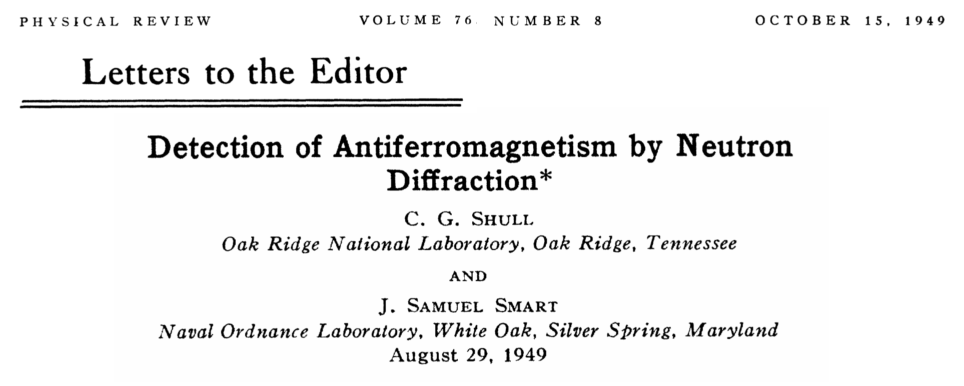
Concepts of Neutron Scattering

Andrew Boothroyd
University of Oxford

- Basic features of neutron scattering
- Neutron diffraction
- Neutron spectroscopy
- Correlations
- Polarized neutrons

6th PSI Summer School on Condensed Matter Research
Zuoz, Aug 18–25, 2007

Nearly 60 years of magnetic diffraction!



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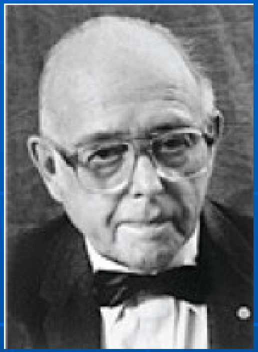
Letters to the Editor

Detection of Antiferromagnetism by Neutron Diffraction*

C. G. Shull
Oak Ridge National Laboratory, Oak Ridge, Tennessee
AND
J. Samuel Smart
Naval Ordnance Laboratory, White Oak, Silver Spring, Maryland

August 29, 1949



C.G. Shull
(1915–2001)

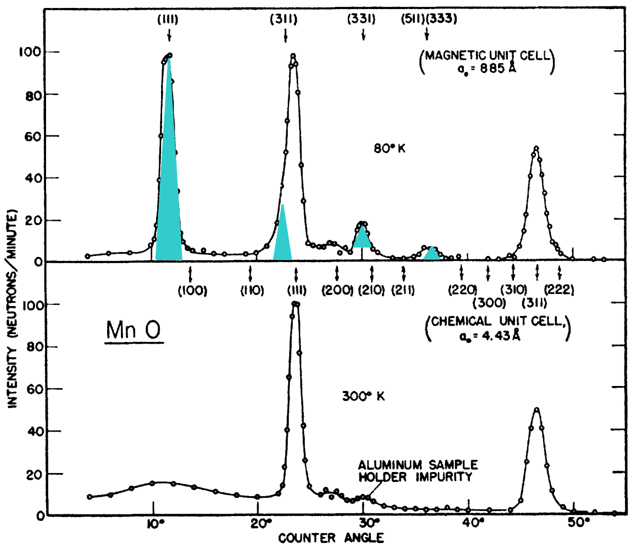
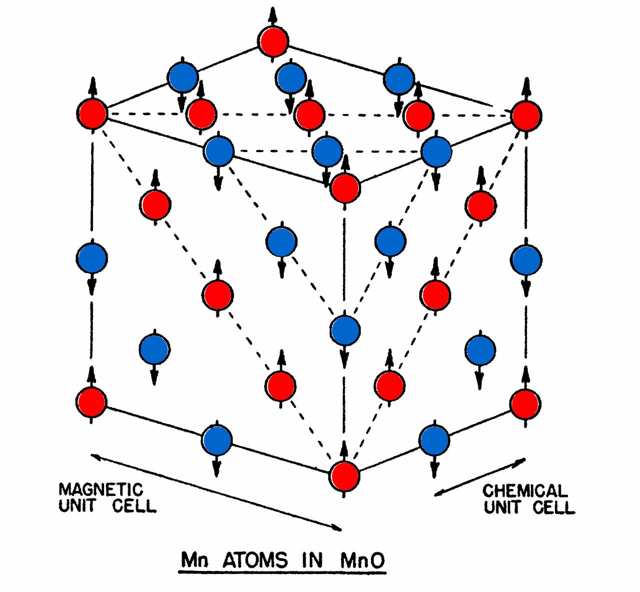


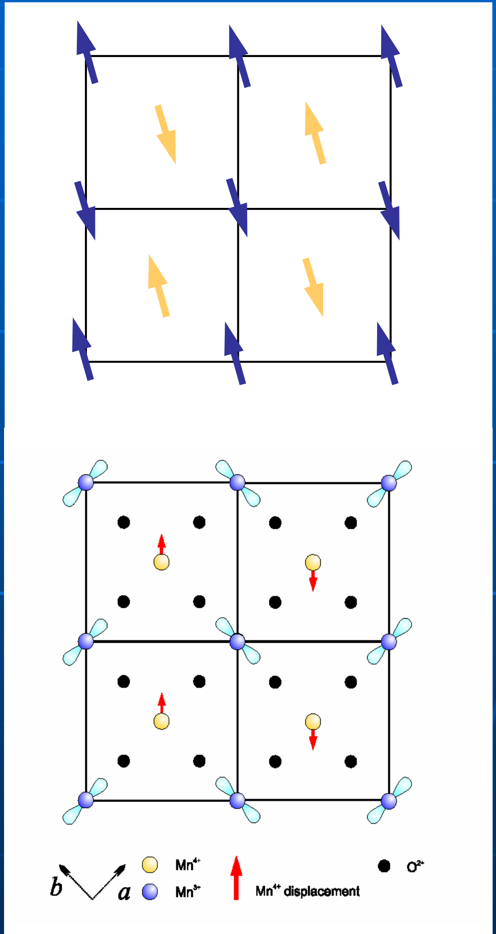
FIG. 1. Neutron diffraction patterns for MnO at room temperature and at 80°K.



Correlations in interacting electron systems

	Neutrons?
◇ magnetic	✔
◇ charge *	✘
◇ orbital *	✘
◇ lattice distortions	✔

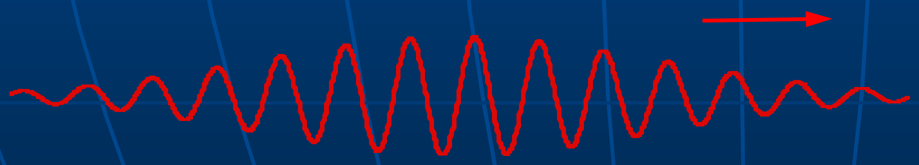
*BUT neutrons can detect charge and orbital correlations indirectly through their effect on the lattice



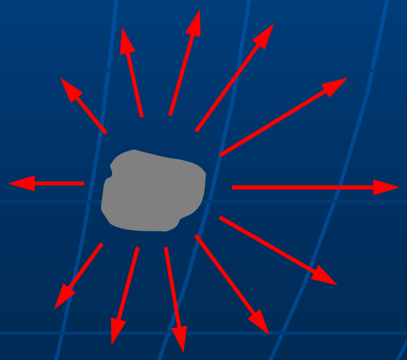
$La_{1/2}Sr_{3/2}MnO_4$

Scattering 'nuts and bolts'

- Neutrons, photons, electrons, atoms
- Measure distribution of radiation scattered from a sample
- Interaction potential determines property measured
- Radiation must be coherent to measure correlations



