty}^{+\infty}\langle A^*_{j'}(0)A_j(t)e^{-i\vec{Q}\vec{R}_{j'}(0)}e^{i\vec{Q}\vec{R}_j(t)}\rangle e^{-i\omega t}dt$$



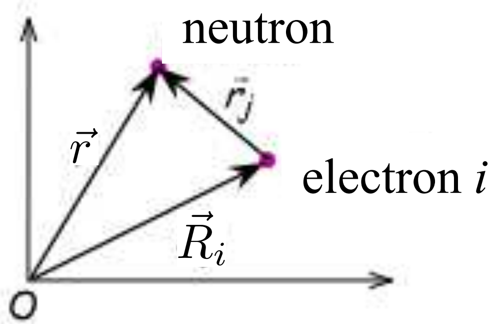
nuclear

b_j

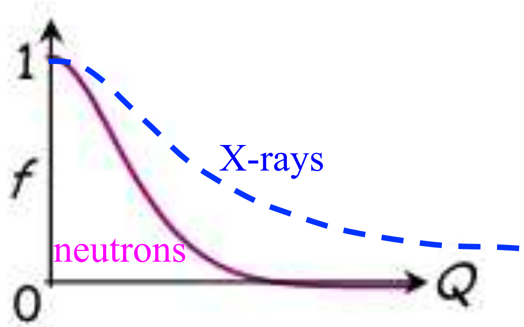
magnetic

$p f_j(Q)\vec{M}_{j\perp}(\vec{Q},t)$

p= 0.2696x10^{-12} cm



Magnetic form factor of the free ion

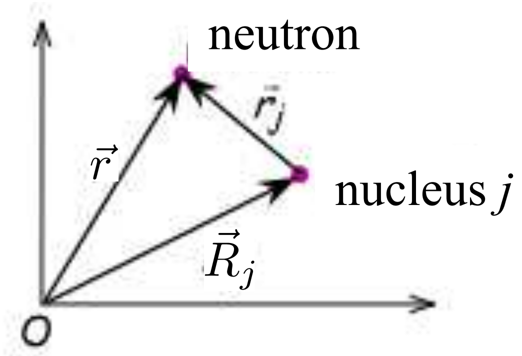


Neutron-matter interaction processes

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'}\int_{-\infty}^{+\infty}\langle A_{j'}^{*}(0)A_j(t)e^{-i\vec{Q}\vec{R}_{j'}(0)}e^{i\vec{Q}\vec{R}_j(t)}\rangle e^{-i\omega t}dt$$

nuclear

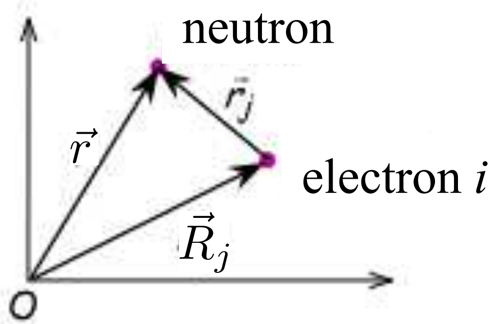
magnetic

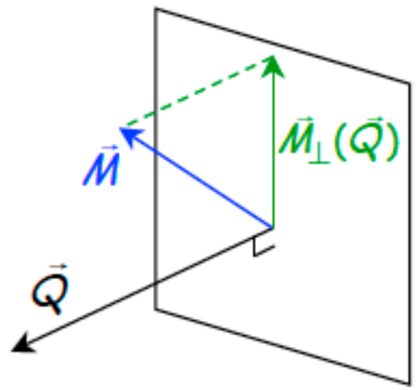


$$b_j$$

$$pf_j(Q)\vec{M}_{j\perp}(\vec{Q},t)$$

Projection of the magnetic moment $\perp \vec{Q}$





Neutron-matter interaction processes

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'}\int_{-\infty}^{+\infty}\langle A^*_{j'}(0)A_j(t)e^{-i\vec{Q}\vec{R}_{j'}(0)}e^{i\vec{Q}\vec{R}_j(t)}\rangle e^{-i\omega t}dt$$

nuclear | magnetic

b_j | $p f_j(Q)\vec{M}_{j\perp}(\vec{Q},t)$

= Double FT in space and time of
the pair correlation function of the
nuclear density | magnetic density $\perp \vec{Q}$

Neutron-matter interaction processes

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'}\int_{-\infty}^{+\infty}\langle A^*_{j'}(0)A_j(t)e^{-i\vec{Q}\vec{R}_{j'}(0)}e^{i\vec{Q}\vec{R}_j(t)}\rangle e^{-i\omega t}dt$$

<u>Separation elastic/inelastic:</u>
Keeps only the time-independent terms
in the cross-section and integrate over energy
→ elastic scattering (resulting from static order)

$$\frac{d\sigma}{d\Omega} = \sum_{jj'}\langle A^*_{j'}A_j e^{-i\vec{Q}(\vec{R}_{j'}-\vec{R}_j)}\rangle$$

Diffraction by a crystal: nuclear and magnetic structures

Diffraction by a crystal: nuclear and magnetic structures

NUCLEAR DIFFRACTION

Crystal = lattice + basis

crystal lattice $\vec{R}_n = u_n\vec{a} + v_n\vec{b} + w_n\vec{c}$

reciprocal lattice $\vec{H} = h\vec{a}^* + k\vec{b}^* + l\vec{c}^*$

$$\frac{d\sigma}{d\Omega} = \sum_{j,j'} < b_j b_{j'} e^{-i\vec{Q}(\vec{R}_{j'} - \vec{R}_j)} >$$

$$\frac{d\sigma}{d\Omega} = \frac{(2\pi)^3}{V} \sum_{\vec{H}} |F_N(\vec{Q})|^2 \delta(\vec{Q} - \vec{H})$$

diffraction condition (lattice)

Coherent elastic scattering from crystal
→Bragg peaks at nodes of reciprocal lattice

Diffraction by a crystal: nuclear and magnetic structures

NUCLEAR DIFFRACTION

$$\frac{d\sigma}{d\Omega} = \frac{(2\pi)^3}{V} \sum_{\vec{H}} |F_N(\vec{Q})|^2 \delta(\vec{Q} - \vec{H})$$

Nuclear structure factor

for a Bravais lattice:
1 atom/unit cell

$$F_N(\vec{Q}) = b$$

For a non Bravais lattice:
ν atoms/unit cell

$$\vec{R}_{n\nu} = \vec{R}_n + \vec{r}_\nu \quad \text{with}$$

$$\vec{r}_\nu = x_\nu \vec{a} + y_\nu \vec{b} + z_\nu \vec{c}$$

$$F_N(\vec{Q}) = \sum_\nu b_\nu e^{i\vec{Q}.\vec{r}_\nu}$$

Information on atomic
arrangement inside unit cell

MAGNETIC DIFFRACTION

Magnetic ordering may not have same periodicity as nuclear one
→ **propagation vector** $\vec{\tau}$ → periodicity and propagation direction

Moment distribution is a periodic function of space → can be Fourier expanded:

$$\vec{\mu}_{n,\nu} = \sum_{\vec{\tau}} \vec{m}_{\nu,\vec{\tau}} e^{-i\vec{\tau}.\vec{R}_n}$$

Magnetic moment of atom *ν* in n^th unit cell

Fourier component associated to $\vec{\tau}$

Example: For a unique propagation vector $\vec{\tau} = (1/2, 0, 0) = \vec{a^*}/2$

$$\vec{\mu}_{n,\nu} = \vec{m}_{\nu} e^{-i2\pi n/2}$$

Staggered magnetic moments = doubling of the nuclear cell

Diffraction by a crystal: nuclear and magnetic structures

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Magnetic moment of atom *ν* in n^th^ unit cell

Fourier component associated to $\vec{\tau}$

Magnetic periodicity = x times nuclear periodicity → $\vec{\tau} = (1/x, 0, 0)$

Diffraction by a crystal: nuclear and magnetic structures

MAGNETIC DIFFRACTION

Magnetic ordering may not have same periodicity as nuclear one
→ **propagation vector** $\vec{\tau}$ → periodicity and propagation direction

For a non-Bravais lattice

$$\frac{d\sigma}{d\Omega} = \frac{(2\pi)^3}{V} \sum_{\vec{H}} \sum_{\vec{\tau}} |\vec{F}_{M\perp}(\vec{Q})|^2 \delta(\vec{Q} - \vec{H} - \vec{\tau})$$

diffraction condition

→ Bragg peaks at satellites positions $\vec{Q} = \vec{H} \pm \vec{\tau}$

MAGNETIC DIFFRACTION

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Magnetic structure factor: information on magnetic arrangement in unit cell

$$\vec{F}_M(\vec{Q} = \vec{H} + \vec{\tau}) = p \sum f_\nu(\vec{Q}) \vec{m}_{\nu,\vec{\tau}} e^{i\vec{Q}.\vec{r}_\nu}$$

Fourier component

Diffraction by a crystal: nuclear and magnetic structures

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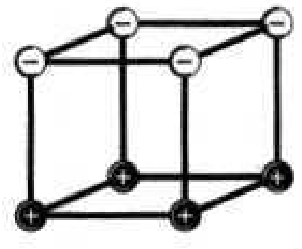


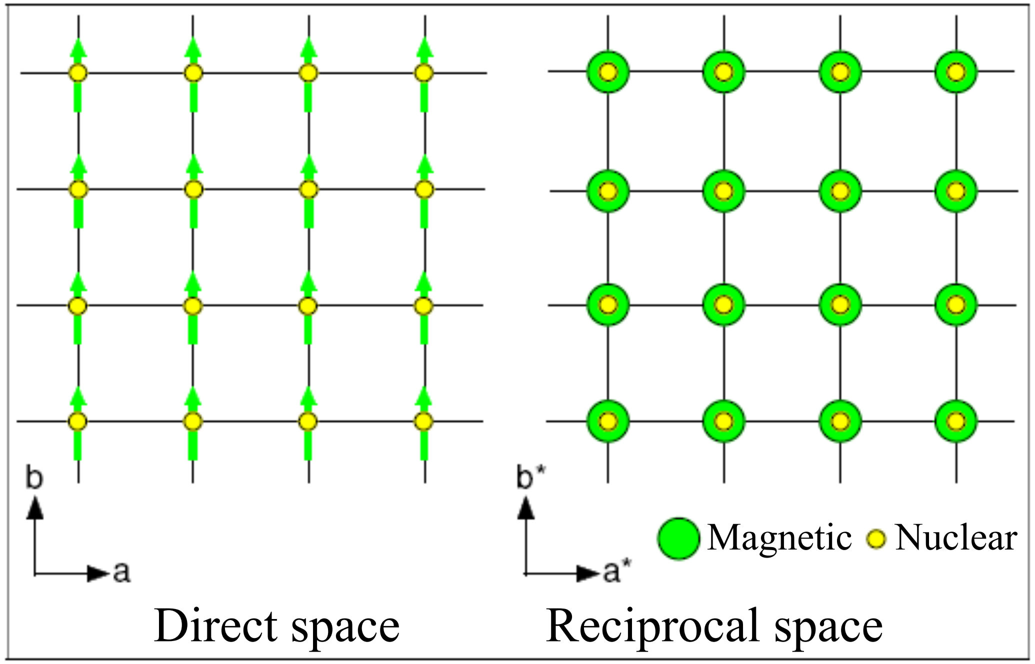
- **What is the magnetic structure described by a zero propagation vector?**
- **What is the propagation vector describing a type A antiferromagnet?**
- **What is the propagation vector associated to a magnetic helix of periodicity 8a?**

Diffraction by a crystal: nuclear and magnetic structures

MAGNETIC DIFFRACTION: CLASSIFICATION OF THE MAGNETIC STRUCTURES

If $\vec{\tau} = 0$, magnetic/nuclear structures same periodicity
→ Bragg peaks at reciprocal lattice nodes $\vec{Q} = \vec{H}$

- Bravais lattice $\vec{\tau}$=0 ⇒ ferromagnetic structure



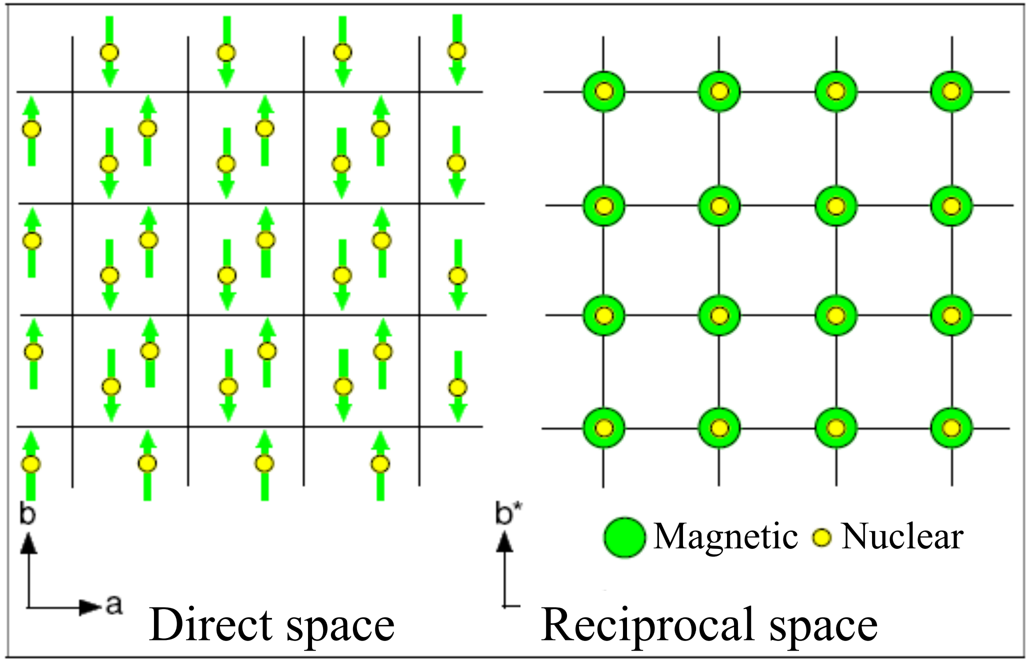
Diffraction by a crystal: nuclear and magnetic structures

MAGNETIC DIFFRACTION: CLASSIFICATION OF THE MAGNETIC STRUCTURES

If $\vec{\tau} = 0$, magnetic/nuclear structures same periodicity
→ Bragg peaks at reciprocal lattice nodes $\vec{Q} = \vec{H}$

•Non Bravais lattice $\vec{\tau}$=0 ⇒ ferromagnetic or antiferromagnetic structure

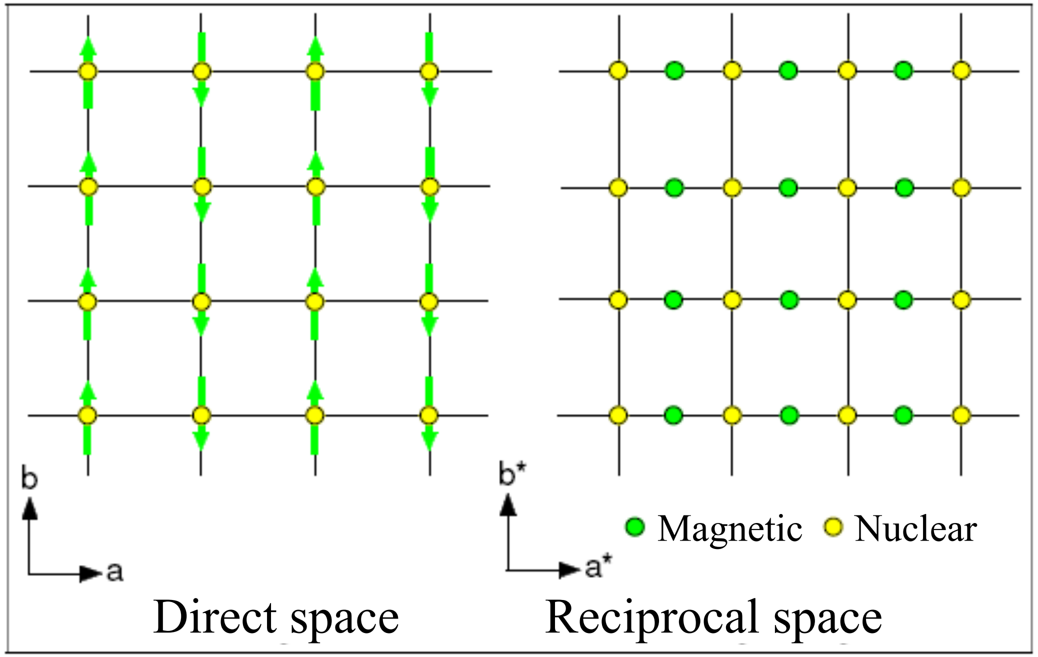
Intensities
of magnetic peaks
→Arrangement of
moments in cell



MAGNETIC DIFFRACTION: CLASSIFICATION OF THE MAGNETIC STRUCTURES

If $\vec{\tau} \neq 0$, magnetic satellites at $\vec{Q} = \vec{H} \pm \vec{\tau}$

