

- 1. Introduction
- 2. The neutron interaction with magnetism
- 3. The Born Approximation
- 4. Elastic scattering
- 5. Bragg scattering
- 6. Neutron polarization analysis
- 7. Diffuse scattering



Neutrons and Magnetism

I. Elastic scattering of neutrons

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No time to present:

• Sources

- Techniques and instrumentation
- Resolution
- Analysis methods



Magnetization and the size of the magnetic moment per atom the spatial distribution of magnetization

Magnetic correlations in space and time

magnetic structures, from Å to μ m the influence of impurities, frustration on magnetism magnetic phase transitions and critical exponents

Energy states associated with magnetic electrons and dynamics eigenstates of Hamiltonians magnetic exchange integrals crystal field transitions

Coupling between magnetic and chemical/structural properties superconductivity colossal- and giant magnetoresistance magnetostriction and INVAR effect



FLUX!

Measurements are limited by statistics. Normally have to compromise on resolution. It is difficult to measure $< 0.1 \mu_B$ per atom.

Neutrons interact with all the magnetic fields in the sample, it can be difficult to separate different components.

Neutrons don't measure spatial dimensions directly, they measure in Fourier space. Conclusions are often model-dependent This is both a plus and a minus!



PUT

NEUTRONS

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Neutrons are *matter waves*.

- They have a de Broglie wavelength of λ

 (λ ~ 1.8 Å is a 'standard' for 'thermal' neutrons,
 but experiments can use ~ 0.1 to 100 Å neutrons)
- They have a momentum of $\mathbf{p} = h/\lambda = \hbar k$ and a kinetic energy of $E = h^2/2m\lambda^2 = \hbar^2 k^2/2m$
- They have a magnetic dipole moment given by $-\gamma \mu_N \hat{\sigma}$ (where $\hat{\sigma}$ is the Pauli spin operator)
- The kinetic energy of a thermal neutron is about the same as the energy of a lattice or magnetic vibration

We must solve the Wave equation for the neutron/sample ensemble.

$$\left[-\left(\hbar^2/2M\right)\nabla^2 + \hat{V}(\mathbf{r})\right]\psi = E\psi$$

 $\hat{V}(\mathbf{r})$ is the potential energy operator. For neutrons it is called the *Fermi pseudo-potential*.

$$\hat{V}(\mathbf{r}) = V_n(\mathbf{r}) + V_m(\mathbf{r})$$
where
$$V_n(\mathbf{r}) = \frac{2\pi\hbar^2}{m_n} (b + B\hat{\mathbf{I}} \cdot \hat{\boldsymbol{\sigma}}) \delta(\mathbf{r})$$

W

$$V_m(\mathbf{r}) = -\gamma \mu_N \hat{\boldsymbol{\sigma}} \cdot \mathbf{B}(\mathbf{r})$$

b is the nuclear scattering length

 $B\hat{\mathbf{I}}$ is the nuclear spin

B(**r**) is the magnetic induction

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$$V_m(\mathbf{r}) = -\gamma \mu_N \hat{\boldsymbol{\sigma}} \cdot \mathbf{B}(\mathbf{r})$$
b is the nuclear scattering length
B\hat{\mathbf{l}} is the nuclear spin
B(\mathbf{r}) is the magnetic field
magnetic!

Neutron scattering

Most neutron magnetic experiments are *scattering* experiments.



Constructive and destructive interference leads to peaks in the intensity as a function of:

- 1) the momentum transfer: $\mathbf{Q} = \mathbf{k} \mathbf{k}'$
- 2) the energy transfer: $\Delta E = (\hbar^2/2m)(|k|^2 |k'|^2)$
 - 3) the change in the neutron spin orientation



Most neutron experiments are scattering experiments.

Fig. 1.2 Geometry for scattering experiment.



The target volume is initially in state ζ .

A neutron enters with wave vector *k* and spin *s* It interacts with the target.

The final neutron wave vector is k' and spin s'. The final target state is ζ' .