Neutrons are waves and particles



Scattering techniques probe correlations

1 Born approximation:

2 Coherence + superposition:

Detected amplitude:

$$\Phi = \ \ldots \ \phi_1 + \phi_2 \ldots$$

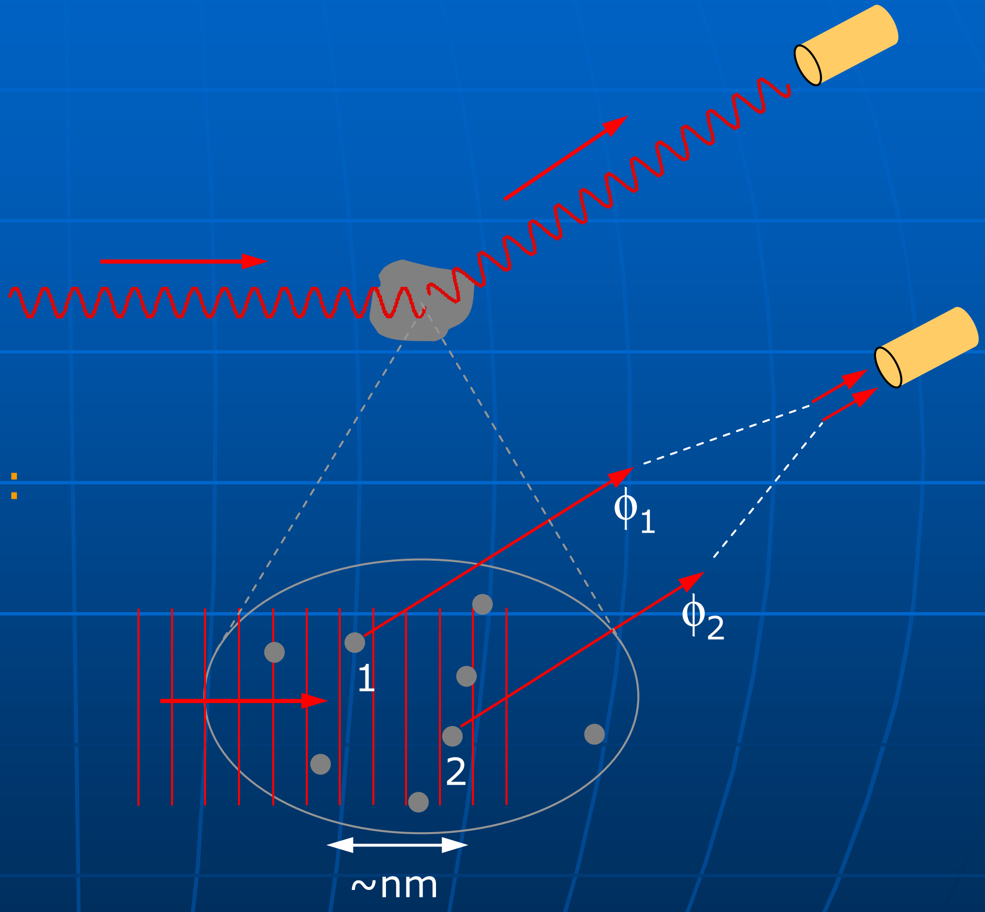
Detected intensity:

$$I = \ |\ldots \ \phi_1 + \phi_2 \ \ldots|^2$$

$$= \ \ldots \ |\phi_1|^2 + |\phi_2|^2 + \underline{\phi_1^* \ \phi_2 + \phi_2^* \ \phi_1} \ \ldots$$

depends on relative positions of 1 and 2

→ correlations



Properties of the Neutron

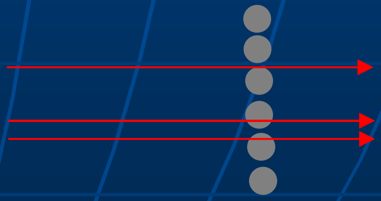
- ◇ Mass = 1.675×10^{-27} kg
- ◇ Charge = 0
- ◇ Mean lifetime ≈ 15 min
- ◇ Spin = ½
- ◇ Magnetic moment = 1.91 μ_N (~$0.001\mu_B$)

Neutrons interact with

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

In both cases, the interaction is very weak

E.g. consider close-packed layer of atoms:



Probability of scattering ~ 1 in 10^8
→ mean free path ~ 1 cm

Weak interaction potential

Advantages:

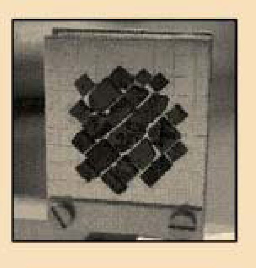
1. Neutrons probe the bulk (~1 cm)
2. Neutrons do not damage the sample
3. Born approximation holds
   → scattering depends on Fourier transform of interaction potential
   → system responds linearly
   → measure equilibrium properties
4. Intensity can be calibrated
5. Theory is quantitative

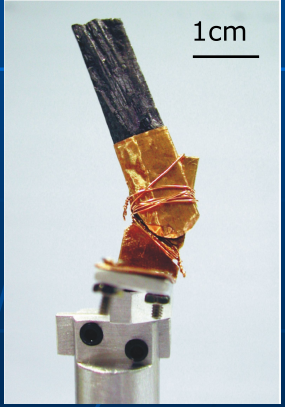
Disadvantages:

1. Sample size is an important consideration

single crystals: ~1 mm^3 (diffraction) ~1 cm^3 (spectroscopy)

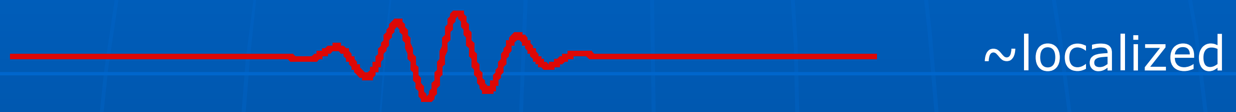
powder samples: ~1 g (diffraction) ~10 g (spectroscopy)

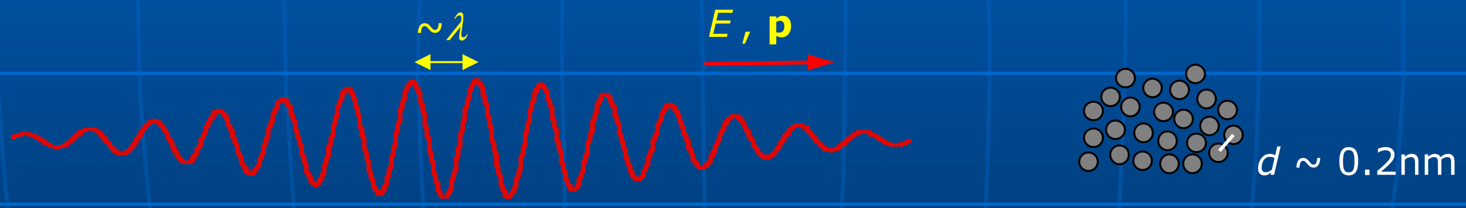




Neutron energy and wavelength

Neutrons are particles and waves:





de Broglie: $p = h/\lambda = \hbar k$ $(k = 2\pi/\lambda)$

$$E = h^2/(2m\lambda^2) = \hbar^2k^2/2m$$

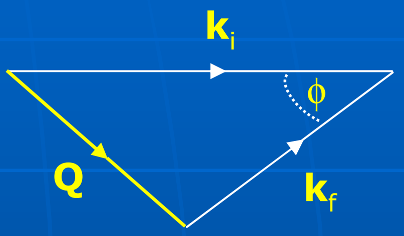
→ E = 25 meV corresponds to v = 2,200 ms^{-1} and λ = 0.18 nm

comparable to energy and length scales of static and dynamic correlations in condensed matter

Neutron kinematics

- momentum transfer

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



scattering triangle

- energy transfer

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

- a scattering event is characterised by $(\mathbf{Q}, \omega)$

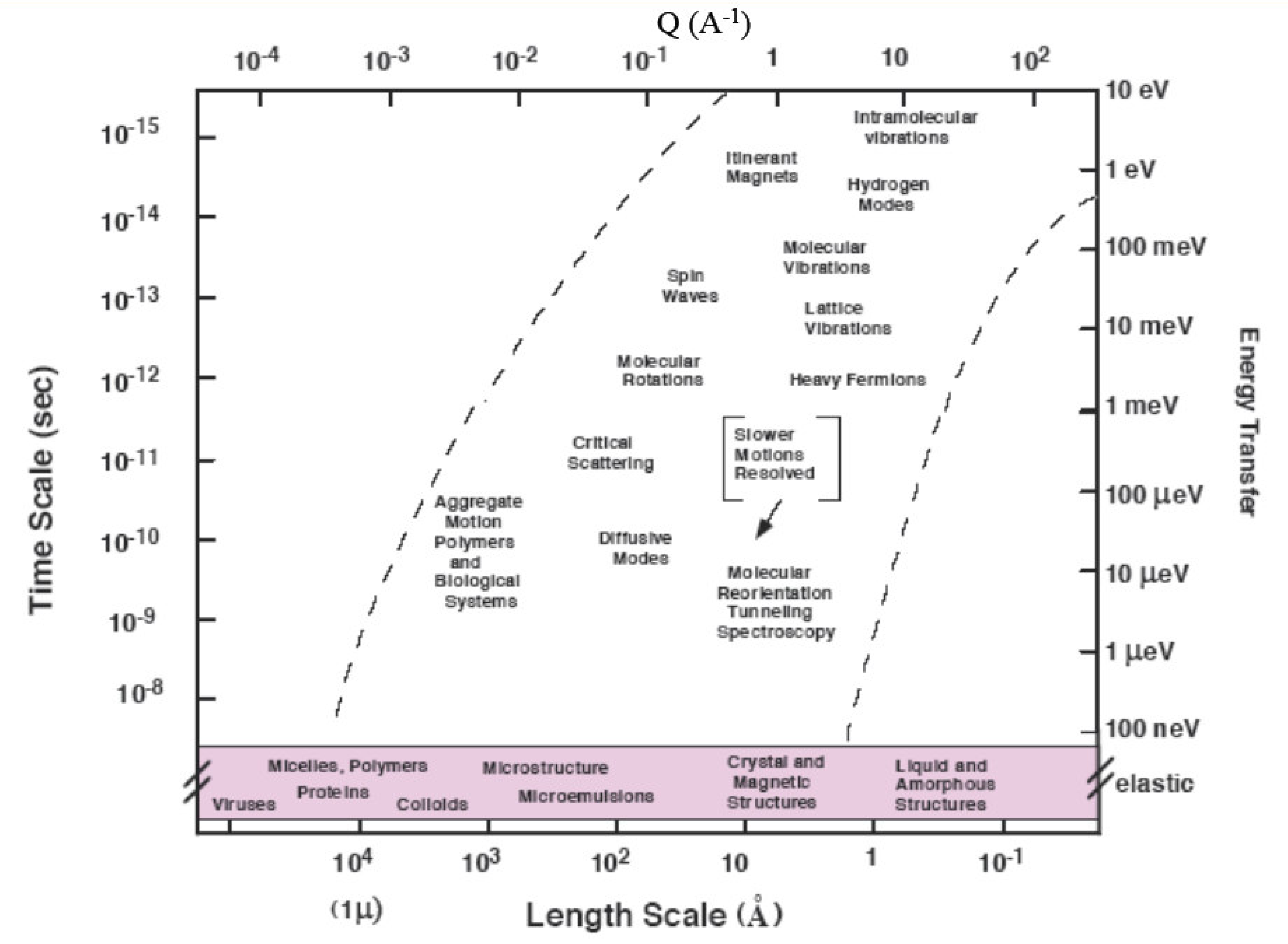
  Elastic scattering (diffraction): $\hbar\omega = 0$