Ex. $\vec{\tau} = \vec{H}/2$=(1/2,0,0) , collinear antiferromagnetic structure

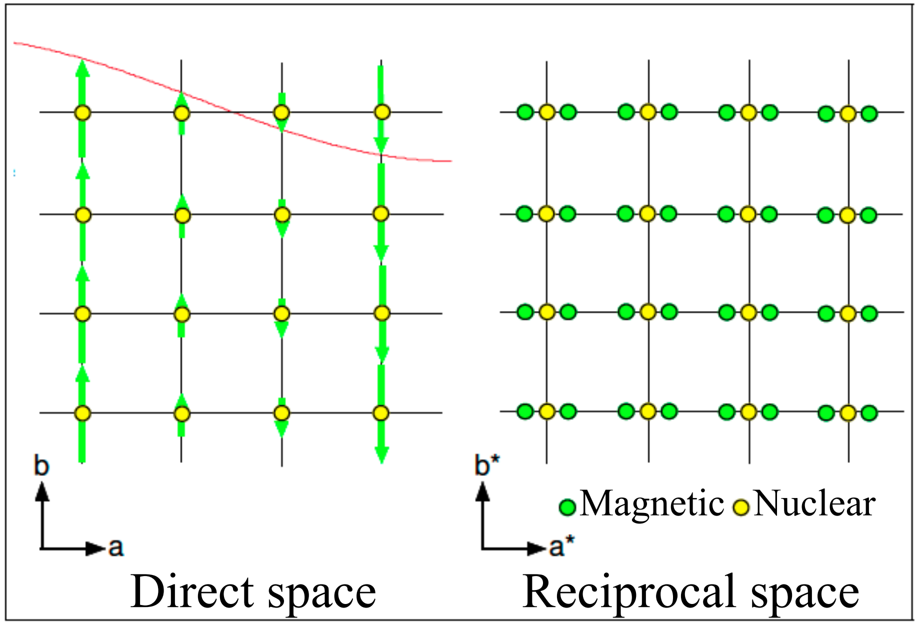


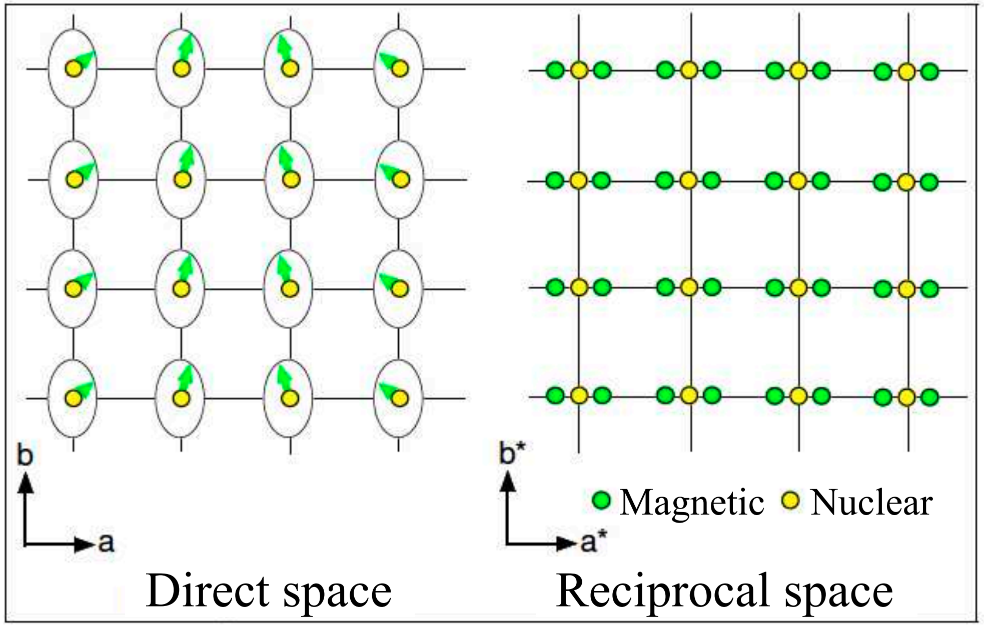
Diffraction by a crystal: nuclear and magnetic structures

MAGNETIC DIFFRACTION: CLASSIFICATION OF THE MAGNETIC STRUCTURES

If $\vec{\tau} \neq 0$ and $\vec{\tau} \neq \vec{H}/2$, magnetic satellites at $\vec{Q} = \vec{H} \pm \vec{\tau}$

Sine wave amplitude modulated and spiral structures $\vec{\tau} = (\tau, 0, 0)$





$$\vec{\mu}_{n\nu} = \mu_\nu \hat{u} \cos(\vec{\tau}.\vec{R}_n + \Phi_\nu)$$

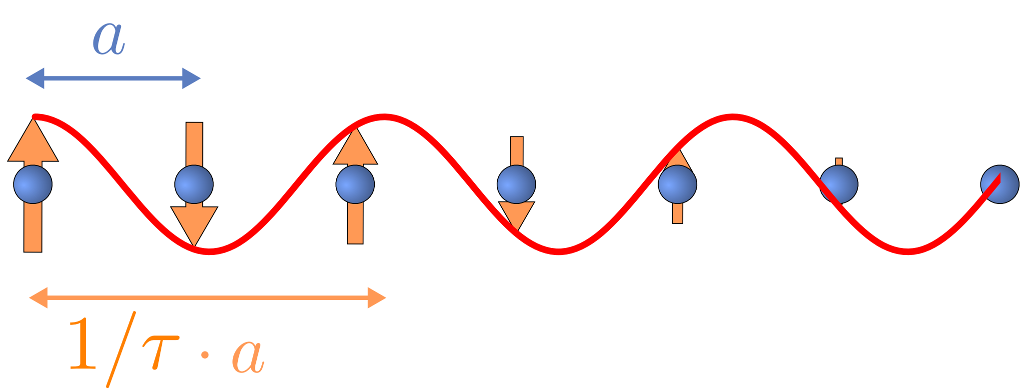
$$\vec{\mu}_{n\nu} = \mu_{1\nu} \hat{u} \cos(\vec{\tau}.\vec{R}_n + \Phi_\nu) + \mu_{2\nu} \hat{v} \sin(\vec{\tau}.\vec{R}_n + \Phi_\nu)$$

Diffraction by a crystal: nuclear and magnetic structures

MAGNETIC DIFFRACTION: CLASSIFICATION OF THE MAGNETIC STRUCTURES

If $\vec{\tau} \neq 0$ and $\vec{\tau} \neq \vec{H}/2$, magnetic satellites at $\vec{Q} = \vec{H} \pm \vec{\tau}$

Sine wave amplitude modulated and spiral structures $\vec{\tau} = (\tau, 0, 0)$



Rational/irrational $\vec{\tau}$ = **commensurate/incommensurate** magnetic structure

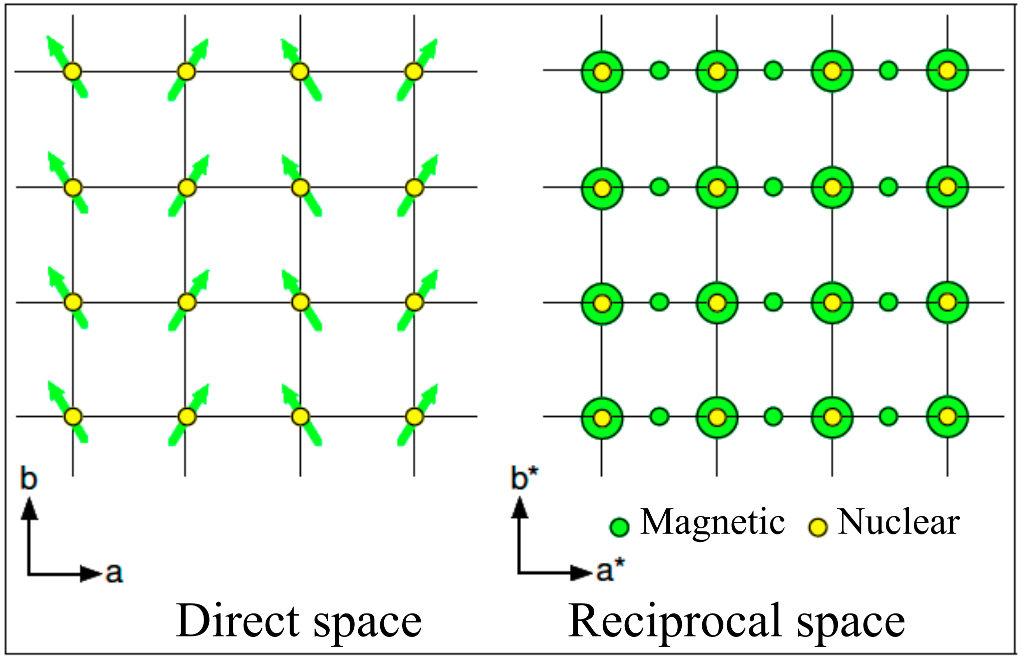
Diffraction by a crystal: nuclear and magnetic structures

MAGNETIC DIFFRACTION: CLASSIFICATION OF THE MAGNETIC STRUCTURES

Multi-$\vec{\tau}$ magnetic structure

$$\frac{d\sigma}{d\Omega} = \frac{(2\pi)^3}{V} \sum_{\vec{H}} \sum_{\vec{\tau}} |\vec{F}_{M\perp}(\vec{Q})|^2 \delta(\vec{Q} - \vec{H} - \vec{\tau})$$

Ex. canted structure with $\vec{\tau} = 0$ and $\vec{\tau} = \vec{H}/2$ = (1/2,0,0)

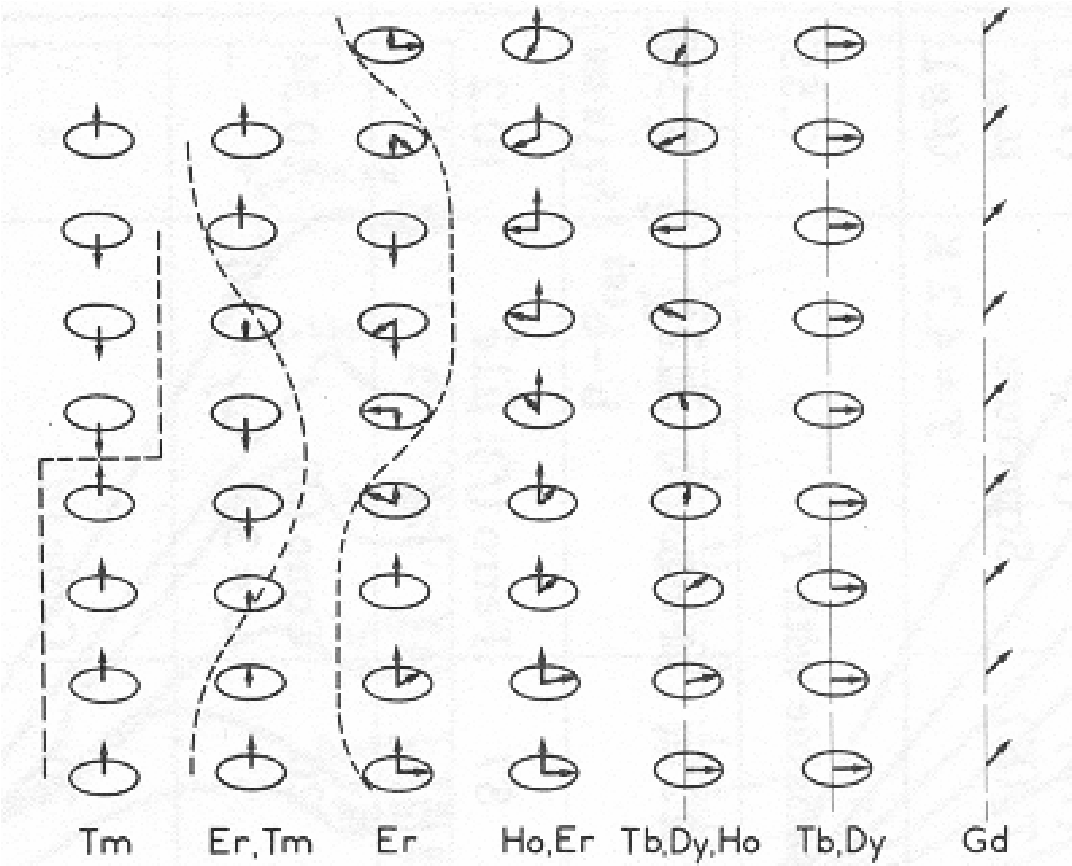


Diffraction by a crystal: nuclear and magnetic structures

MAGNETIC DIFFRACTION: CLASSIFICATION OF THE MAGNETIC STRUCTURES

Ex. in rare earth metals

Complex magnetic structures :
Sine wave amplitude modulated
spiral (helix, cycloid), canted structures
→ due to frustration, competition of interactions, Dzyaloshinskii-Moryia/ anisotropic interaction…



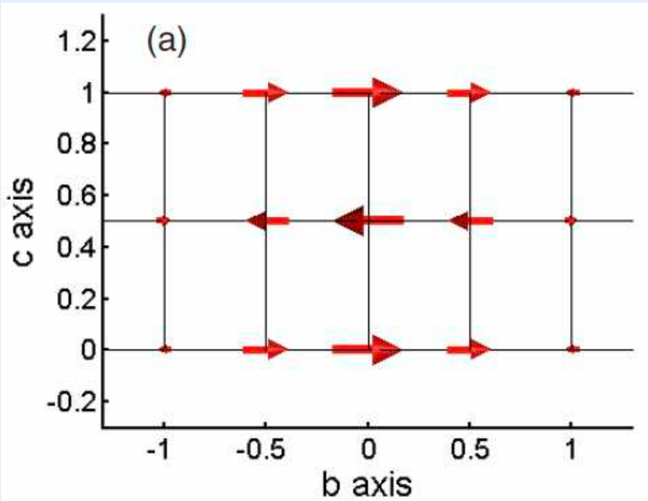
MAGNETIC DIFFRACTION: CLASSIFICATION OF THE MAGNETIC STRUCTURES

Sine wave amplitude modulated
spiral (helix, cycloid), canted structures

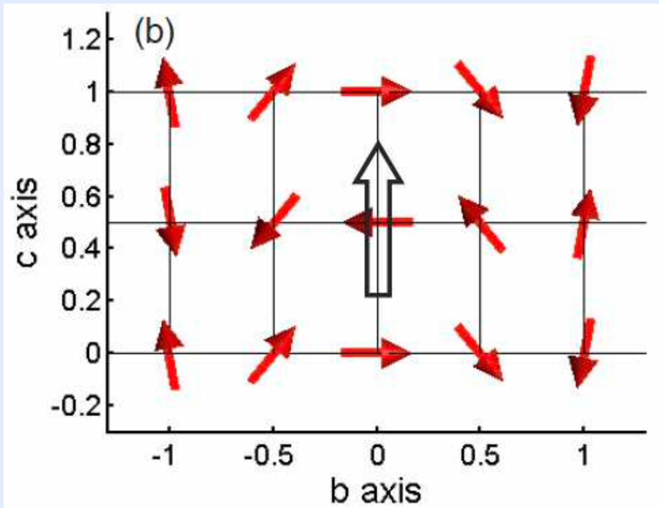
Ex. in multiferroics

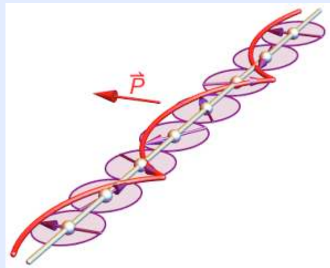
$TbMnO_3$

Kenzelmann et al., PRL 2007



28 K<T<41 K :
incommensurate sine wave modulated
→paraelectric





T<28 K :
commensurate spiral (cycloid)
→ferroelectric

Diffraction by a crystal: nuclear and magnetic structures

MAGNETIC DIFFRACTION: TECHNIQUES

Powder diffraction $I(|\vec{Q}|)$

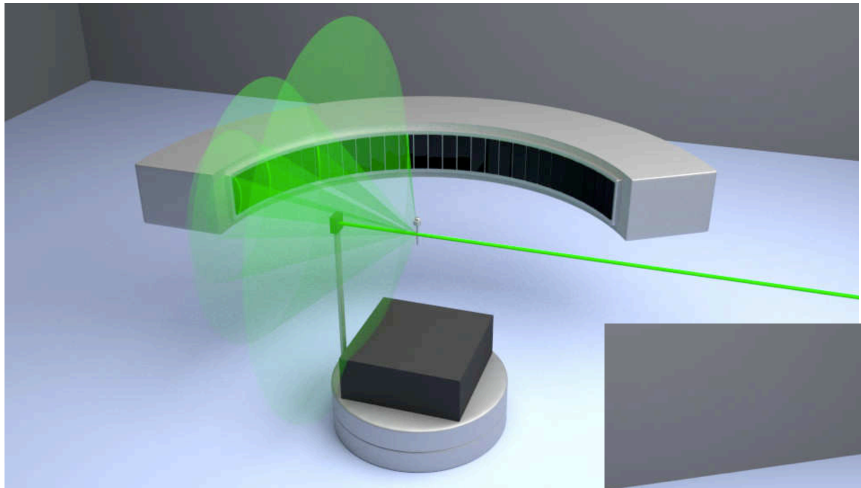
Bragg's law $Q = \dfrac{2\sin\theta}{\lambda}$