If the neutron has a *plane wave function*, if the interaction is *weak*, then the wave equation can be solved using first order perturbation theory, i.e. Fermi's Golden Rule

$$\frac{\mathrm{d}^{2}\sigma}{\mathrm{d}\Omega\cdot\mathrm{d}E} = \frac{k'}{k} \left(\frac{m_{n}}{2\pi\hbar^{2}}\right)^{2} \sum_{\zeta,s} p_{\zeta} p_{s} \sum_{\zeta',s'} \left|\langle \boldsymbol{k}',s',\zeta' | \hat{V}(\mathbf{r}) | \boldsymbol{k},s,\zeta \rangle \right|^{2} \delta(\hbar\omega + E_{\zeta} - E_{\zeta'})$$

This is known as the *cross-section*, and gives the probability that a neutron will scatter in to a certain solid angle with a certain change in energy

The two assumptions form the first *Born approximation*



The cross-section

Appropriate averaging over the target energy states, the positions **r**, and the neutron spin directions is necessary to find the measured cross-section

G. L. Squires, *Introduction to the theory of thermal neutron scattering*, Dover Publications, New York, 1978 W. Marshall and S. W. Lovesey, *Theory of thermal neutron scattering*, Oxford University Press, Oxford, 1971 S. W. Lovesey, *Theory of neutron scattering from condensed matter*, Oxford University Press, Oxford, 1986



Elastic scattering

If the incident neutron energy = the final neutron energy, the scattering is *elastic*.

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \sum_{\zeta',s'} p_s \left| \langle \boldsymbol{k}',s' | \hat{V}(\mathbf{r}) | \boldsymbol{k},s \rangle \right|^2$$

Forget about the spins for the moment and integrate over all **r**:

$$\langle \mathbf{k}' | \hat{V}(\mathbf{r}) | \mathbf{k} \rangle = \int \hat{V}(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot d\mathbf{r}$$

Momentum transfer $\mathbf{Q} = \mathbf{k} - \mathbf{k}'$

The elastic cross-section is then directly proportional to the *Fourier transform squared* of the potential. Neutron scattering thus works in Fourier space, otherwise called *reciprocal space*.

Elastic neutron scattering is also referred to as neutron diffraction





$$\int V_m(\mathbf{r})e^{i\mathbf{Q}\cdot\mathbf{r}}\cdot d\mathbf{r} = -\gamma\mu_N\hat{\boldsymbol{\sigma}}\int \mathbf{B}(\mathbf{r})e^{i\mathbf{Q}\cdot\mathbf{r}}\cdot d\mathbf{r} = -\gamma\mu_N\hat{\boldsymbol{\sigma}}\cdot\mathbf{B}(\mathbf{Q})$$

B(Q) is related to the *magnetization* of the sample, M(r), through the equation:

$$\mathbf{B}(\mathbf{Q}) = \int \hat{\mathbf{Q}} \times \left(\mathbf{M}(\mathbf{r}) \times \hat{\mathbf{Q}} \right) e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot d\mathbf{r} = \mathbf{M}_{\perp}(\mathbf{Q})$$

Neutron scattering therefore probes the components of the sample magnetization that are *perpendicular* to the neutron's momentum transfer,**Q**.

and
$$\frac{\int V_m(\mathbf{r})e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot d\mathbf{r} = \mathbf{M}_{\perp}(\mathbf{Q})}{d\Omega} = \left\langle \mathbf{M}_{\perp}^*(\mathbf{Q}) \right\rangle \left\langle \mathbf{M}_{\perp}(\mathbf{Q}) \right\rangle$$

Neutron scattering measures the *correlations* in magnetization, i.e. the influence a magnetic moment has on its neighbours.

It is capable of doing this over all length scales, limited only by wavelength.

Elastic scattering





Crystalline structures $\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} \propto \int \left| \left\langle \hat{V} \right\rangle \right|^2 e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot d\mathbf{r}$

Recall the Fourier transform from a series of delta-functions



Bragg's Law: $2d\sin\theta = \lambda$ Leads to Magnetic Crystallography

A simple example of magnetic elastic scattering

 MgB_2 is a superconductor below 39K, and expels all magnetic field lines (Meisner effect). Above a critical field, flux lines penetrate the sample.





The momentum transfer, \mathbf{Q} , is roughly perpendicular to the flux lines, therefore all the magnetization is seen.