  Inelastic scattering (spectroscopy): $\hbar\omega \neq 0$

- $\mathbf{Q}$ and $\hbar\omega$ can be chosen independently (within limits set by $0 < \phi < 180$ deg)

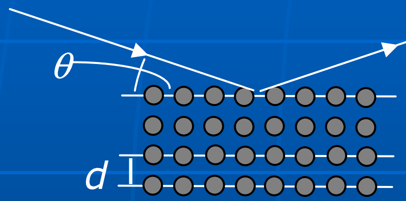
- Fourier transform conjugate variables: $Q \sim 2\pi/(\text{length})$ $\hbar\omega = 2\pi\hbar/(\text{time})$

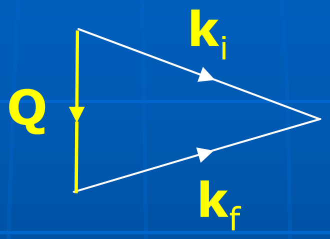
Correlations in space and time



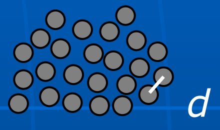
Diffraction

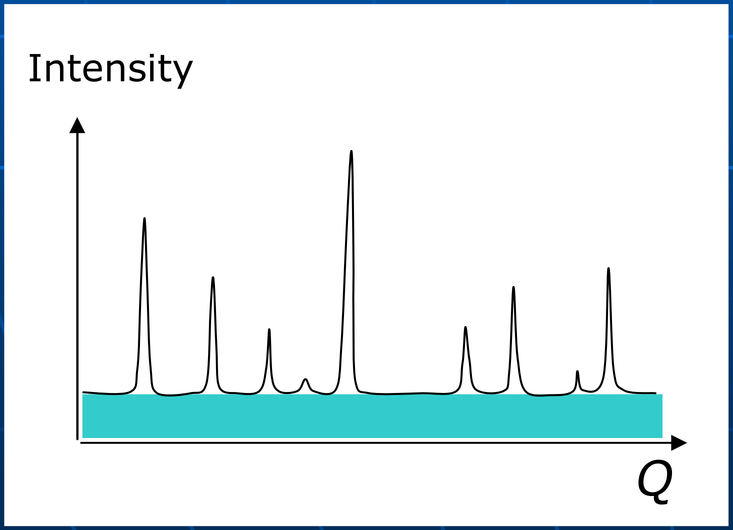
Diffraction from
a crystalline material



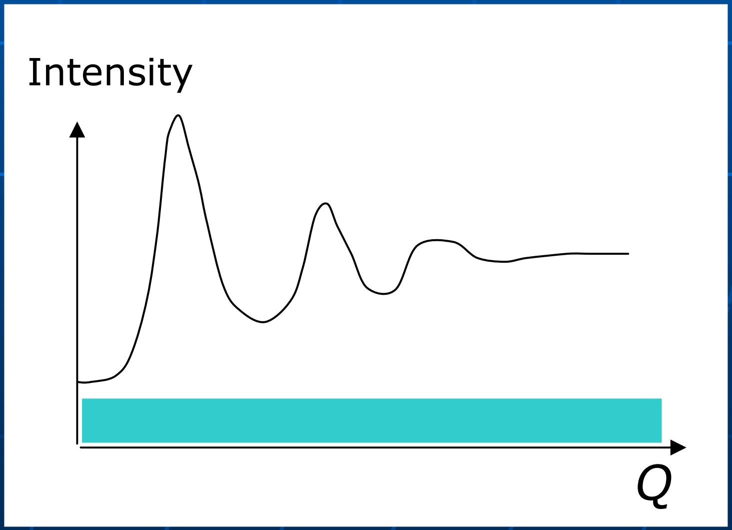


Diffraction from
a disordered material





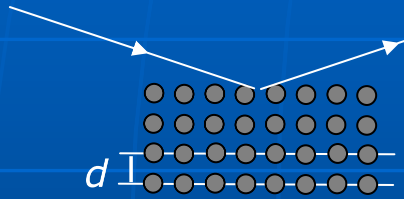
Diffraction peaks when $n\lambda = 2d \sin\theta$
(Bragg's Law)

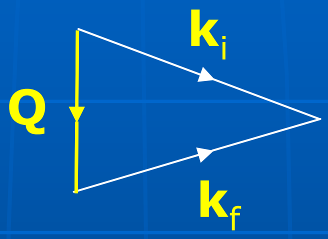


First maximum when $Q \approx 2\pi/d$

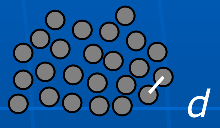
Diffraction

Diffraction from
a crystalline material

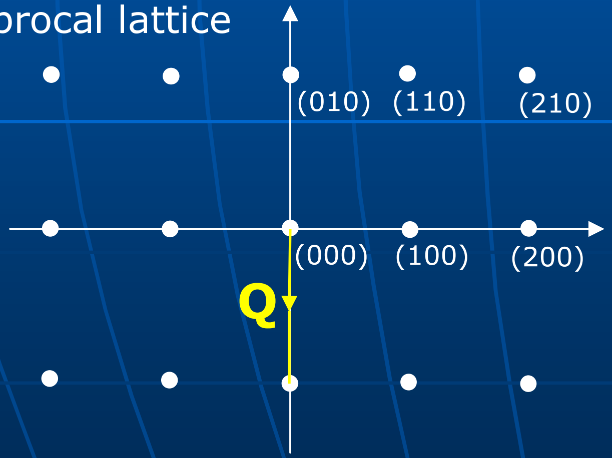


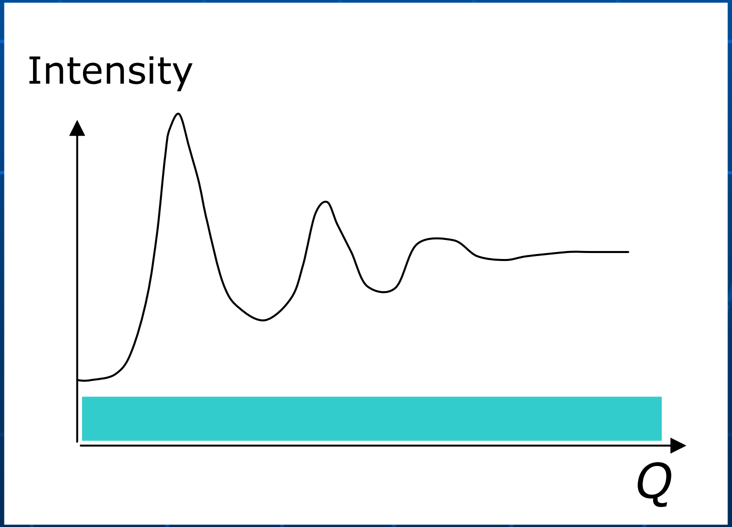


Diffraction from
a disordered material



Reciprocal lattice



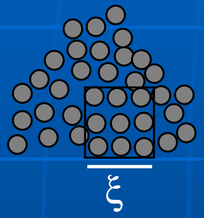


Bragg peaks when **Q** = **G**
(**G** = reciprocal lattice vector)