Ex. Fixed λ and varying θ (or multidetector)

Single-crystal diffraction $I(\vec{Q})$

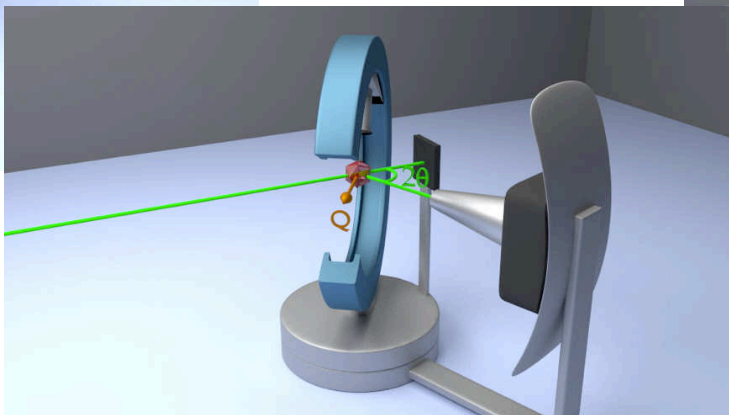
Complex structures, magnetic domains, bulky environments

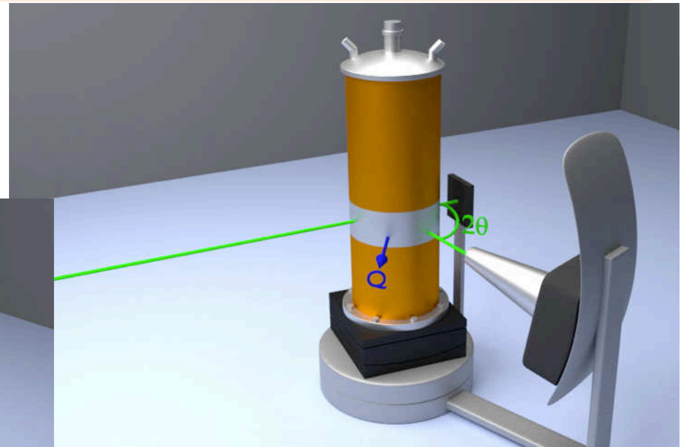
→Bring a reciprocal node in coincidence with $\vec{Q} = \vec{k}_i - \vec{k}_f$ then measure the integrated intensity (rocking curve)



Powder diffratometer

4-circles mode





Lifting arm

MAGNETIC DIFFRACTION: SOLVING A MAGNETIC STRUCTURE

Finding the propagation vector $\vec{\tau}$ (periodicity of magnetic structure):
powder diffraction → difference between measurements below and above T_c.
Indexing magnetic Bragg reflections with $\vec{Q} = \vec{H} \pm \vec{\tau}$

Refining magnetic Bragg peaks intensities (powder and single-crystal)
and domain populations (single-crystal)
→ moment amplitudes and magnetic arrangement of atoms in the cell
(use scaling factor from nuclear structure refinement) with programs like Fullprof

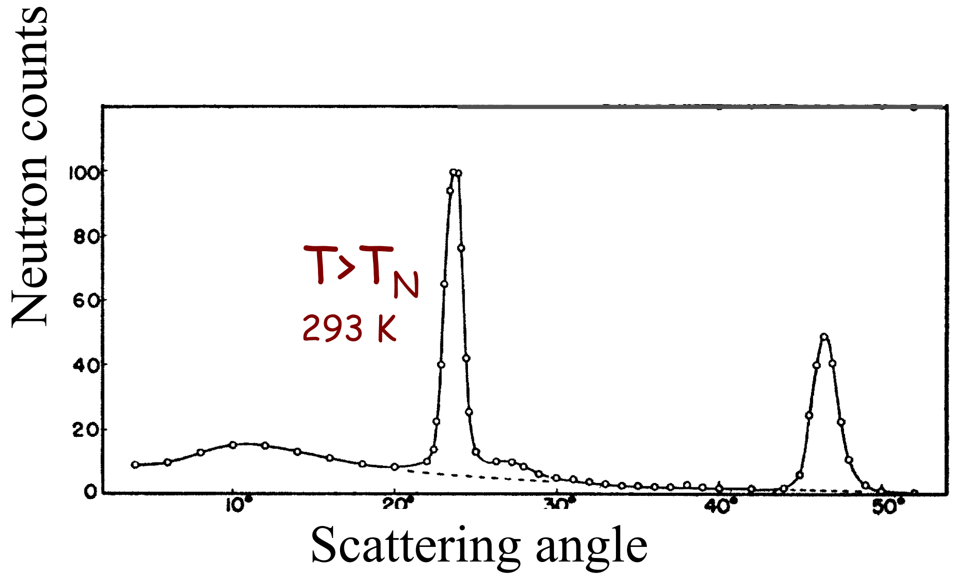
https://www.ill.eu/sites/fullprof/

Help from group theory and representation analysis
Use of rotation/inversion symmetries to infer possible magnetic arrangements compatible with the symmetry group that leaves the propagation vector invariant → constrains the refinement

MAGNETIC DIFFRACTION: EXAMPLES

Original powder diffraction experiment in **MnO** from Shull *et al.*
Phys. Rev. (1951)

T_N=116 K



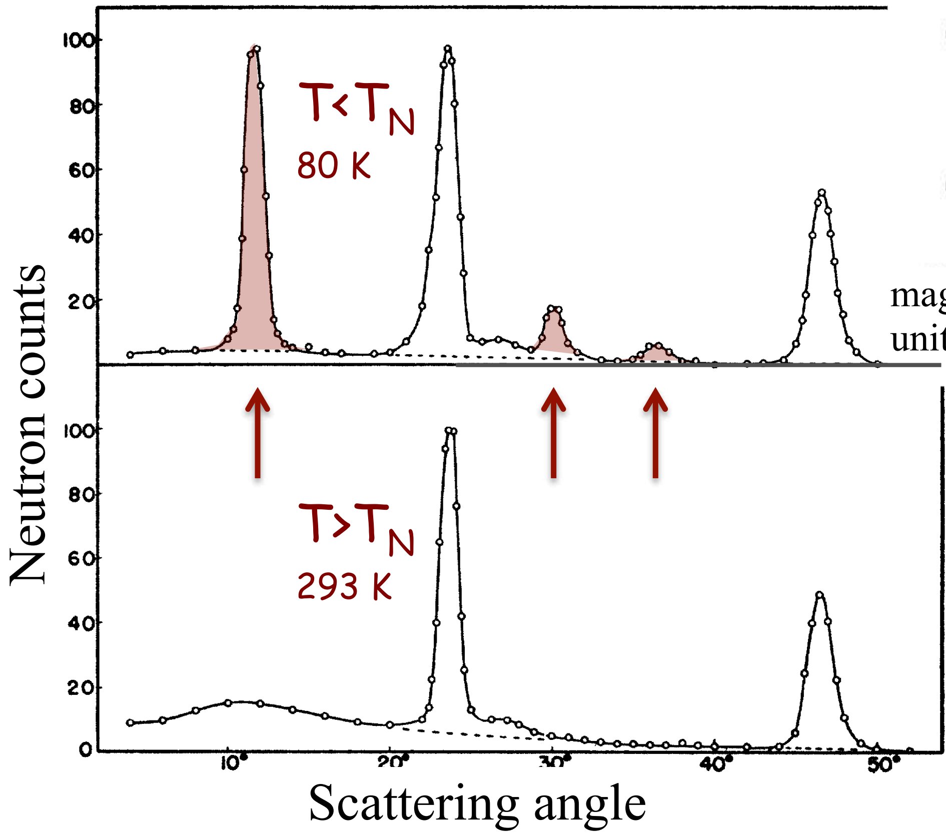
Diffraction by a crystal: nuclear and magnetic structures

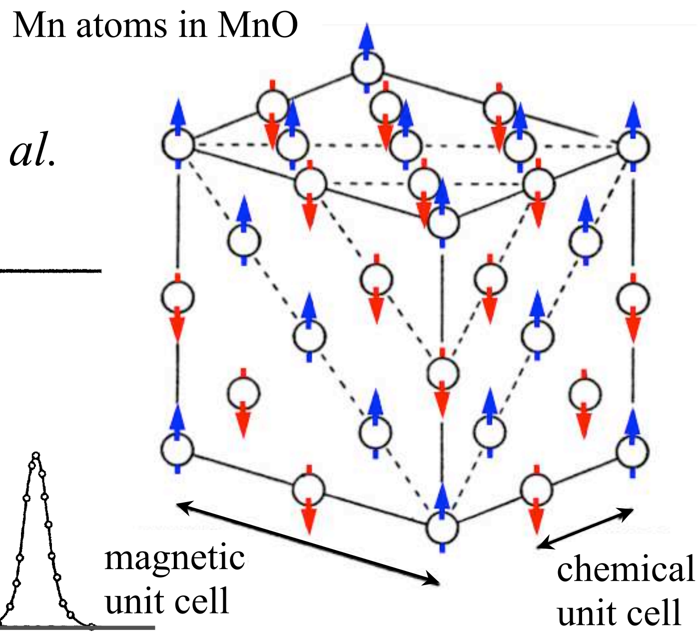
MAGNETIC DIFFRACTION: EXAMPLES

Original powder diffraction experiment in **MnO** from Shull *et al.*
Phys. Rev. (1951)

T_N=116 K

Propagation vector
(½, ½, ½)

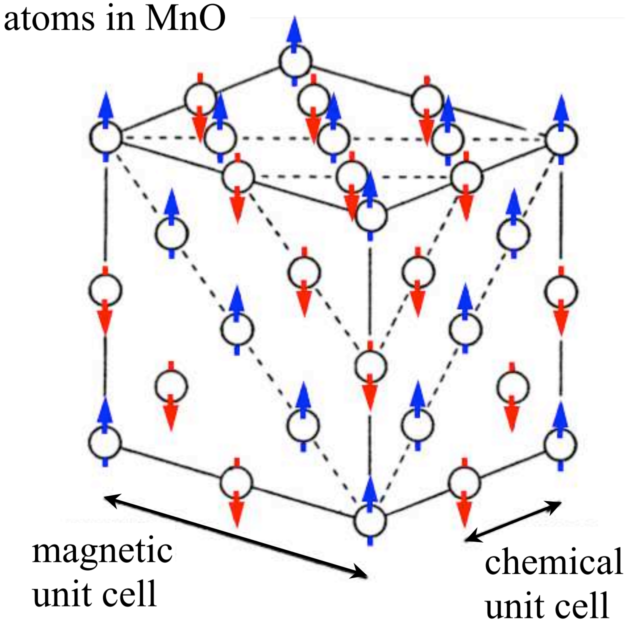




MAGNETIC DIFFRACTION: EXAMPLES

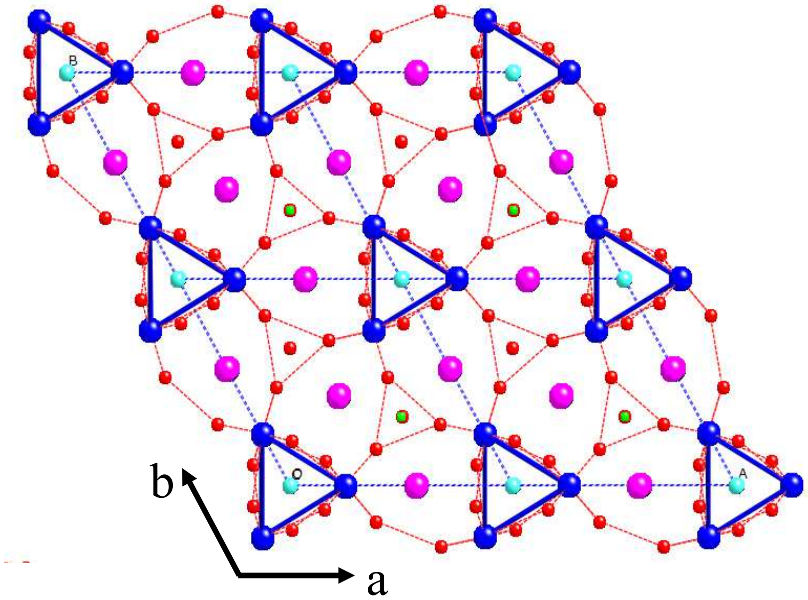
Original powder diffraction experiment in **MnO** from Shull *et al.* *Phys. Rev. (1951)*

- Confirmation of antiferromagnetism
  Predicted by *Louis Néel* in 1936



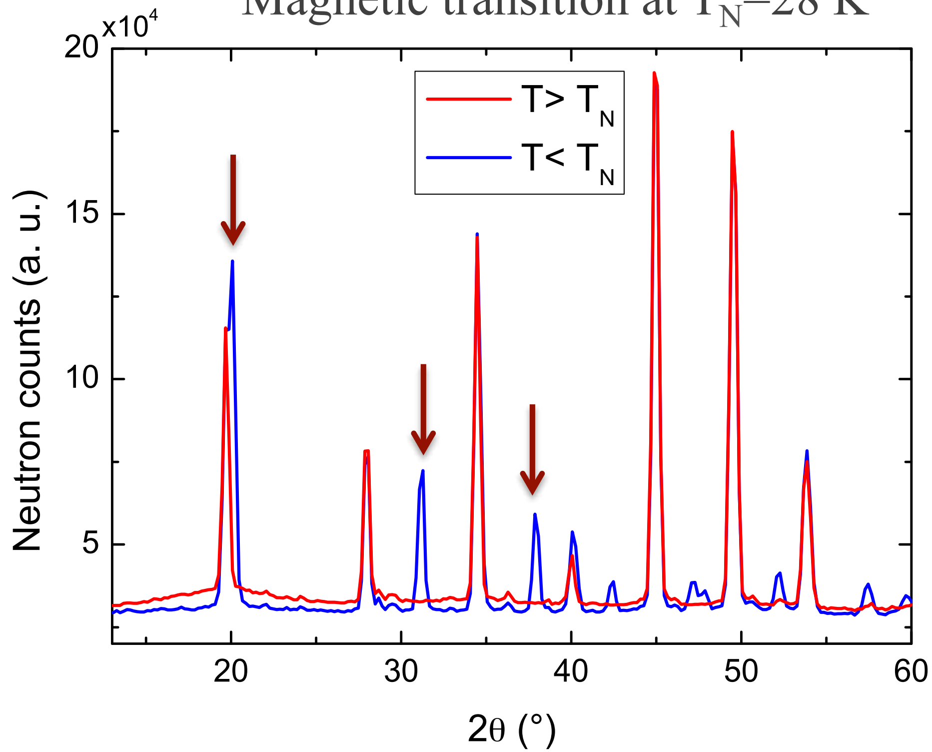
MAGNETIC DIFFRACTION: EXAMPLES

$Ba_3NbFe_3Si_2O_{14}$ *Marty et al., PRL 2008*



triangular lattice of Fe^{3+} triangles, S=5/2

Powder diffraction



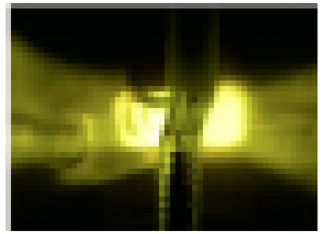
→ Propagation vector $\vec{\tau}$ = (0, 0, 1/7)

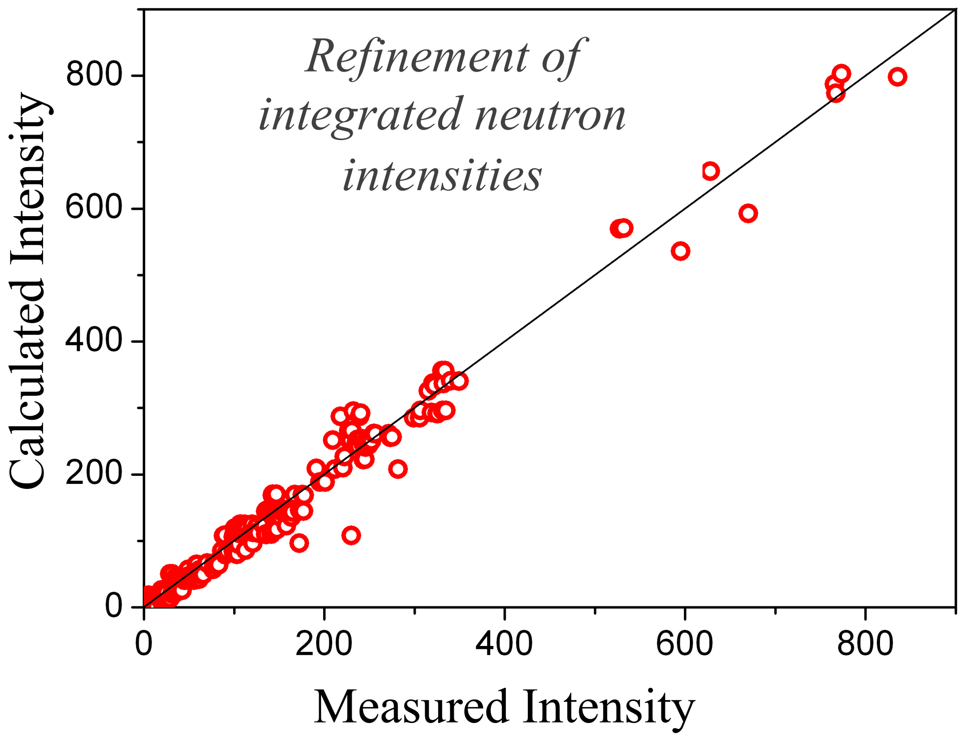
Diffraction by a crystal: nuclear and magnetic structures