(recall
$$\frac{d\sigma_{magnetic}}{d\Omega} = \langle \mathbf{M}_{\perp}^{*}(\mathbf{Q}) \rangle \langle \mathbf{M}_{\perp}(\mathbf{Q}) \rangle$$
)

A simple example of magnetic elastic scattering

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Antiferromagnetism in MnO



temperature and at 80°K.

C. G. Shull & J. S. Smart, Phys. Rev. 76 (1949) 1256

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C. G. Shull & J. S. Smart, Phys. Rev. 76 (1949) 1256



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C. G. Shull *et al.*, Phys. Rev. **83** (1951) 333H. Shaked *et al.*, Phys. Rev. B **38** (1988) 11901

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C. G. Shull *et al.*, Phys. Rev. **83** (1951) 333H. Shaked *et al.*, Phys. Rev. B **38** (1988) 11901

Chromium is an example of an *itinerant* antiferromagnet Reciprocal space Real space



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a Spin Density wave

(Recall the Fourier Transform for two Delta functions)





E.Fawcett, Rev. Mod. Phys. 60 (1988) 209 INSTITUT MAX VON LAUE - PAUL LANGEVIN

Magnetic structure of Holmium



W. C. Koehler, in Magnetic Properties of Rare Earth Metals, ed. R. J. Elliot (Plenum Press, London, 1972) p. 81

R. A. Cowley and S. Bates, J. Phys. C 21 (1988) 4113

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A. R. Mackintosh and J. Jensen, Physica B 180 & 181 (1992) 1



A map in reciprocal space...leads to the structure in real space





(complicated!)

O. Zaharko et al., Phys. Rev. Lett. 93 (2004) 217206

Nuclear magnetic order

The *nuclear* magnetic phase diagram as a function of field at 60 nK $B | [01\overline{1}]$

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A. S. Oja and O. V. Lounasmaa, Rev. Mod. Phys. 69 (1997) 1

Polarized neutrons

- all and

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Polarized neutrons

1.400

$$\frac{d\sigma}{d\Omega} = \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \sum_{\zeta',s'} p_s |\langle \mathbf{k}', s' | \hat{V}(\mathbf{r}) | \mathbf{k}, s \rangle|^2$$
$$U^{ss'} = \langle s' | \hat{V} | s \rangle = \langle s' | b + B \hat{\mathbf{l}} \cdot \hat{\boldsymbol{\sigma}} - \mathbf{M}_{\perp} \cdot \hat{\boldsymbol{\sigma}} | s \rangle$$
Neutron polarization coordinates:
z is parallel to **P**

P. 2019 55710

Neglect nuclear spin for the moment

$$U^{++} = b - M_{\perp z}$$

$$U^{--} = b + M_{\perp z}$$

$$U^{+-} = -(M_{\perp x} + iM_{\perp y})$$

$$U^{-+} = -(M_{\perp x} - iM_{\perp y})$$

REMEMBER: the only visible components of the magnetization are PERPENDICULAR to **Q**

R. M. Moon, T. Riste and W. K. Koehler, Phys. Rev. 181 (1969) 920



- The non-spin flip cross-sections have only nuclear components
- The spin-flip cross-sections have only the magnetic components
- There is a *complete separation* of nuclear from magnetic scattering.
- The cross term in the spin flip cross-sections usually cancels, i.e. $M_{\perp}(\mathbf{r}_i) \times M_{\perp}^*(\mathbf{r}_j) = -(M_{\perp}(\mathbf{r}_i) \times M_{\perp}^*(\mathbf{r}_j))$

 α -Fe₂O₃ is antiferromagnetic. Powder diffraction gives Bragg peaks with mixed nuclear and magnetic intensities

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R. M. Moon, T. Riste and W. K. Koehler, Phys. Rev. 181 (1969) 920

Elastic scattering, $\mathbf{P} \perp \mathbf{Q}$ $d\sigma^{\pm\pm} = \left| \int (b(\mathbf{r}) \mp M_{\perp z}(\mathbf{r})) e^{i\mathbf{Q}\cdot\mathbf{r}} d\mathbf{r} \right|^2$ $\frac{d\sigma^{\pm\pm}}{d\Omega} = \left| \int M_{\perp y}(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} d\mathbf{r} \right|^2$

• The non-spin flip cross-sections probe the components of the magnetization parallel to the neutron polarization

• The spin-flip cross-sections probe the components of the magnetization that are perpendicular to *both* the polarization and to \mathbf{Q}

• There is a difference between the two non-spin flip cross-sections, the difference is called nuclear-magnetic interference.