First maximum when $Q \approx 2\pi/d$

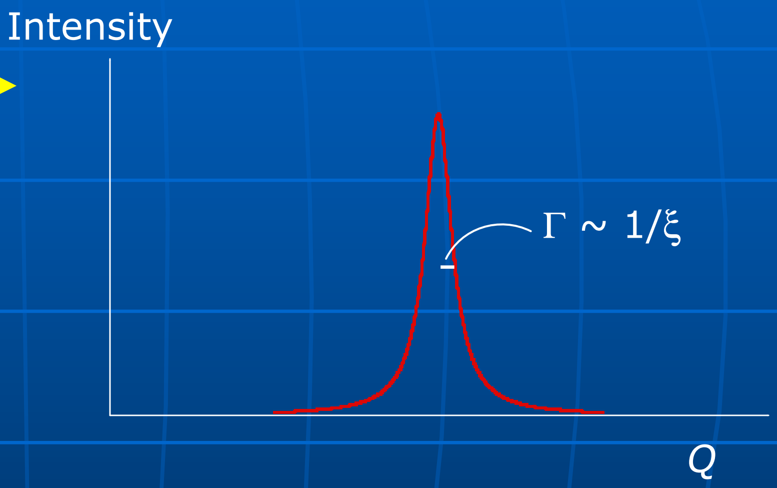
Short-range order and diffuse scattering

Short-range order



ξ = correlation length

Diffuse scattering



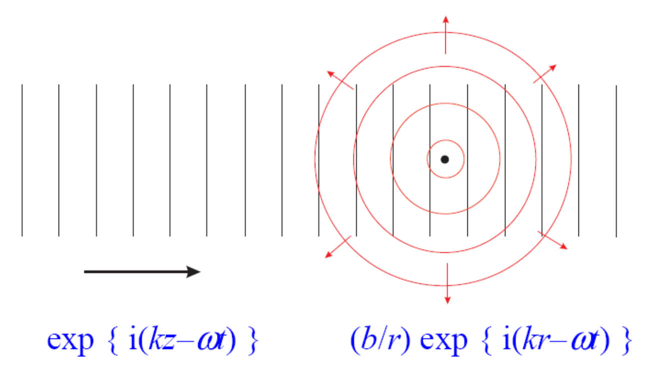
Γ = half width at half maximum

If the system is correlated over a range $\Delta x \sim \xi$,

the scattering features are broadened by $\Delta Q \sim 1/\xi$

Nuclear neutron diffraction

Nuclear interaction (pseudo-)potential



$$V_N(\mathbf{r}) = \frac{2\pi\hbar^2}{m_n} b\,\delta(\mathbf{r})$$

- ◇ very short range
- ◇ scalar potential → isotropic scattering
- ◇ b = scattering amplitude/length (~10^{-12} cm)
- ◇ diffraction intensity:

$$S(\mathbf{Q}) = |\sum_j b_j \exp(i\mathbf{Q} \cdot \mathbf{r}_j)|^2$$ (sum over all nuclei in sample)

- ◇ Rigid crystal:

$$S(\mathbf{Q}) = N \frac{(2\pi)^3}{v_0} \sum_{\mathbf{G}} |F(\mathbf{G})|^2 \delta(\mathbf{Q} - \mathbf{G})$$ (sum over all reciprocal lattice vectors **G**)

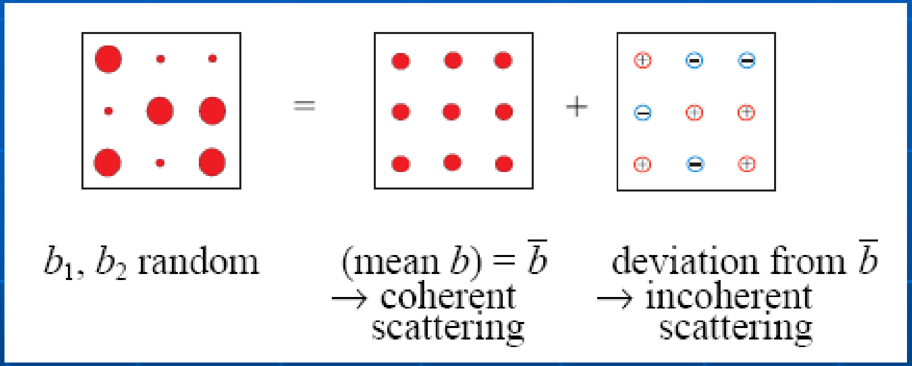
where $F(\mathbf{G}) = \sum_j b_j \exp(i\mathbf{G} \cdot \mathbf{r}_j)$ (sum over nuclei in unit cell)

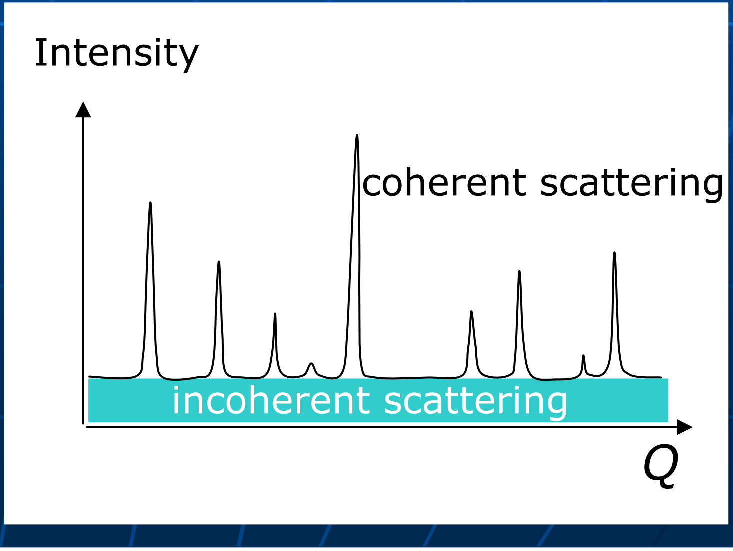
is the structure factor,
v_0 is the volume of the unit cell and N is the no. unit cells in crystal

Coherent and incoherent nuclear scattering

b varies with isotope and with nuclear spin orientation

e.g. sample with two isotopes with scattering lengths b_1(●) and b_2(•)





Coherent scattering: replace b_j by $\overline{b_j}$

Incoherent scattering: additional 'flat background'

$$S_{\text{inc}}(\mathbf{Q}) = \sum_j \frac{(\sigma_{\text{inc}})_j}{4\pi}$$

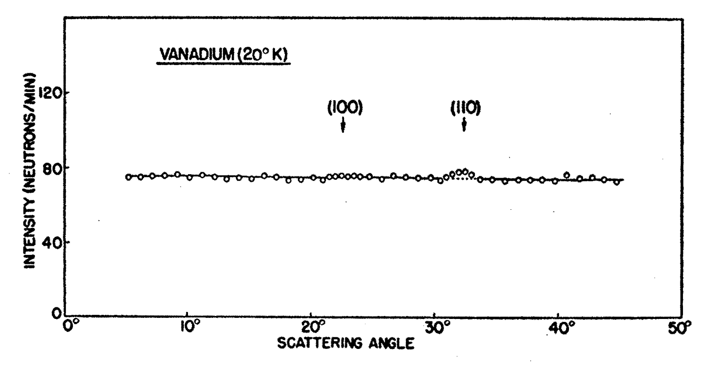
Coherent and incoherent nuclear scattering

Values of $\bar{b}$ and σ_{inc} can be found in tables,
e.g. http://www.ncnr.nist.gov/resources/n-lengths/

Examples:

Nucleus	$\bar{b}$ ($\times 10^{-12}$ cm)	σ_{inc} ($\times 10^{-24}$ cm^2)
^{1}H (proton)	–0.374	79.9
^{2}H (deuteron)	0.667	2.0
O	0.581	0.0
V	–0.040	4.9
La	0.827	1.6
U	0.842	0.0

Strong incoherent scattering from vanadium is used for calibration and normalisation of detectors



Shull & Wilkinson 1953

Magnetic neutron diffraction

Magnetic interaction potential

$V_M(\mathbf{r}) = -\boldsymbol{\mu}_n \,.\, \mathbf{B}(\mathbf{r})$ [$\boldsymbol{\mu}_n$ = neutron magnetic moment]

- ◇ $\mathbf{B}(\mathbf{r})$ originates from distribution of electron spin and orbital currents
- ◇ Vector interaction
- ◇ Anisotropic scattering
- ◇ Depends on orientation of neutron spin $\mathbf{s}_n$: $\boldsymbol{\mu}_n = -2\gamma\mu_N\mathbf{s}_n$ $(\gamma = 1.913)$