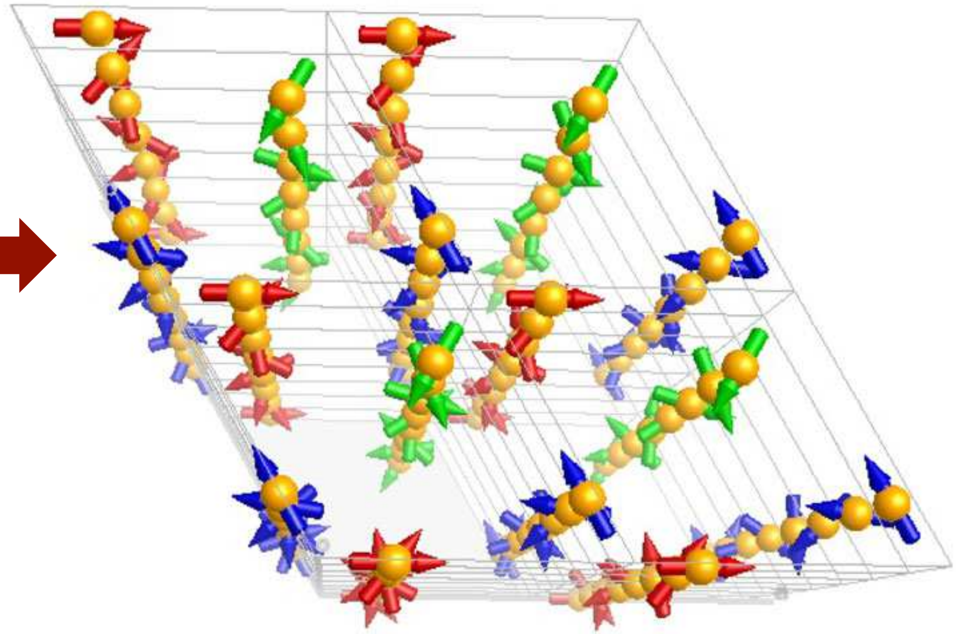
MAGNETIC DIFFRACTION: EXAMPLES

$Ba_3NbFe_3Si_2O_{14}$ *Marty et al., PRL 2008*

Single-crystal diffraction





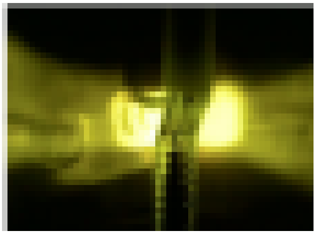


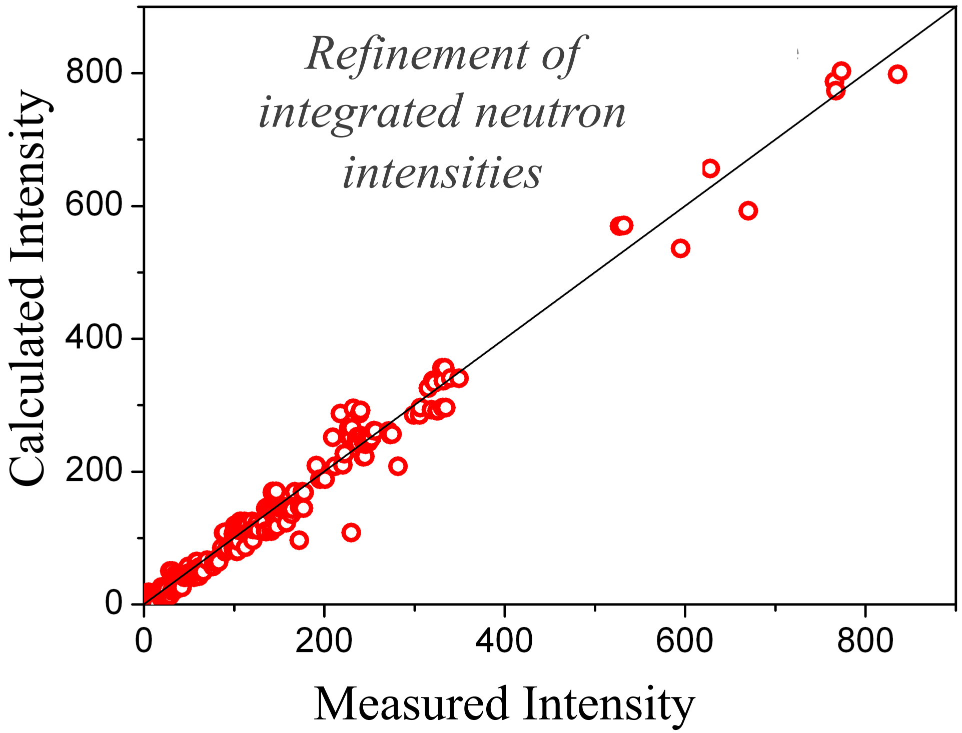
Diffraction by a crystal: nuclear and magnetic structures

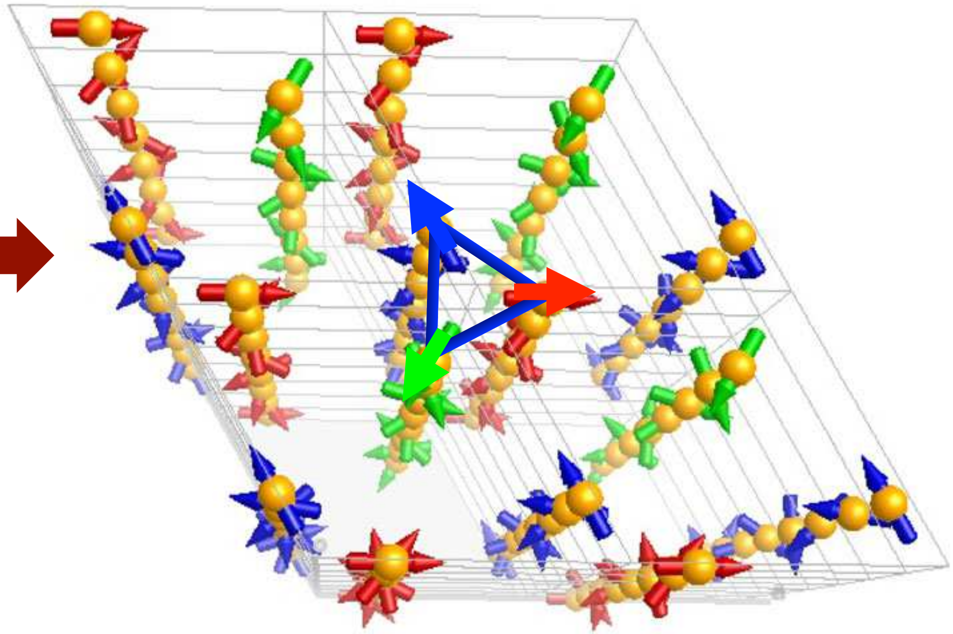
MAGNETIC DIFFRACTION: EXAMPLES

$Ba_3NbFe_3Si_2O_{14}$ *Marty et al., PRL 2008*

Single-crystal diffraction



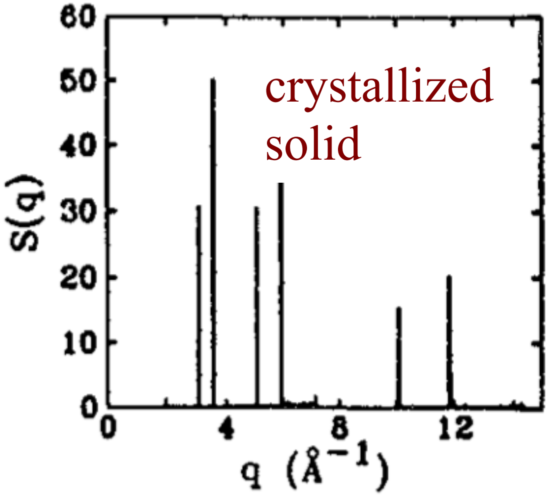


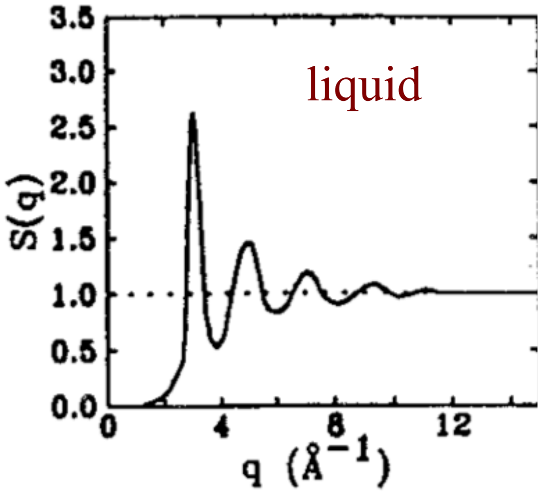


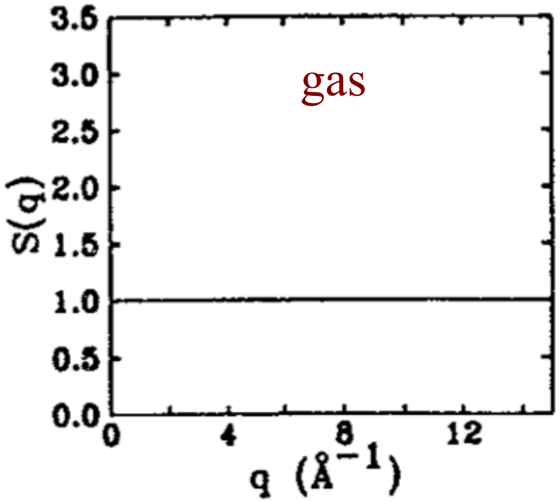
- ➢ Triangles of magnetic moments in (a, b) plane
- ➢ Magnetic helices propagating along *c* with period ≈7c

Diffraction by a ill-ordered magnetic systems

atomic states







Magnetic states

Magnetic order

Spin liquid

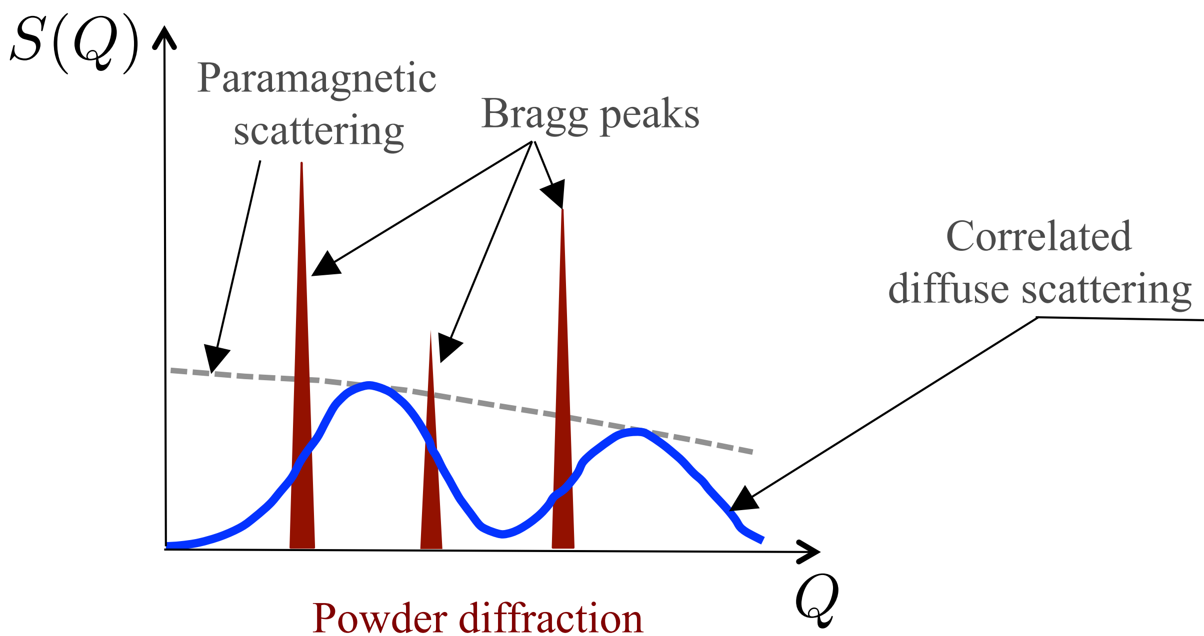
paramagnet

Diffraction by a ill-ordered magnetic systems

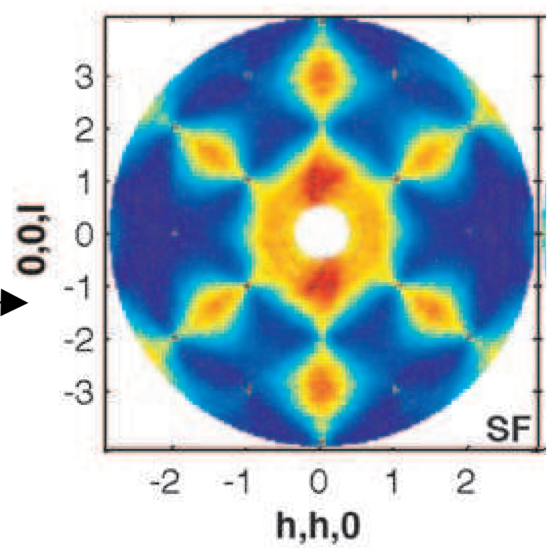
Ex.: Spin liquid = no order/strong fluctuations despite presence of spin pair correlations

Diffuse neutron scattering map of spin ice

Fennell et al., Science 2009



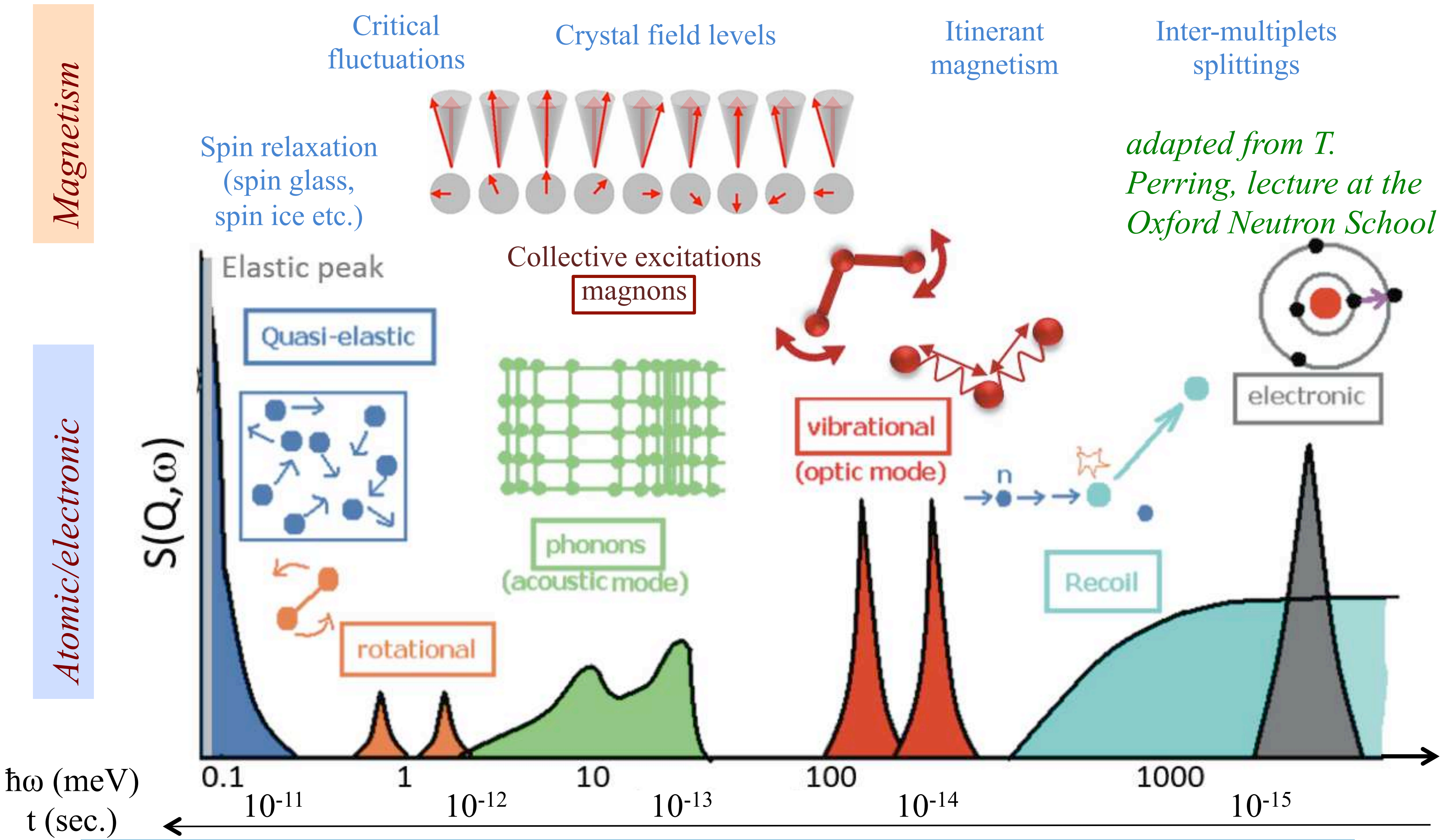
Powder diffraction



Single-crystal diffraction

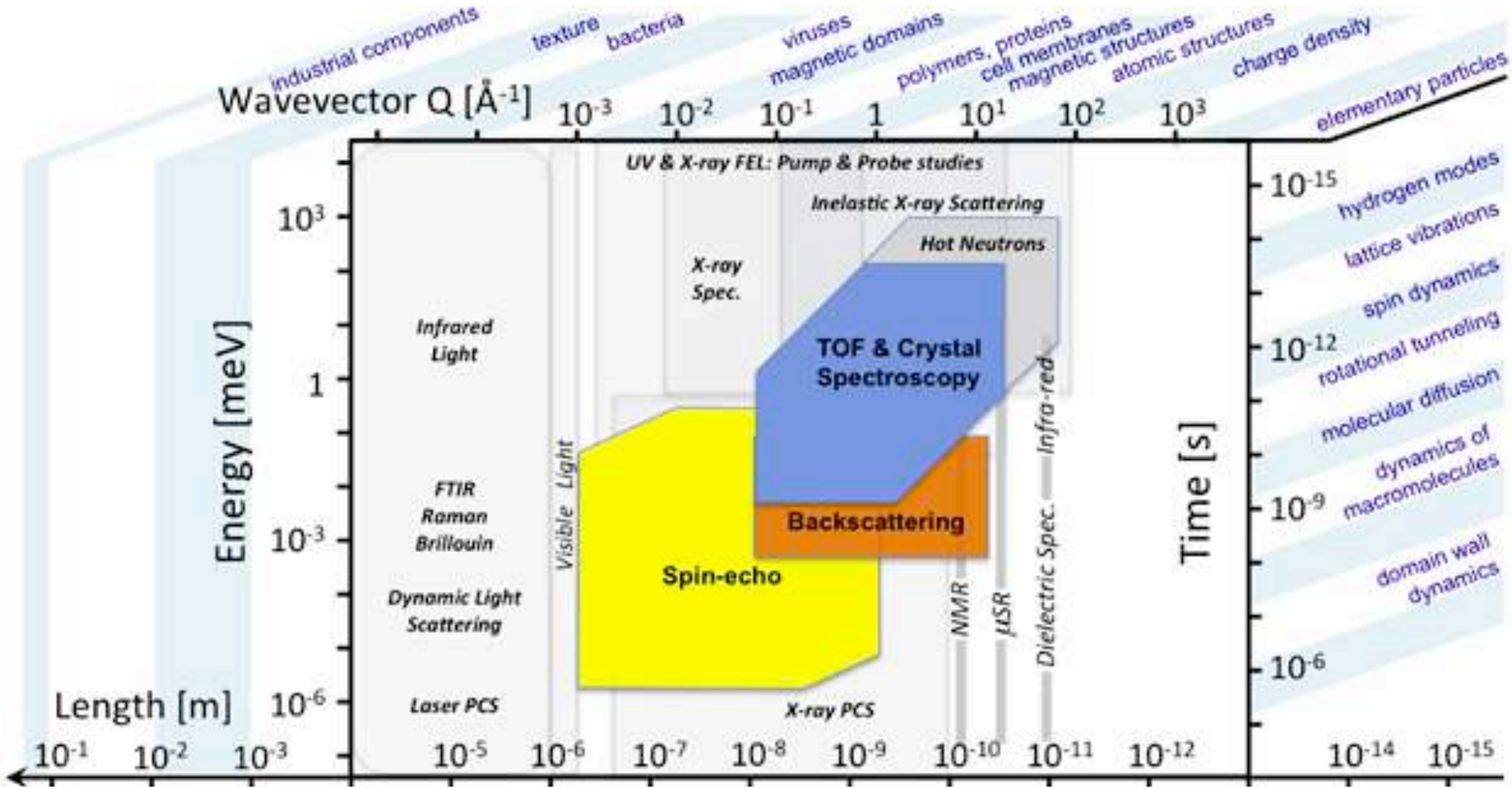
Inelastic neutron scattering: magnetic excitations

Inelastic neutron scattering: nuclear and magnetic excitations



adapted from T. Perring, lecture at the Oxford Neutron School

Inelastic neutron scattering: magnetic excitations



https://europeanspallationsource.se/science-using-neutrons

Inelastic neutron scattering: magnetic excitations

INELASTIC SCATTERING: MAGNETIC

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'}\int_{-\infty}^{+\infty}\langle A^*_{j'}(0)A_j(t)e^{-i\vec{Q}\vec{R}_{j'}(0)}e^{i\vec{Q}\vec{R}_j(t)}\rangle e^{-i\omega t}dt$$

with $A_j(t) = pf_j(Q)\vec{M}_{j\perp}(\vec{Q},t)$

again some algebra

$$\frac{d\sigma^2}{d\Omega dE} = (\gamma r_0^2)\frac{k_f}{k_i}f^2(\vec{Q})\sum_{\alpha,\beta}\left[\delta_{\alpha,\beta} - \frac{Q_\alpha Q_\beta}{Q^2}\right]S^{\alpha,\beta}(\vec{Q},\omega)$$

Inelastic neutron scattering: magnetic excitations

INELASTIC SCATTERING: MAGNETIC

$$\frac{d\sigma^2}{d\Omega dE} = (\gamma r_0^2)\frac{k_f}{k_i}\underbrace{f^2(\vec{Q})}_{\text{Magnetic form factor (squared)}}\sum_{\alpha,\beta}\Bigg[\underbrace{\delta_{\alpha,\beta} - \frac{Q_\alpha Q_\beta}{Q^2}}_{\text{Polarization factor}}\Bigg]\underbrace{S^{\alpha,\beta}(\vec{Q},\omega)}$$

Scattering function: spin-spin correlation function $\langle \vec{S}^{\alpha}_{j'}(0)\vec{S}^{\beta}_{j}(t)\rangle$

related to the dynamical susceptibility via the fluctuation-dissipation theorem

$$S^{\alpha,\beta}(\vec{Q},\omega) = \frac{1}{1-\exp(-\hbar\omega/k_BT)}\chi''^{\alpha,\beta}(\vec{Q},\omega)$$

Inelastic neutron scattering: magnetic excitations

INELASTIC SCATTERING: SPIN WAVES

Quantum description: spin wave mode = quasi-particle called magnon

Creation/annihilation processes in cross-section

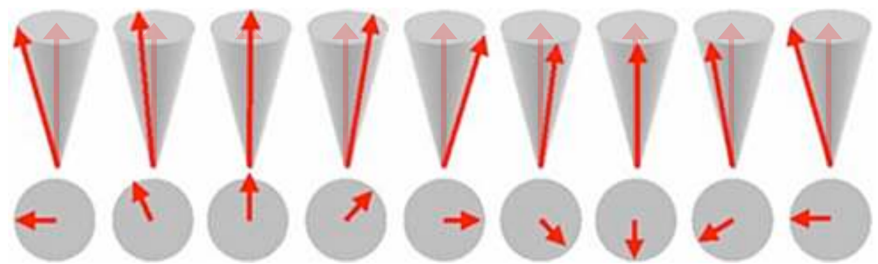
dynamical magnetic structure factor

$$\frac{d^2\sigma}{d\Omega dE} = (\gamma r_0)^2 \frac{k_f}{k_i} \frac{(2\pi)^3}{V} \sum_{\vec{H}} \sum_{\vec{q}} f(Q)^2 |F(\vec{Q})|^2 \boxed{< n_\pm > \delta(\omega \mp \omega_{\vec{q}})} \begin{cases} \delta(\vec{Q} - \vec{H} - \vec{q}) \\ \delta(\vec{Q} - \vec{H} + \vec{q}) \end{cases}$$

INELASTIC SCATTERING: SPIN WAVES

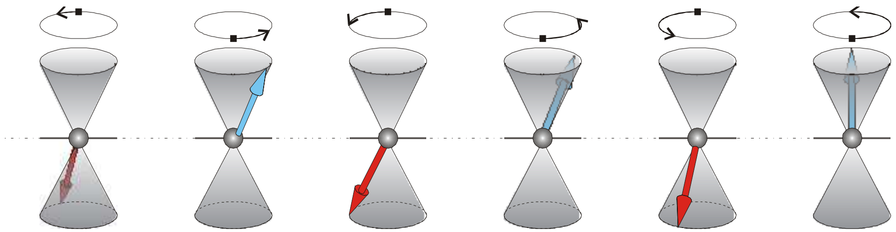
Spin waves (magnons): elementary excitations of magnetic compounds= transverse oscillations in relative orientation of the spins

Characterized by wave vector $\vec{q}$, a frequency ω
Only certain spin components involved



Ferromagnetic
J >0

$$\mathcal{H} = -\sum_{i,j} J_{ij}\vec{S}_i.\vec{S}_j$$



Antiferromagnetic
J<0

Inelastic neutron scattering: magnetic excitations

INELASTIC SCATTERING: SPIN WAVES

Spin waves (magnons): elementary excitations of magnetic compounds= transverse oscillations in relative orientation of the spins

Characterized by wave vector $\vec{q}$, a frequency ω
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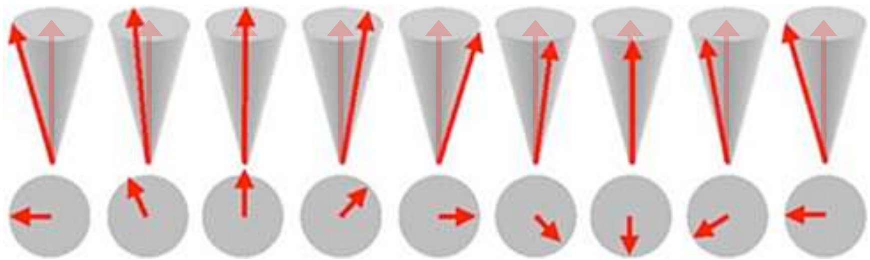
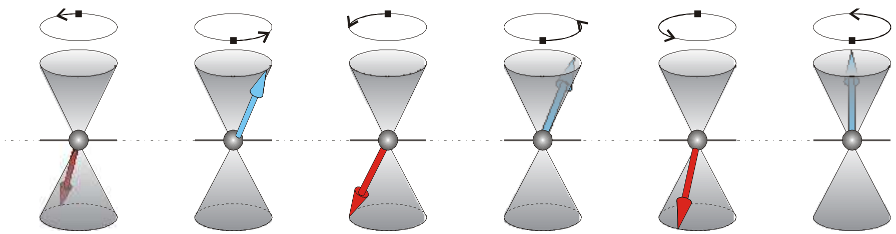
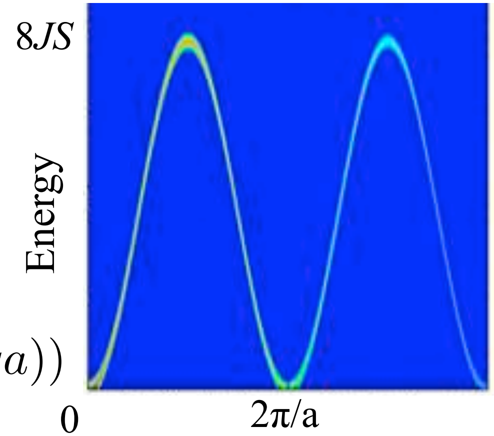




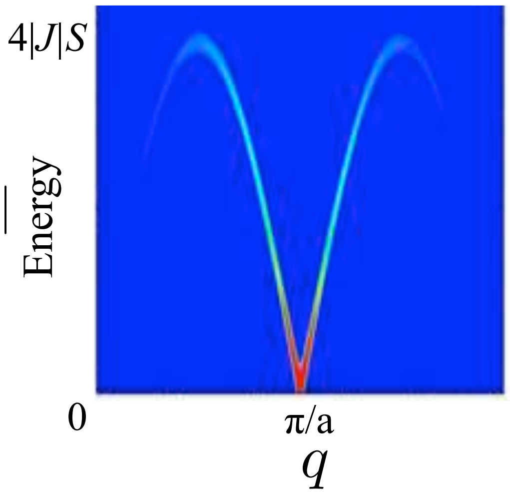
Dispersion relation $\omega(\vec{q})$

Crystal with p atoms/unit cell:
→p branches



$$E(q) = 4JS(1 - \cos(qa))$$

Ferromagnetic
J >0



$$E(q) = -4JS|\sin(qa)|$$

Antiferromagnetic
J<0

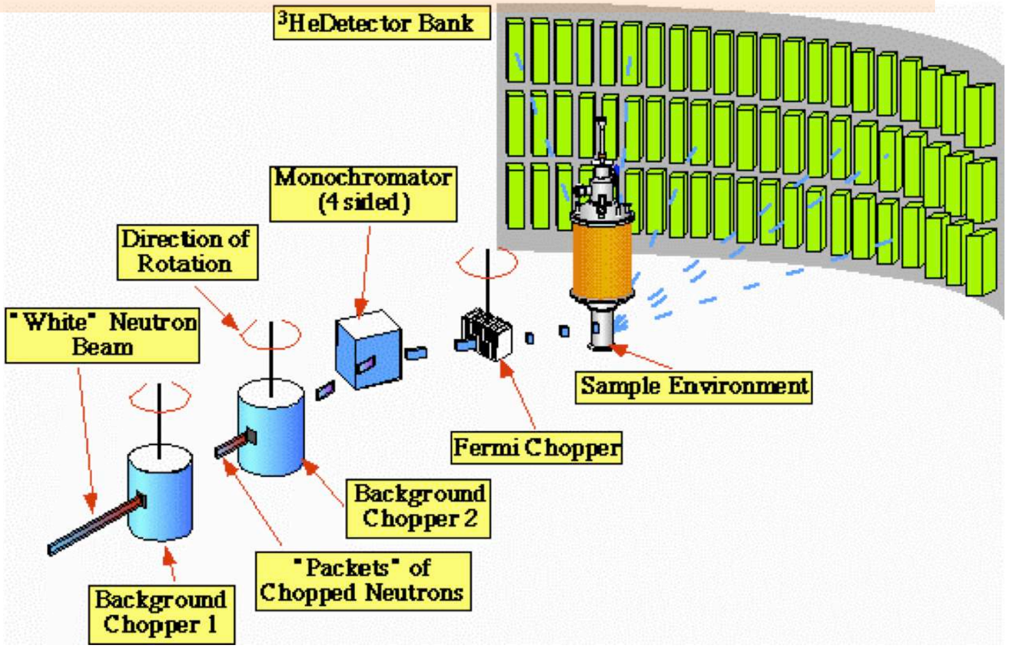
Inelastic neutron scattering: magnetic excitations

INELASTIC SCATTERING: TECHNIQUES

Instrument time-of-flight

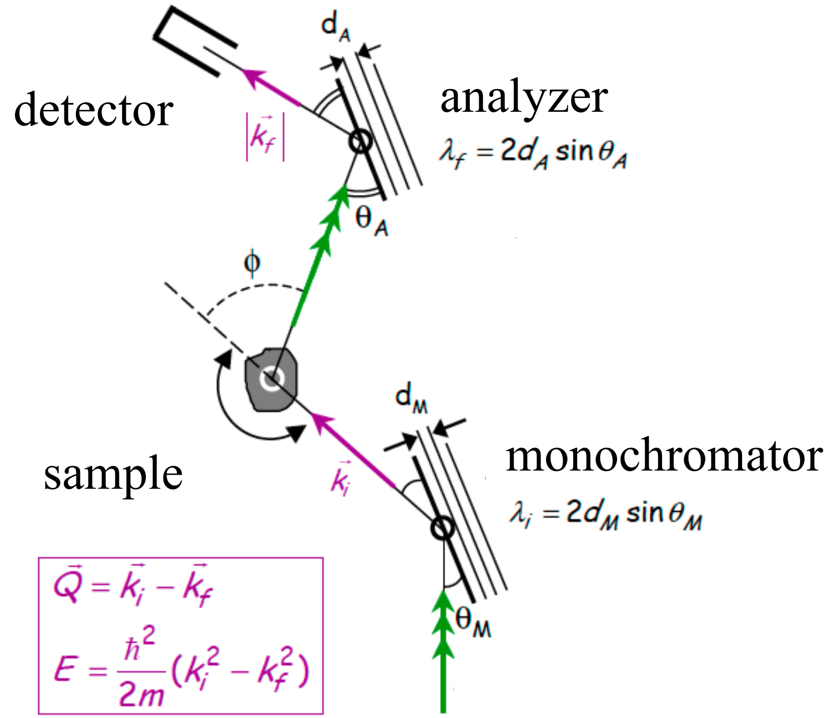
-Neutron pulses (spallation/chopped): time and position on multidetector give final E and $\vec{Q}$