→ polarised neutrons can be used

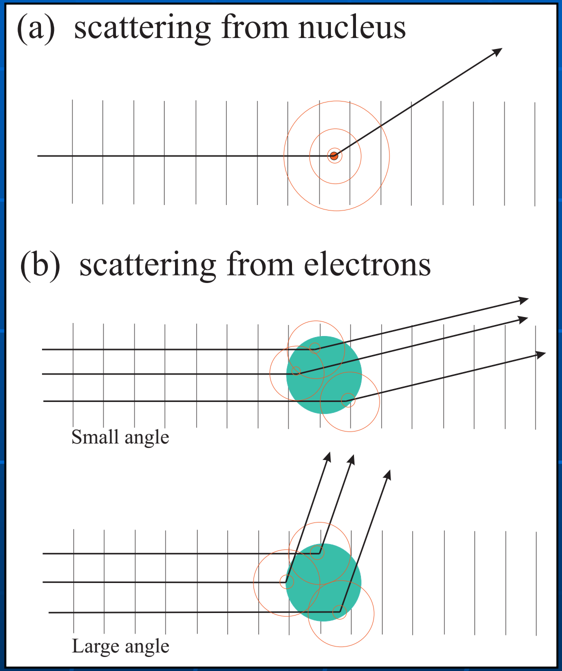
to probe different magnetic components

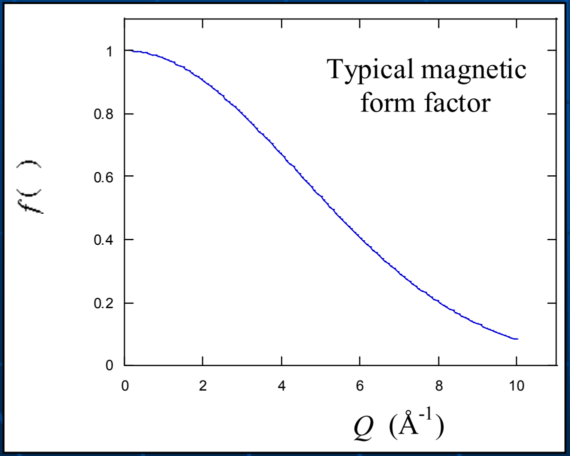
Magnetic form factor

$$V_{\mathrm{M}} = -\boldsymbol{\mu}_{\mathrm{n}} \;.\; \mathbf{B}(\mathbf{r})$$

B(**r**) derives from electron spin and orbital motion, which is distributed over volume of atom

Scattering decreases with increasing Q due to intra-atomic interference





Fourier transform of magnetic potential

Magnetic scattering probes magnetic moments perpendicular to **Q**

Magnetic scattering depends on Fourier transform of $V_M(\mathbf{r})$:

$$V_M(\mathbf{Q}) = -\boldsymbol{\mu}_n \,.\, \mathbf{B}(\mathbf{Q})$$

James Clerk Maxwell
(1831 – 1879)

Maxwell's eq.

$$\nabla .\, \mathbf{B}(\mathbf{r}) = 0$$

$$\xrightarrow{\text{F.T.}} \quad i\mathbf{Q} \,.\, \mathbf{B}(\mathbf{Q}) = 0$$

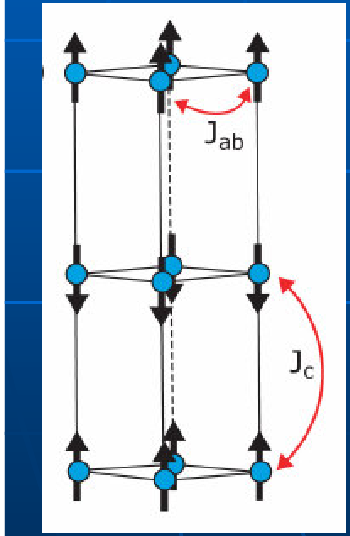
$$\longrightarrow \quad \mathbf{B}(\mathbf{Q}) \text{ is perpendicular to } \mathbf{Q}$$

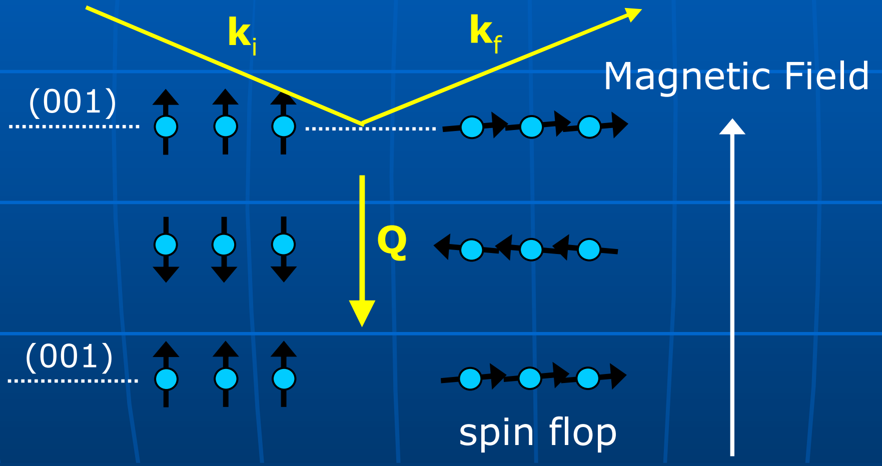
$\longrightarrow$ Neutrons scatter from $m_\perp$, the component of the (atomic) magnetic moment perpendicular to **Q**

Diffraction from moments perpendicular to **Q**

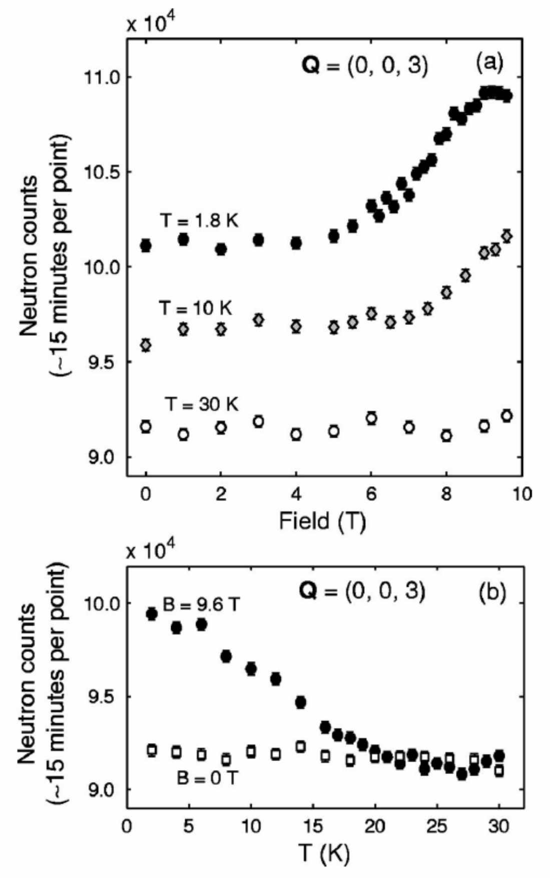
Example: Metamagnetic transition in Na_xCoO_2

(Lucy Helme, ATB, et al., 2006)



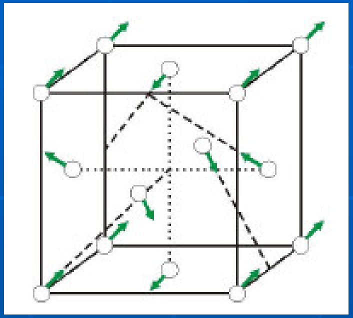


H = 0	$H = H_{SF}$
$I_{(00L)} = 0$	$I_{(00L)} > 0$



Magnetic diffraction intensity

Diffraction of unpolarized neutrons from general magnetic structure (dipole approx.):



$$S_\mathrm{M}(\mathbf{Q}) \;= C \sum_{\mathbf{G}_\mathrm{M}} |\mathbf{F}_\mathrm{M}(\mathbf{G})|^2 \,\delta(\mathbf{Q} - \mathbf{G}_\mathrm{M})$$ (sum over all magnetic reciprocal lattice vectors $\mathbf{G_M}$)

where

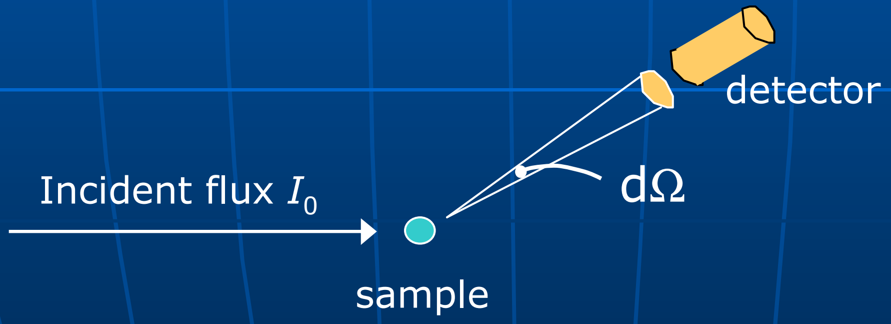
$$\mathbf{F}_\mathrm{M}(\mathbf{G}) = \sum_j f_j(\mathbf{Q})\, \mathbf{m}_{\perp j} \exp\, (i\mathbf{G} \,.\, \mathbf{r}_j)$$ (sum over ordered moments in magnetic unit cell)

is the magnetic structure factor

Inelastic scattering cross section

double differential cross section

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E_\mathrm{f}} = \frac{\begin{pmatrix}\text{no. neutrons scattered per sec. into solid angle } \mathrm{d}\Omega \\ \text{with final energy between } E_\mathrm{f} \text{ and } E_\mathrm{f} + \mathrm{d}E_\mathrm{f}\end{pmatrix}}{I_0 \times \mathrm{d}\Omega \times \mathrm{d}E_\mathrm{f}}$$



Detector has energy analysis capability

Inelastic scattering cross section

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{\begin{pmatrix}\text{no. neutrons scattered per sec. into solid angle } d\Omega \\ \text{with final energy between } E_f \text{ and } E_f + dE_f\end{pmatrix}}{I_0 \times d\Omega \times dE_f}$$

Numerator depends implicitly on 5 factors:

1 $d\Omega$
2 dE_f
3 $v_f = \hbar k_f/m$ — speed of scattered neutrons
4 ρ_i — density of incident neutrons
5 $S(\mathbf{Q},\omega)$ — transition probability for process in which system changes its energy by $\hbar\omega$ and momentum $\hbar\mathbf{Q}$ while scattering a neutron

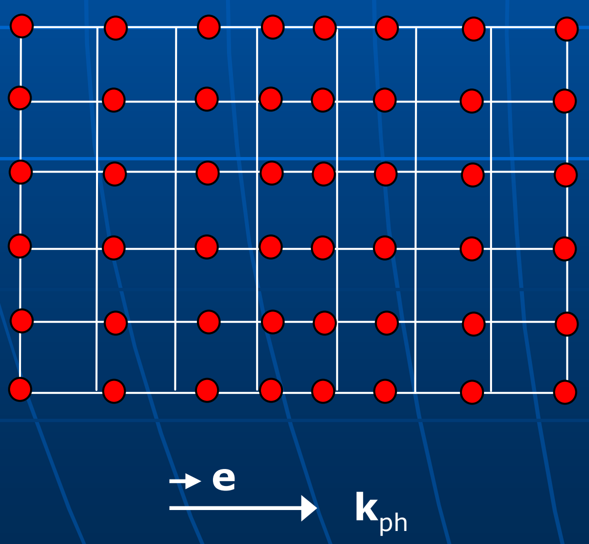
In denominator, $I_0 = \rho_i v_i = \rho_i \hbar k_i/m$

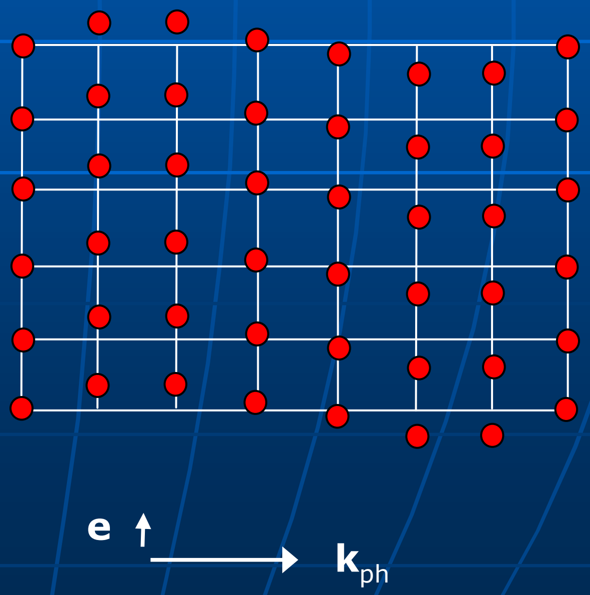
$$\longrightarrow \quad \boxed{\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i} S(\mathbf{Q},\omega)}$$

Lattice vibrations

Normal mode — all atoms vibrate at same frequency

Phonon — quantum $\hbar\omega_{ph}$ of lattice vibrational energy

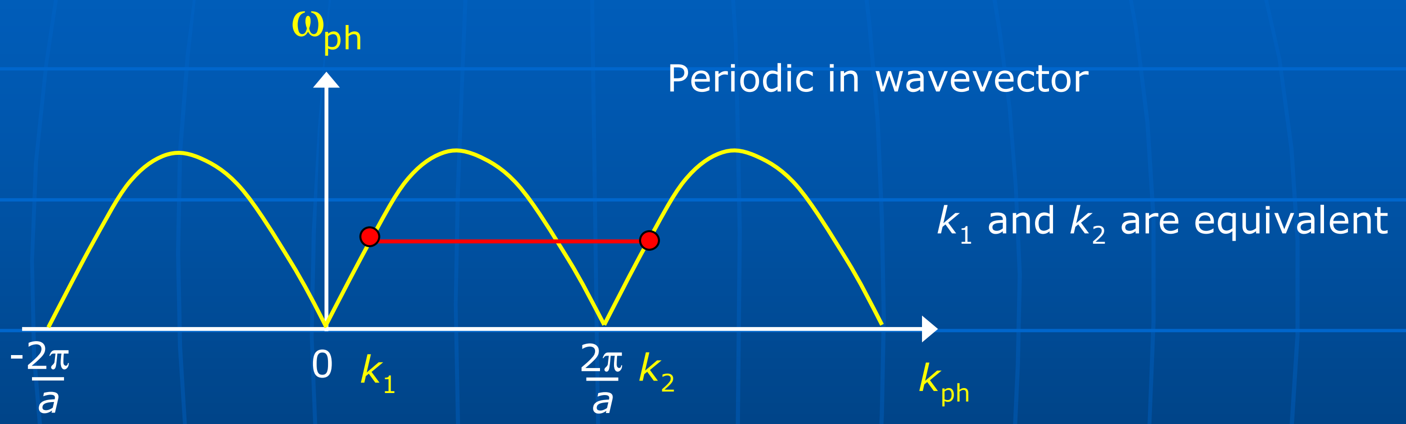

Longitudinal mode


Transverse mode

e = polarization

Scattering from lattice vibrations

◇ Phonon dispersion curve



◇ Observe peaks in neutron scattering when

$\hbar\omega = \pm\hbar\omega_{ph}$

$\hbar\mathbf{Q} = \hbar(\mathbf{k}_{ph} \pm \mathbf{G})$ (**G** is a reciprocal lattice vector; $\mathbf{k}_{ph}$ is in 1st Brillouin zone)

◇ Intensity ~ $b^2(\mathbf{Q}.\mathbf{e})^2$ Intensity increases with Q

Principle of Detailed Balance

◇ neutron energy loss and energy gain processes

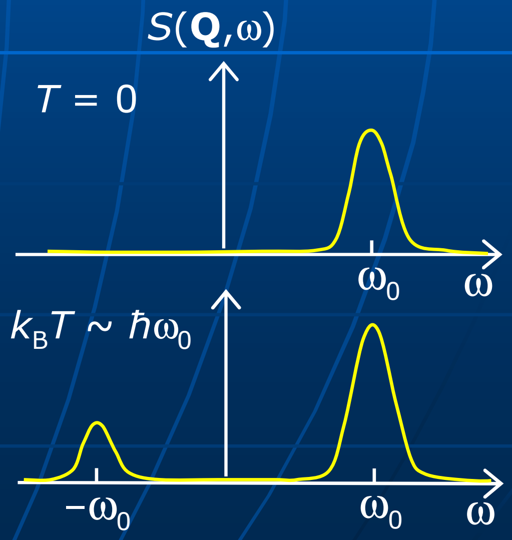
‘neutron energy loss’ ‘neutron energy gain’