-Powder and single-crystal: access to wide region of reciprocal space



Instrument triple-axis

Position at $\vec{Q}$ point and energy analyzer: single-crystal, bulky sample environment, polarized neutrons



Inelastic neutron scattering: magnetic excitations

INELASTIC SCATTERING: NUCLEAR *VERSUS* MAGNETIC

Nuclear excitations (phonons)

Form factor $\propto Q^2$
Intensity max for $\vec{Q}||\vec{e}$ and zero for $\vec{Q}\perp\vec{e}$
with $\vec{e}$ the polarization of the mode

Magnetic excitations (spin waves)

Form factor ↘ with Q
Intensity maximum for $\vec{M}\perp\vec{Q}$

Purposes of inelastic scattering experiments:

Nuclear: Information on elastic constants, sound velocity, structural instabilities…

Magnetic: Information on magnetic interactions and microscopic mechanisms yielding the magnetic properties…

In multiferroics: Spin-lattice coupling, hybrid modes ex. electromagnons

Inelastic neutron scattering: magnetic excitations

INELASTIC SCATTERING: EXAMPLES

Spin waves in MnO

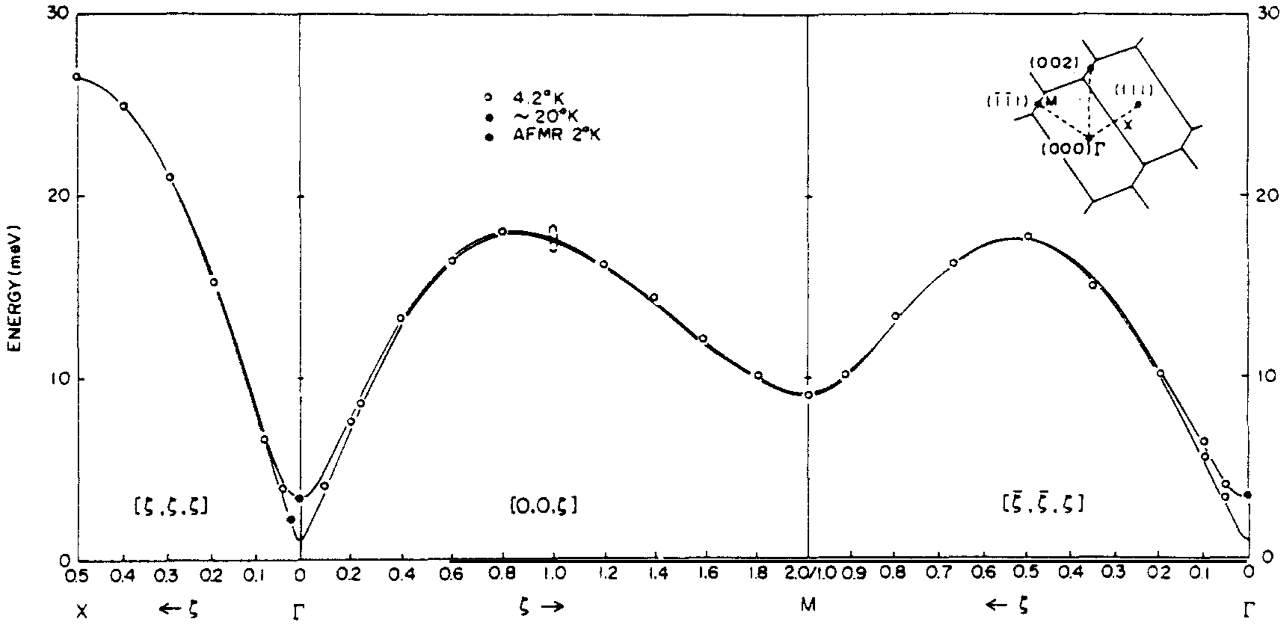


FIG. 1. Dispersion relation $E(q)$ of the spin waves in MnO measured along [111], [001] and [$\bar{1}\bar{1}$1].

➡ J_1=0.77±0.02, J_2=0.89±0.02 meV, + (anisotropies, exchange striction…)

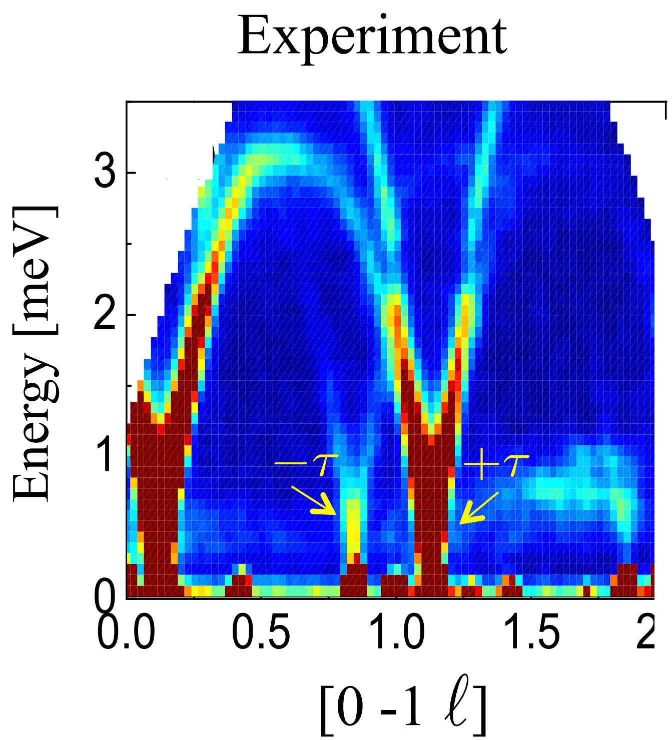
Bonfante et al. Solid State Com. 1972
Kohgi et al. Solid State Com. 1972

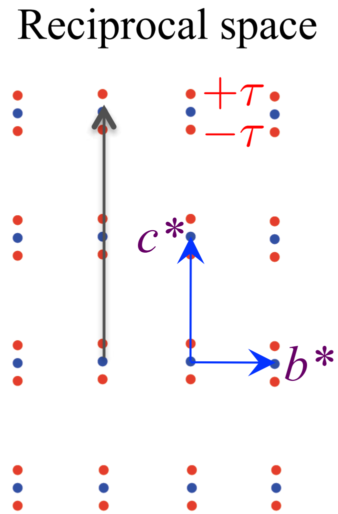
Inelastic neutron scattering: magnetic excitations

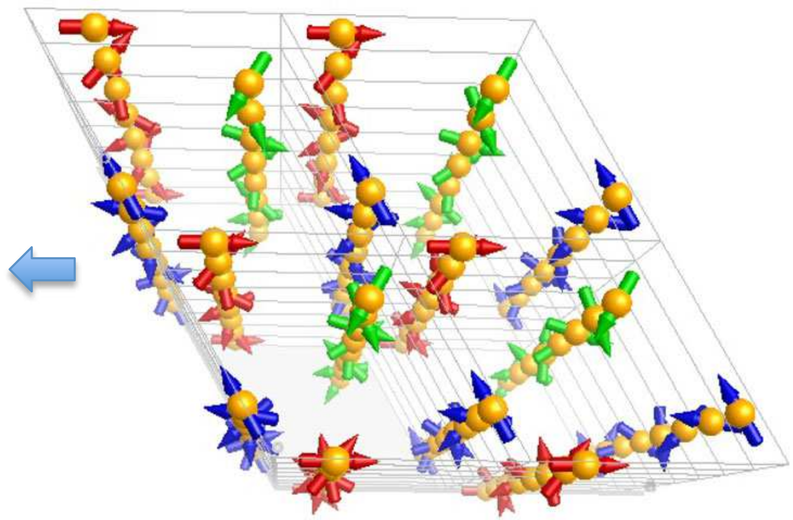
INELASTIC SCATTERING: EXAMPLES

Spin waves in $Ba_3NbFe_3Si_2O_{14}$ single crystal

Loire et al. PRL 2011
Chaix et al. PRB 2016







$\vec{\tau} = (0, 0, 1/7)$

http://www-llb.cea.fr/logicielsllb/SpinWave/SW.html
https://www.psi.ch/de/spinw

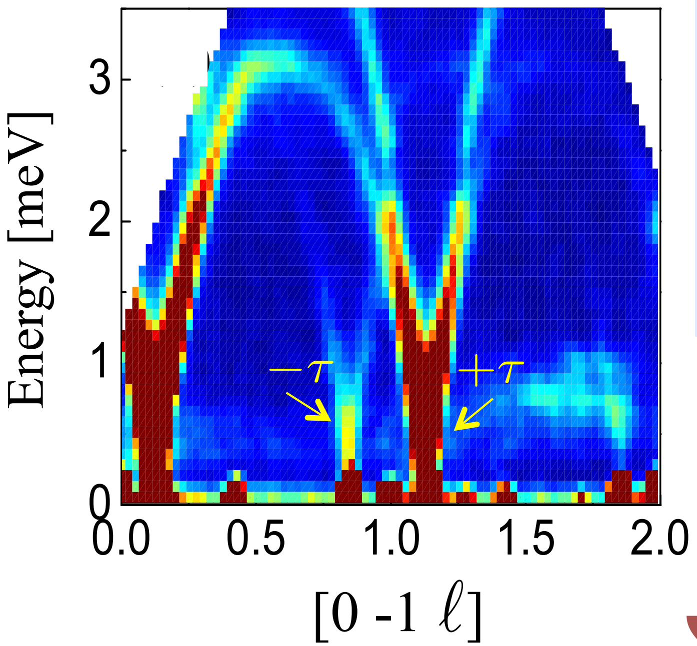
Analysis of spin waves dispersion using Holstein-Primakov formalism in linear approximation
Magnetic structure and Hamiltonian are inputs of existing programs (SpinWave, SpinW)

Inelastic neutron scattering: magnetic excitations

INELASTIC SCATTERING: EXAMPLES

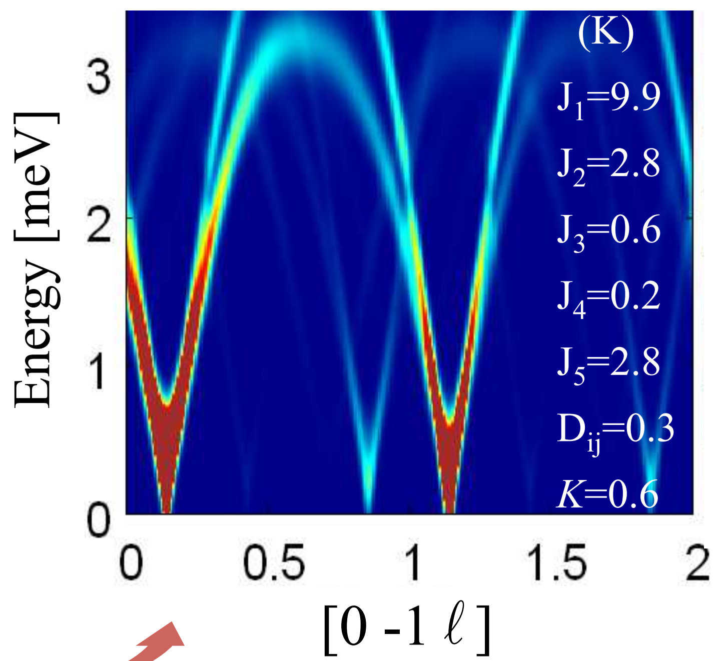
Spin waves in $Ba_3NbFe_3Si_2O_{14}$ single crystal

Experiment



- Determination of the Hamiltonian
- Interpretation of multiferroic properties

Calculation



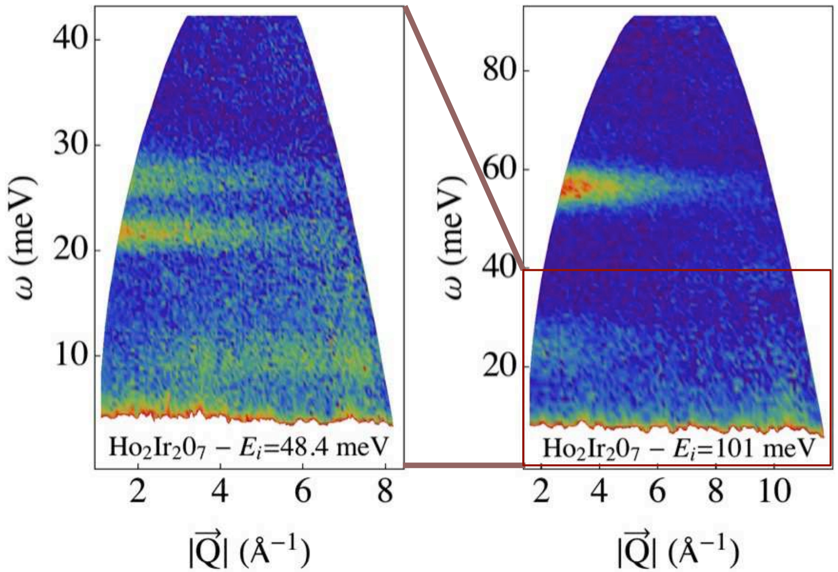
$$H = \sum_{ij} J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j + \sum_{ij\triangle} \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \sum_{i,\alpha} K_\alpha (\hat{\mathbf{n}}_\alpha \cdot \mathbf{S}_i)^2$$

Inelastic neutron scattering: magnetic excitations

INELASTIC SCATTERING: EXAMPLES OTHER THAN SPIN WAVES

Localized excitations

Transition between energy levels :
Discrete non dispersive signal
Example crystal field excitations in rare-earth ions



Ho^{3+} in $Ho_2Ir_2O_7$, *Lefrançois et al. Nat. Com. 2017*

Inelastic neutron scattering: magnetic excitations

INELASTIC SCATTERING: EXAMPLES OTHER THAN SPIN WAVES

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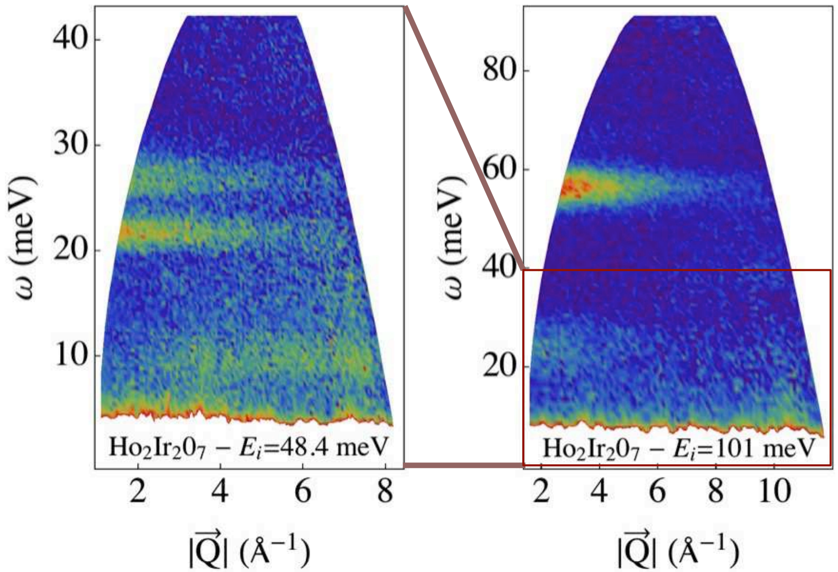
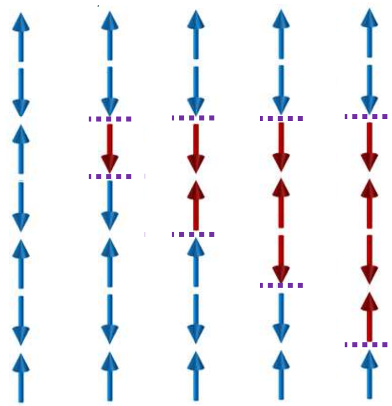


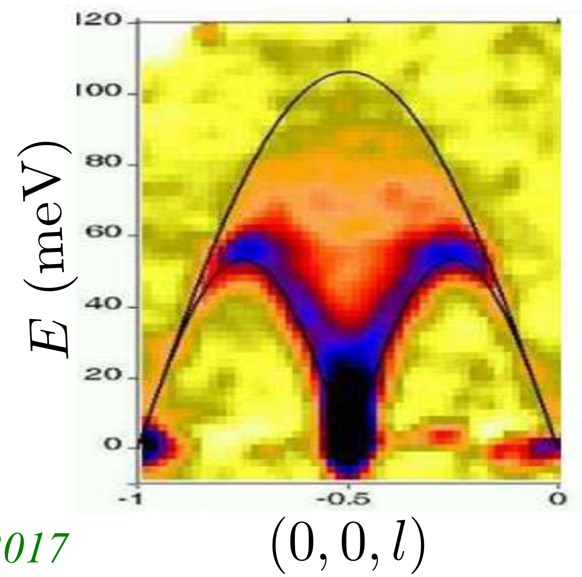
Ho^{3+} in $Ho_2Ir_2O_7$, *Lefrançois et al. Nat. Com. 2017*

Quantum excitations

Spinons (≈ domains walls)
→ Freely propagate
→ Ungapped **continuum**



Dispersion relation



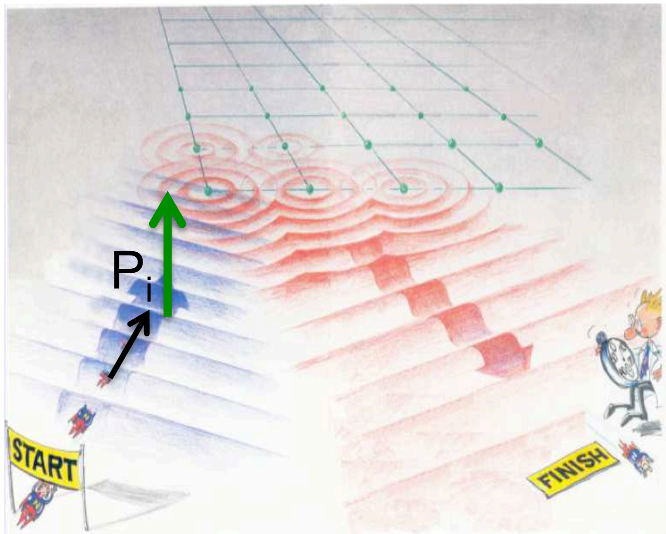
$KCuF_3$: 1D antiferromagnets with spin S=1/2, *Nagler et al. PRB 1991*

Use of Polarized neutrons

Use of Polarized neutrons

Cross section depends on the spin state of the neutron. Polarized neutron experiment uses this spin state and its change upon scattering process to obtain additional information.

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}\left(\frac{m_N}{2\pi\hbar^2}\right)^2 \sum_{\lambda,\sigma_i} \sum_{\lambda',\sigma_f} p_\lambda p_{\sigma_i} |\langle k_f \sigma_f \lambda_f | V | k_i \sigma_i \lambda_i \rangle|^2 \delta(\hbar\omega + E - E')$$



Different techniques using polarized neutrons depending on the way initial P_i and final P_f polarizations are analyzed:
-Half polarized experiments (either P_i or P_f)

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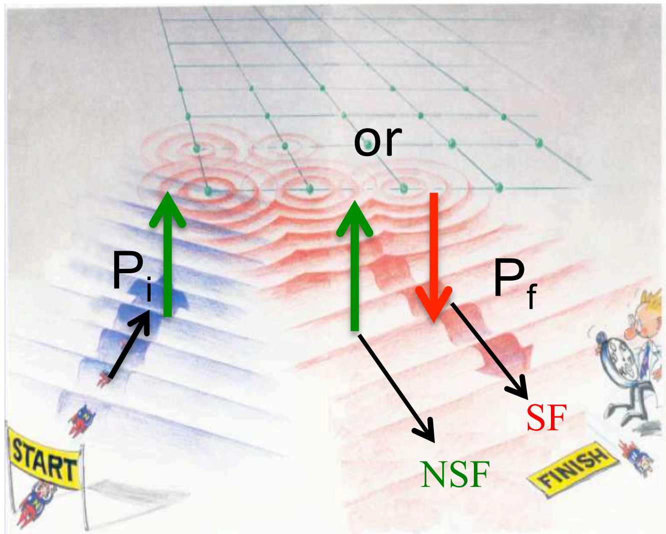

Different techniques using polarized neutrons depending on the way initial P_i and final P_f polarizations are analyzed:

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-Longitudinal polarization analysis

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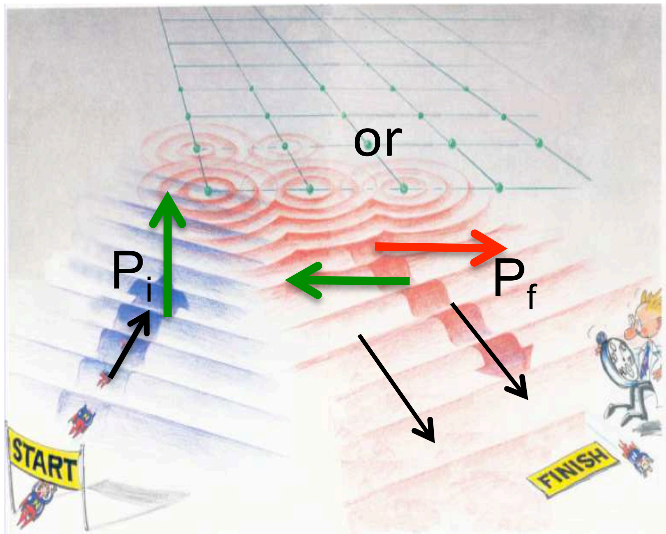


Different techniques using polarized neutrons depending on the way initial P_i and final P_f polarizations are analyzed:

-Half polarized experiments (either P_i or P_f)

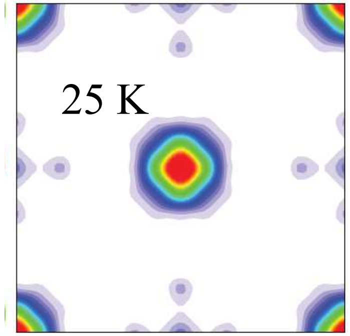
-Longitudinal polarization analysis

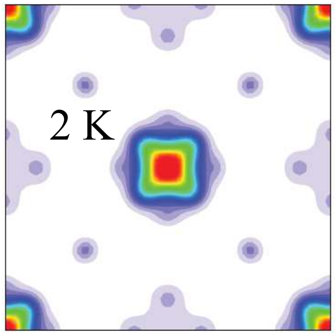
-Spherical polarization analysis

→ Used in diffraction and inelastic scattering

Use of Polarized neutrons

→ Amplifies magnetic signal
→ Measurement of magnetic form factor
→ Atomic site susceptibility tensor
→ Magnetization density map





Spin density maps in URu_2Si_2
Ressouche et al PRL 2012

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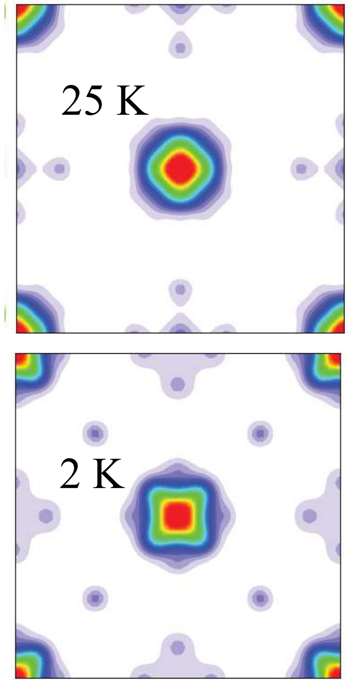
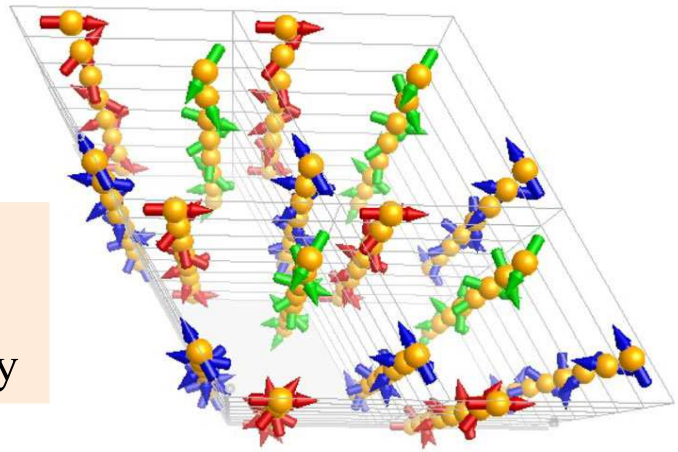
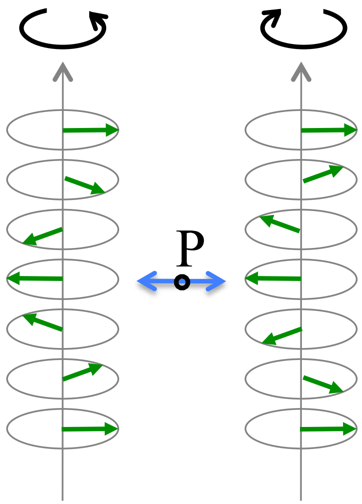


Spin density maps in URu_2Si_2
Ressouche et al PRL 2012

→ Separation magnetic/nuclear
→ Access to spin components M_y, M_z
→ Access to magnetic/nuclear chirality

Unique magnetic chirality
in $Ba_3NbFe_3Si_2O_{14}$ *Marty et al. PRL 2008*





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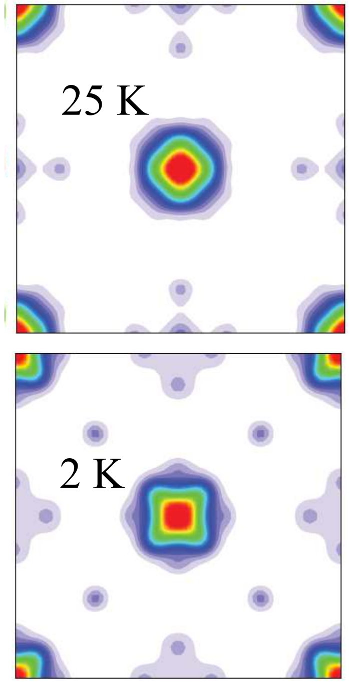

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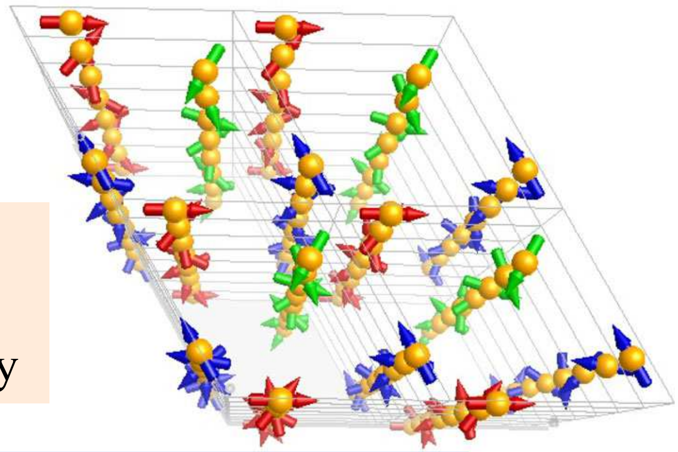
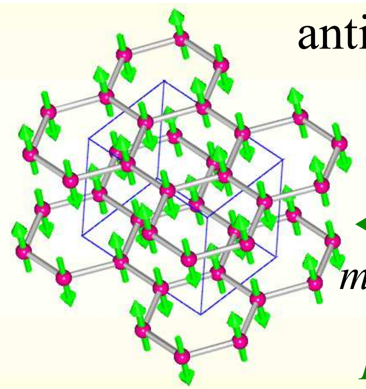
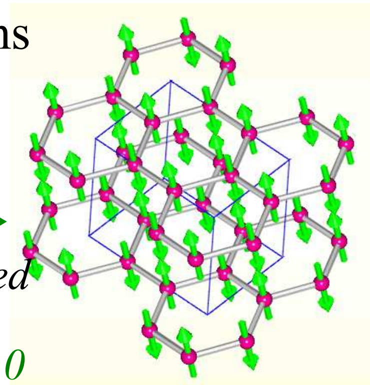


→ Refine complex magnetic structure
→ Probe magnetic domains

antiferromagnetic domains
in $MnPS_3$



magnetic moments reversed



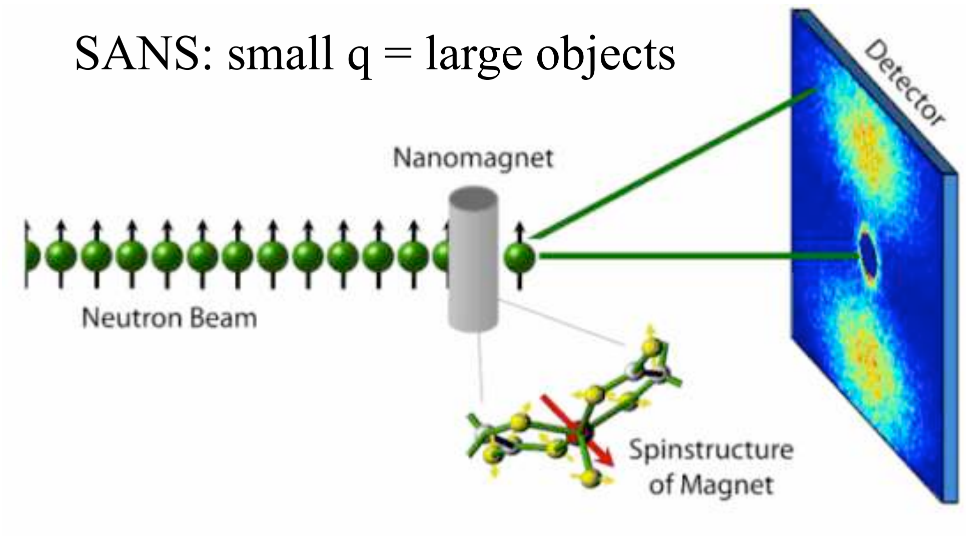
Ressouche et al PRB 2010

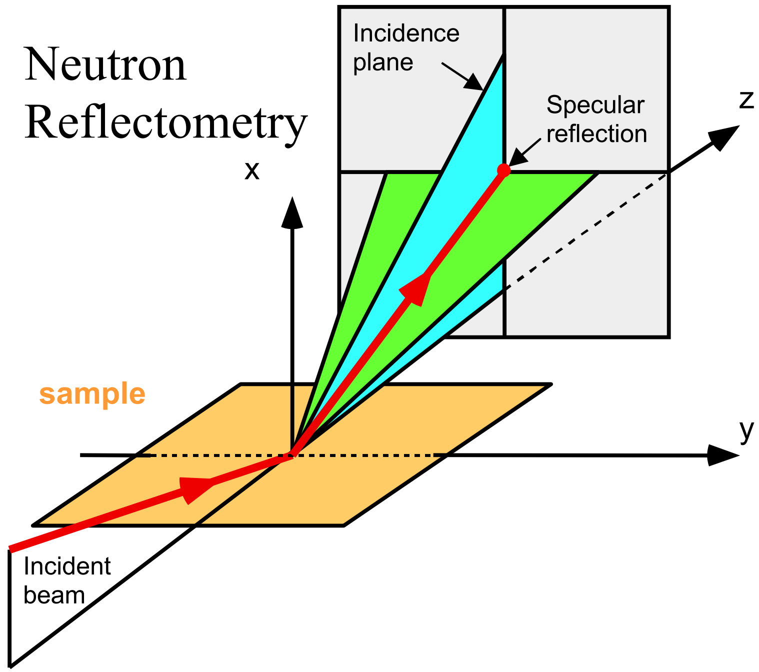
Techniques for studying magnetic nano-objects

Techniques for studying magnetic nano-objects

SMALL ANGLE SCATTERING AND REFLECTOMETRY

Techniques to probe various kinds of nanostructures
Use of polarized neutrons
Reflectometry, SANS,
combination of both (GISANS)



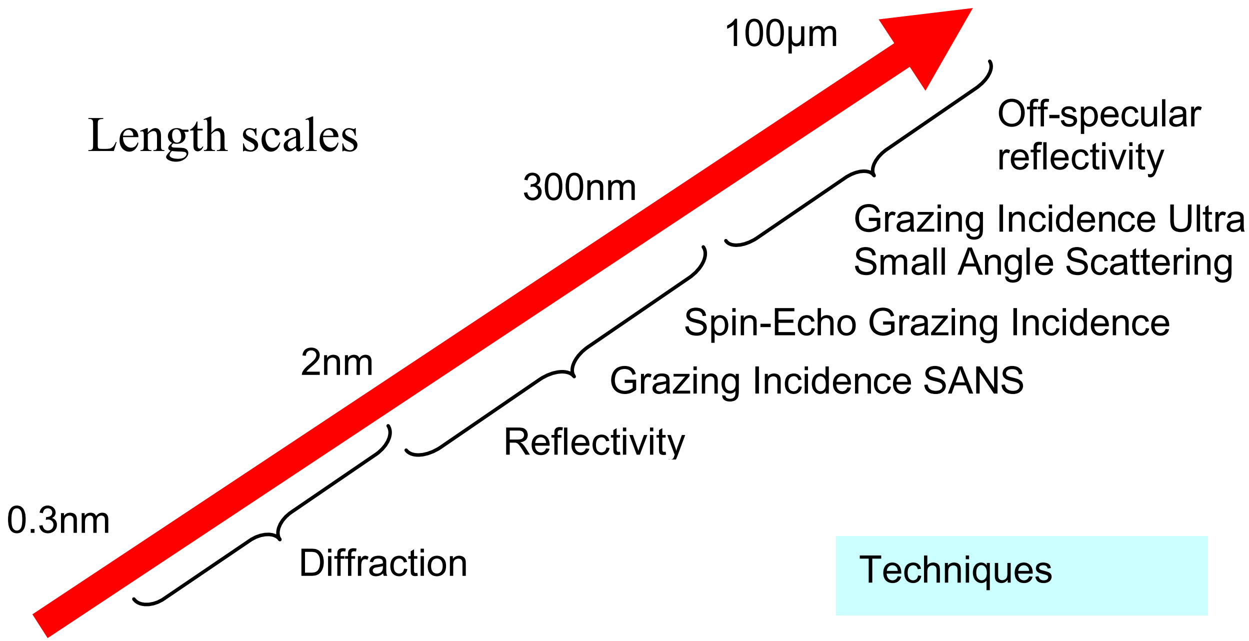


Mühlbauer et al. Rev. Mod. Phys. 2019

Techniques for studying magnetic nano-objects

SMALL ANGLE SCATTERING AND REFLECTOMETRY

Applications: long wavelength spin textures, vectorial magnetization profile of ordered or diluted magnetic nanoparticles/nanowires/domain walls and of magnetic multilayers down to the monolayer (depth and lateral structure) in absolute values.