◇ For any neutron scattering process

$$S(\mathbf{Q}, -\omega) = \exp(-\hbar\omega/k_B T)\, S(\mathbf{Q}, \omega)$$

neutron energy gain neutron energy loss

Principle of Detailed Balance



Expressions for $S(\mathbf{Q}, \omega)$ — 1

1 Transition probability

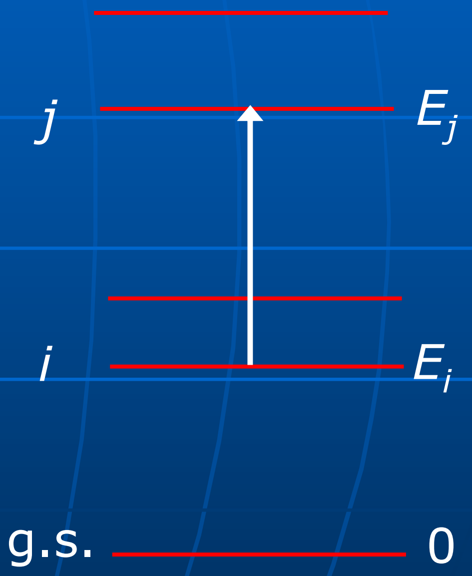
$$S(\mathbf{Q},\omega) = \sum_i p_i \sum_j |M_{ij}|^2 \delta(\hbar\omega - E_j + E_i)$$

where

P_i = thermal occupancy of initial state

M_{ij} = transition matrix element

$\delta(\hbar\omega - E_j + E_i)$ represents conservation of energy

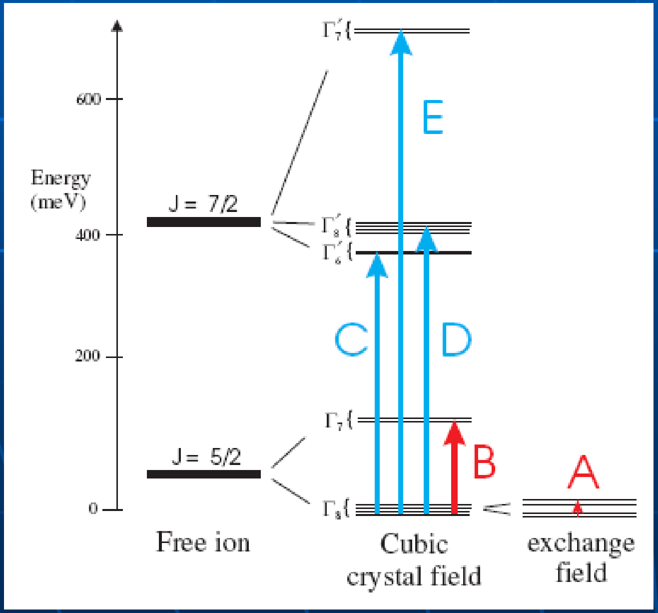


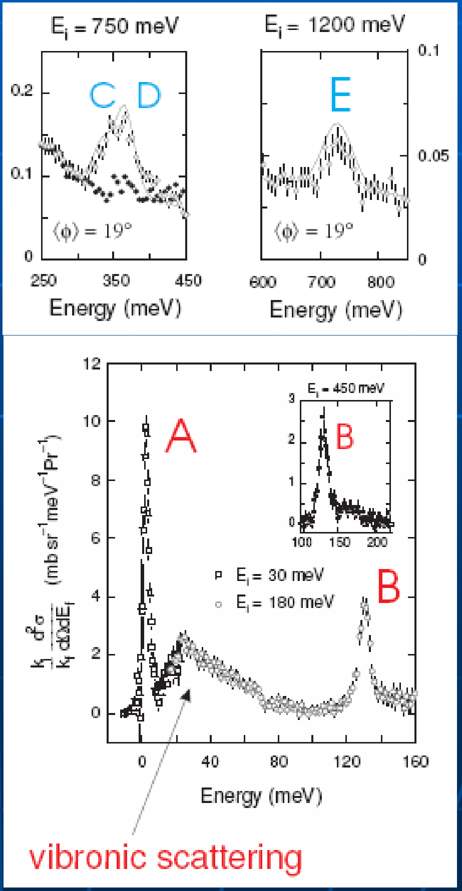
Local magnetic excitations

Example: local magnetic excitations in PrO_2

[ATB *et al*., Phys. Rev. Lett. **86**, 2082 (2001)]

Pr^{4+} $4f^1$ L=3 S=1/2





Expressions for $S(\mathbf{Q}, \omega)$ — 2

2 Pair correlation functions

These are equilibrium properties of the unperturbed system, related to thermodynamic functions

⟶ Theory is simple, direct, exact and quantitative!

◇ Example 1 scattering from lattice of spins of one type

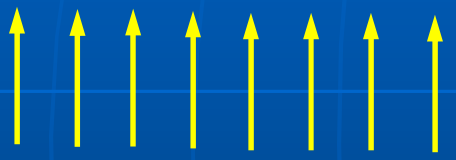
$$S(\mathbf{Q},\omega) = A\int_{-\infty}^{+\infty} dt\, \exp(-i\omega t) \sum_{\mathbf{R}} \exp(i\mathbf{Q}.\mathbf{R})\, \langle \underset{\substack{\uparrow\\ \text{spin–spin correlation function}\\ \text{(measures spectrum of spontaneous spin fluctuations)}}}{S_0^{\perp}(0)} S_{\mathbf{R}}^{\perp}(t)\rangle$$

◇ Example 2 coherent nuclear scattering (one type of atom)

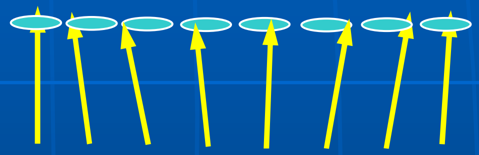
$$S(\mathbf{Q},\omega) = A'\int_{-\infty}^{+\infty} dt\, \exp(-i\omega t) \sum_{\mathbf{R}} \exp(i\mathbf{Q}.\mathbf{R})\, \langle \underset{\substack{\uparrow\\ \text{density–density correlation function}\\ \text{(measures spectrum of spontaneous density fluctuations)}}}{n_0(0)} n_{\mathbf{R}}(t)\rangle$$

Collective Magnetic Excitations

e.g. ferromagnetic spin waves



Ground state

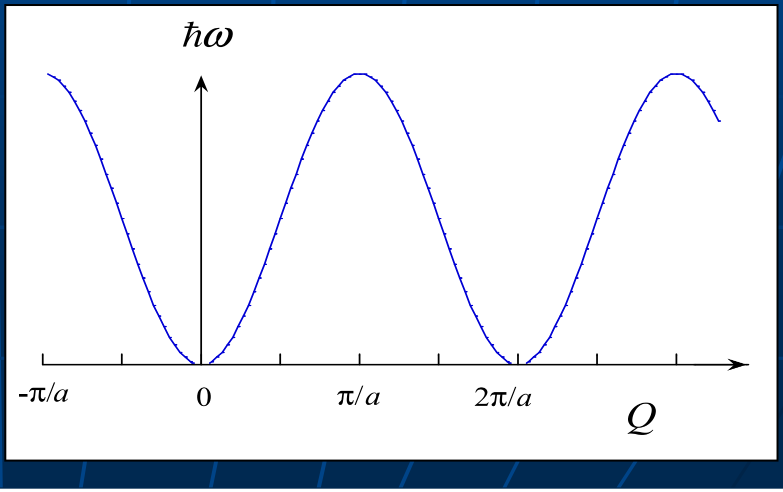


Spin wave

— quantum of energy in a spin wave mode

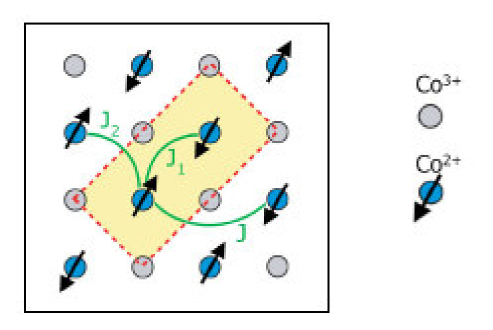
Magnon dispersion curve

(periodic in reciprocal space)



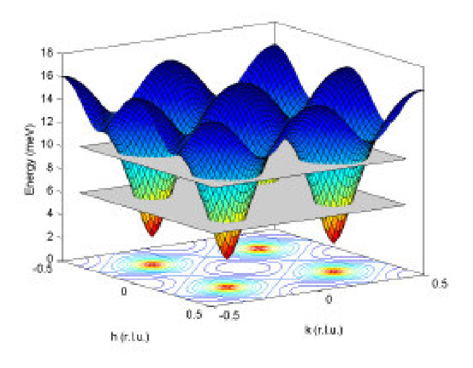
Collective Magnetic Excitations

Example: Antiferromagnons in charge-ordered $La_{3/2}Sr_{1/2}CoO_4$
[Lucy Helme, ATB, *et al*. (2005)]