Techniques for studying magnetic nano-objects

SMALL ANGLE SCATTERING AND REFLECTOMETRY

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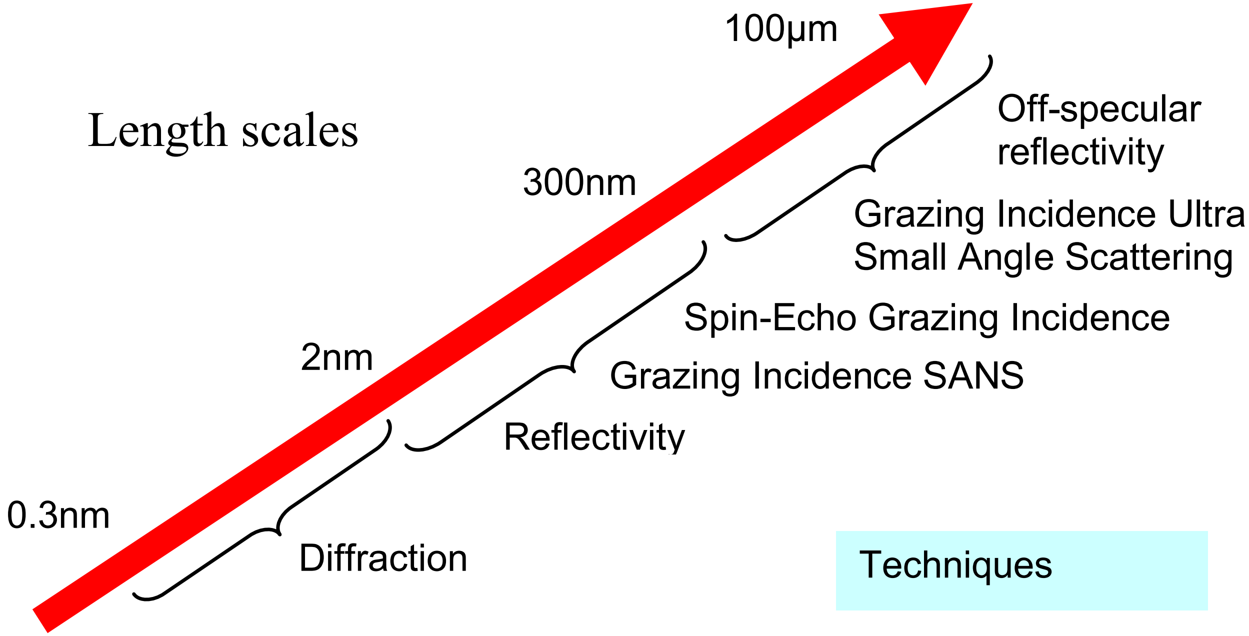
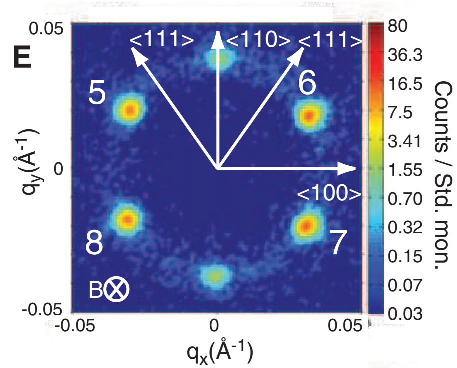
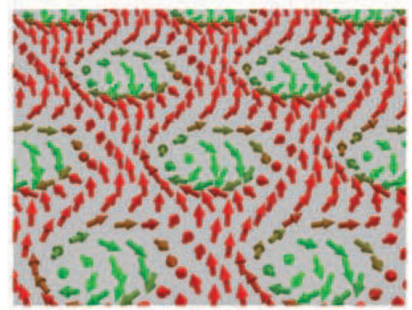
Length scales

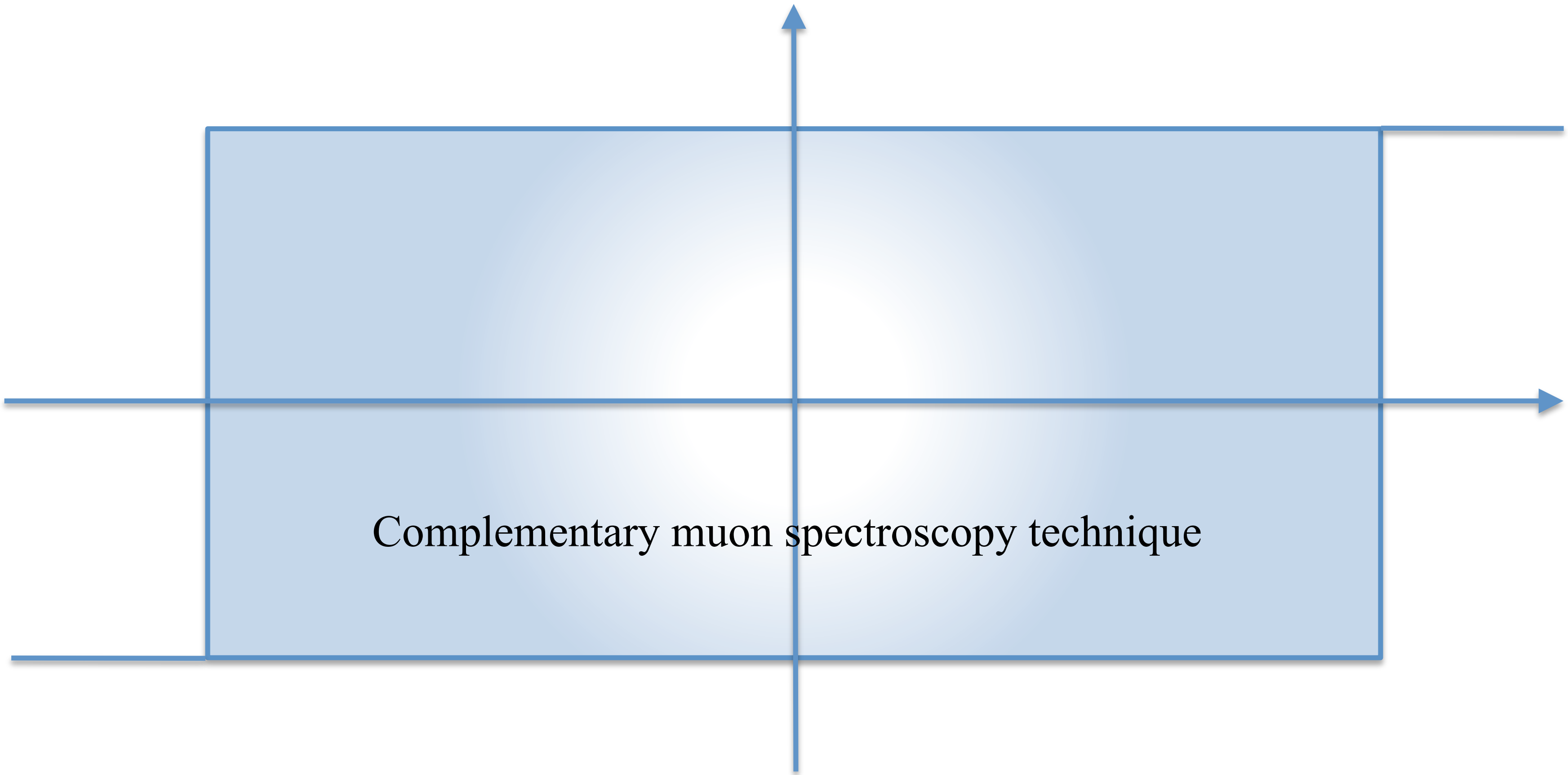


Example SANS in MnSi: ordered lattice of skyrmions





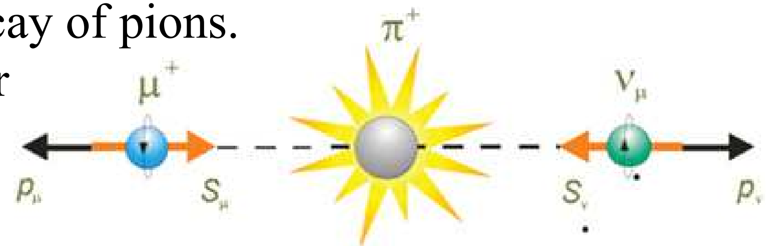
Mühlbauer et al. Science 2019


Complementary muon spectroscopy technique

Complementary muon spectroscopy technique

MUON SPIN SPECTROSCOPY (μSR=muon spin resonance/rotation/relaxation)

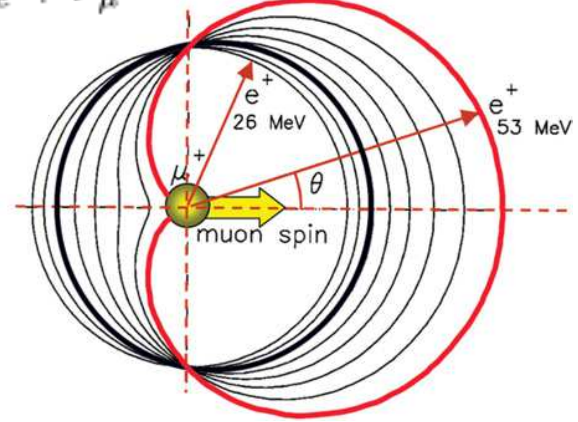
Muons are light elementary particles produced by decay of pions.
Muons have a spin ½, and remain implanted in matter until their decay = **local probe**

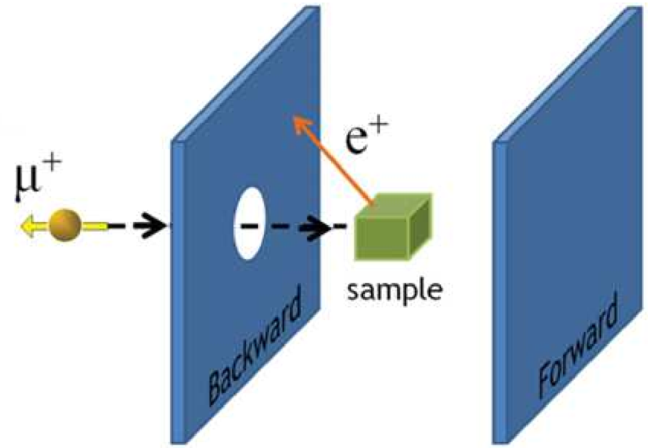


100% polarized muon beam

Muon decay: anisotropic emission of the positron recorded by forward and backward detectors, correlated to muon spin direction.

$$\mu^+ \to e^+ + \nu_e + \bar{\nu}_\mu$$



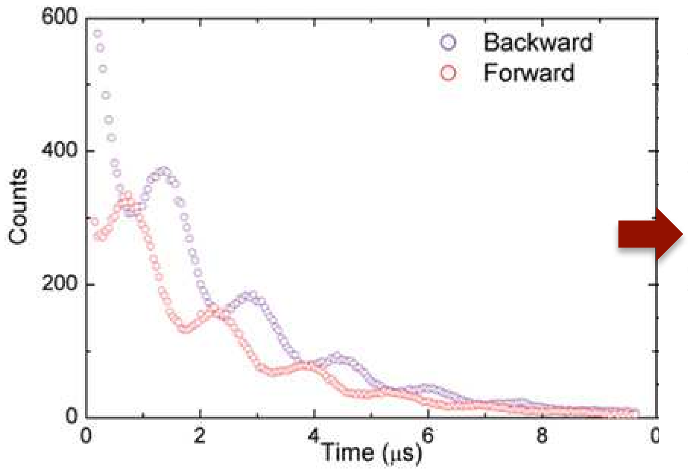


Complementary muon spectroscopy technique

MUON SPIN SPECTROSCOPY (μSR=muon spin resonance/rotation/relaxation)

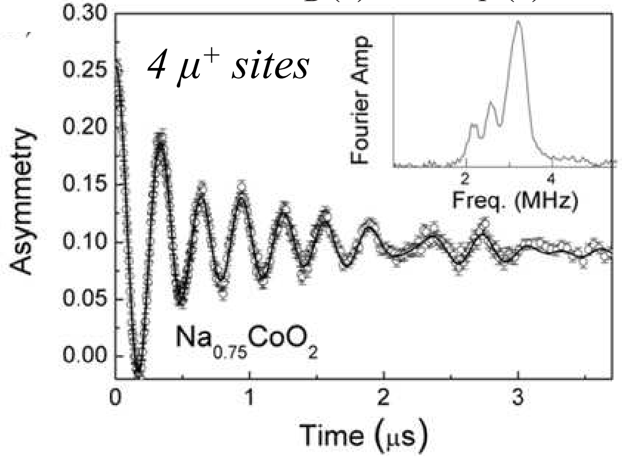
Internal fields → Larmor precession of the muon spin: oscillations on top of asymmetric decay

Number of positrons collected in the two detectors vs time

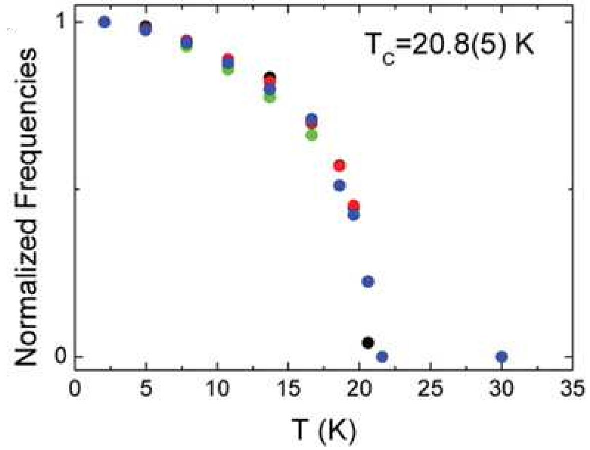


Asymmetry of the muon decay

$$a(t) = \frac{N_B(t) - \alpha N_F(t)}{N_B(t) + \alpha N_F(t)}$$



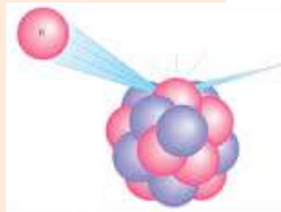
Larmor frequency vs T ∝ internal fields



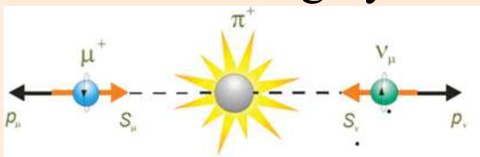
Use of μSR:
Detection of small static/dynamic internals fields (ordered moments or disordered systems) with high sensitivity ≈ 0.01 μ_B → Phase diagrams

Conclusion

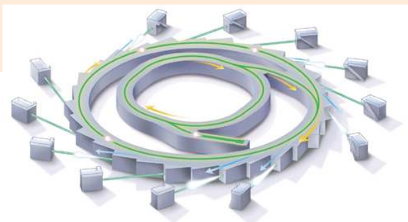
➢**Neutron scattering** = best method to determine the magnetic arrangement in bulk matter, especially for complex orders.
Also unique tool to measure the magnetic excitations especially at low energies.
Drawbacks: needs of big samples → This can be improved with novel sources.
Formalism well established.



➢Internal fields in matter can be measured with alternative highly sensitive techniques such as NMR, Mössbauer, **muon spectroscopy**.



➢**X-ray scattering** complementary tool.
Magnetic scattering rather weak effect (5 orders of magnitude smaller than non-magnetic scattering) compensated by very high brilliance of synchrotron sources and use of resonant techniques (chemically selective)→small samples can be used. Huge progress in RIXS techniques. However still unable to reach low energies accessible by neutron scattering.



Further reading

- Material borrowed from presentations of B. Grenier, L. Chaix, N. Qureshi, E. Ressouche, thanks to them!

- “**Neutrons and magnetism**” JDN20, *collection SFN (2014), EDP Sciences, editors V. Simonet, B. Canals, J. Robert, S. Petit, H. Mutka*,
  in particular lectures from M. Enderle, E. Ressouche, S. Raymond, F. Ott, F. Bert
  free access https://www.neutron-sciences.org/articles/sfn/abs/2014/01/contents/contents.html

- “**Introduction to the theory of Thermal Neutron Scattering**” by G. L. Squires, *Cambridge University Press (1978)*

- “**Theory of Neutron scattering from condensed matter**” by S. W. Lovesey, Oxford Clarendon Press (1984)

- Any questions: virginie.simonet@neel.cnrs.fr