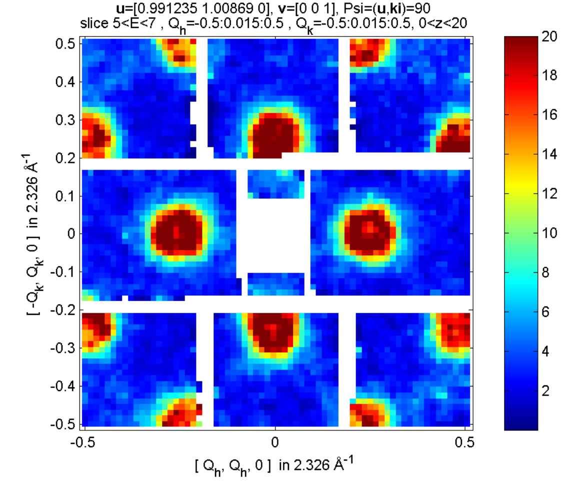


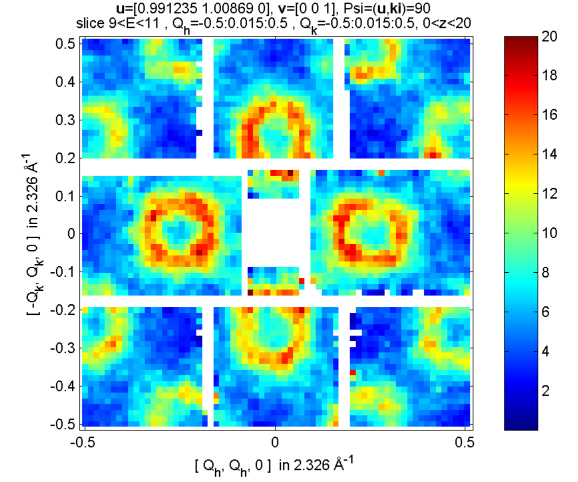
2D square lattice of Co ions
with alternating Co^{2+} and Co^{3+}

Calculated dispersion surface



E = 6 meV



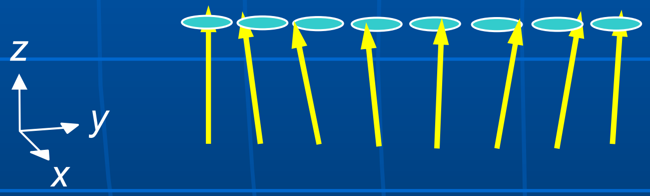


E = 10 meV

Scattering from Spin Waves

S(**Q**,ω) for ferromagnetic spin waves similar to phonons except

◇ Neutron scatters from spin <u>deviations</u> perpendicular to **Q**

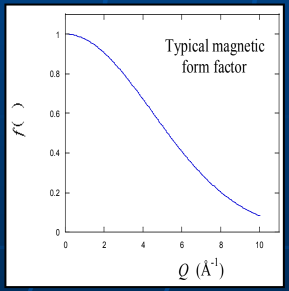


Q Scatter from *x* and *y* spin components

Q Scatter from *x* spin component (no z spin deviation) so intensity weaker by a factor of 2

◇ Instensity decreases with magnetic form factor

◇ No Q^2 variation because lattice is not distorted



Basis for separating scattering from phonons and magnons

Self correlation Function

Example 3 incoherent nuclear scattering

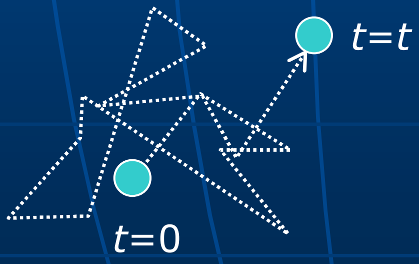
$$S_i(\mathbf{Q},\omega) = A''\int\int dt\, d\mathbf{r}\, \exp(i\mathbf{Q}.\mathbf{r}-i\omega t)\, G_s(\mathbf{r},t)$$

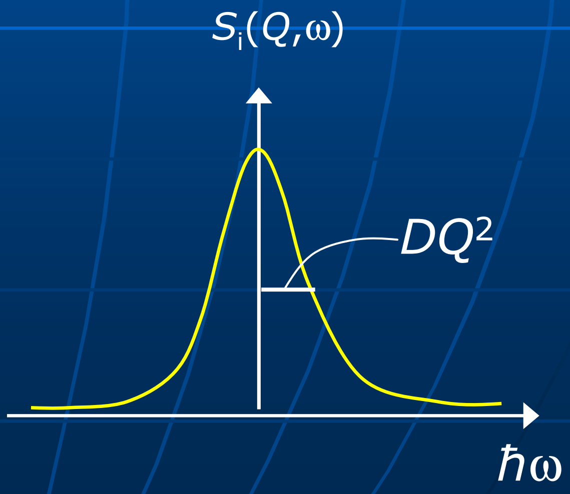
'self' pair correlation function

Measures correlations between
the position of a particle at different times

e.g. diffusion

$\mathbf{J} = D\,\nabla n$





Expressions for $S(\mathbf{Q}, \omega)$ — 3

3 Generalized susceptibility

$$S(\mathbf{Q},\omega) = [1+n(\omega)]\,\frac{1}{\pi}\,\chi''(\mathbf{Q}, \omega)$$

$$\left[n(\omega) = \frac{1}{\exp(\hbar\omega/k_B T) - 1}\right]$$

◇ Magnetic susceptibility

$$\mathbf{M} = \chi\ \mathbf{H}$$

$\mathbf{M}$ = magnetization
$\mathbf{H}$ = applied field
χ = magnetic susceptibility

◇ If H is constant in space and time we measure the zero frequency uniform susceptibility, i.e. $\chi(\mathbf{Q}=0, \omega=0)$

◇ If the applied field varies in space and time we could measure the *generalized* (dynamical) susceptibility $\chi(\mathbf{Q}, \omega)$

$$M_\alpha(\mathbf{Q}, \omega) = \chi_{\alpha\beta}(\mathbf{Q}, \omega)\ H_\beta(\mathbf{Q}, \omega)$$

◇ This applies when the system responds *linearly* to the applied field

Fluctuation–dissipation theorem

In general, *M* is not in phase with *H*, so χ is complex:

$$\chi(\mathbf{Q}, \omega) = \chi'(\mathbf{Q}, \omega) - i\chi''(\mathbf{Q}, \omega)$$

The neutron is a probe that provides a
magnetic perturbation that varies in space and time

Neutron interaction is very weak

→ System responds linearly

→ Fluctuation–dissipation theorem

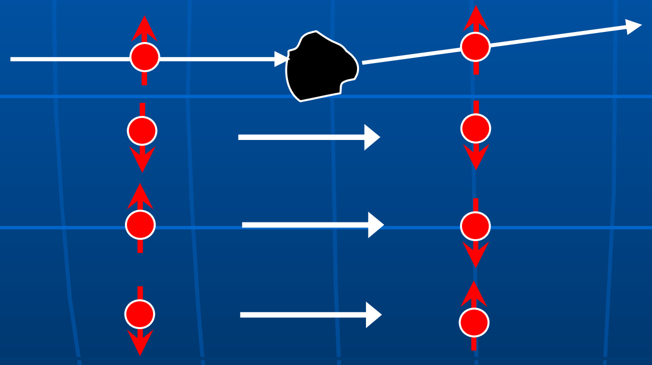
$$S(\mathbf{Q},\omega) = [1+n(\omega)]\ \frac{1}{\pi}\ \chi''(\mathbf{Q}, \omega)$$

Basic excitation spectrum not complicated
by thermal population of states

Polarized neutron scattering

Neutron has spin ½, so is either up (↑) or down (↓) relative to an applied field

◇ Longitudinal polarization analysis



Non-spin-flip (NSF)

Non-spin-flip

Spin-flip (SF)

Spin-flip

◇ Spherical neutron polarimetry

$$\mathbf{P}_i \longrightarrow \mathbf{P}_f$$

Longitudinal polarization analysis

1 Neutrons scatter from spin components ⊥ **Q**

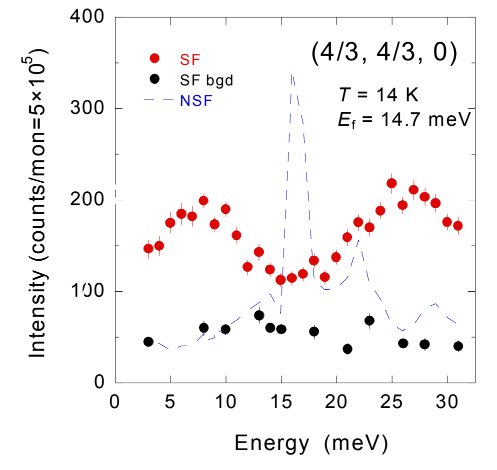
2 Longitudinal neutron polarization:

(i) spin components || **P** do not flip polarization (NSF)
(ii) spin components ⊥ **P** do flip polarization (SF)
(iii) coherent nuclear scattering does not flip polarization (NSF)

Example: separating phonon and magnon scattering

If **P** is parallel to **Q** then:

magnetic scattering is entirely SF
non-magnetic scattering is entirely NSF



Excitations in Stripe ordered $La_{5/3}Sr_{1/3}NiO_4$
ATB *et al*. PRB **67**, 100407(R) (2003)

Summary

Neutron scattering:

(i) is a powerful bulk probe of fundamental electronic and structural correlations in condensed matter

(ii) and provides a direct route to the microscopic origin of the physical properties of materials