• The two spin-flip cross-sections are equivalent

Single domain collinear ferromagnets have a magnetization that is always parallel to **P**.

If
$$\mathbf{P} \perp \mathbf{Q}$$
, $M_{\perp y}(\mathbf{r}) = 0$ and
$$\begin{cases} \frac{d\sigma^{\pm\pm}}{d\Omega} = \left| \int (b(\mathbf{r}) \mp M(\mathbf{r})) e^{i\mathbf{Q} \cdot \mathbf{r}} d\mathbf{r} \right|^2 \\ \frac{d\sigma^{\pm\mp}}{d\Omega} = 0 \end{cases}$$

The difference between the two non-spin flip cross-sections will give information on the ferromagnetic moment.

Plot M(Q) as a function of Q and a characteristic line shape will emerge:



P. Javorsky et al., Phys. Rev. B 67 (2003) 224429

This characteristic line shape is known as the form factor, f(Q)

The form factor arises from the *spatial distribution* of *unpaired electrons* around a magnetic atom. For magnetic scattering due only to electron spin:

Magnetic form factors

$$\mathbf{M}(\mathbf{Q}) = \int \mathbf{M}(\mathbf{r})e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot d\mathbf{r}$$

$$\propto \int s_d e^{i\mathbf{Q}\cdot\mathbf{r}_d} \cdot d\mathbf{r}_d \int S(\mathbf{R})e^{i\mathbf{Q}\cdot\mathbf{R}} \cdot d\mathbf{R}$$

$$= f(Q)\int S(\mathbf{R})e^{i\mathbf{Q}\cdot\mathbf{R}} \cdot d\mathbf{R}$$

$$f(Q) = \int s_d e^{i\mathbf{Q}\cdot\mathbf{r}_d} \cdot d\mathbf{r}_d$$

$$\mathbf{R}$$
Atom at position **R** with a density of unpaired (magnetic) electrons

For magnetic scattering due only to electron spin:

$$\frac{d\sigma_{magnetic}}{d\Omega} = \left\langle \mathbf{M}_{\perp}^{*}(\mathbf{Q}) \right\rangle \left\langle \mathbf{M}_{\perp}(\mathbf{Q}) \right\rangle$$
$$\propto f^{2}(Q) \int S_{\perp}(\mathbf{R}_{i}) S_{\perp}^{*}(\mathbf{R}_{j}) e^{i\mathbf{Q}\cdot(\mathbf{R}_{i}-\mathbf{R}_{j})} \cdot d\mathbf{R}$$

Magnetic electron density



P. Javorsky et al., Phys. Rev. B 67 (2003) 224429

From the form factors, the magnetic moment density in the unit cell can be derived



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magnetic moment density

Elastic scattering

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \sum_{\zeta',s'} p_s \left|\langle \mathbf{k}',s' | \hat{V}(\mathbf{r}) | \mathbf{k},s \rangle\right|^2$$
$$\propto \int \left|\langle \hat{V} \rangle\right|^2 e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot d\mathbf{r} + \left(\left|\langle \hat{V}^2 \rangle\right| - \left|\langle \hat{V} \rangle\right|^2\right) \int z(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot d\mathbf{r}$$

The contribution from deviations from the average structure: *Short-range* order

The contribution from the average structure of the sample: *Long-range* order

and the

The physical meaning of $z(\mathbf{r})$





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R. A. Cowley, Physica B 350 (2004) 1





R. A. Cowley, Physica B 350 (2004) 1

Magnetic and nuclear short-range order are often coupled

At the very least, the diffuse scattering is often a mixture of magnetic and nuclear contributions Neutron polarization analysis is usually essential for the measurement of magnetic short-range order

Recall: if $\mathbf{P} \parallel \mathbf{Q}$, the non-spin flip scattering is all nuclear the spin flip scattering is all magnetic

> if $\mathbf{P} \perp \mathbf{Q}$, the non-spin flip scattering is due to nuclear and magnetic, the magnetic components are parallel to \mathbf{P} . the spin flip scattering is all magnetic, the magnetic components are perpendicular to \mathbf{P} and \mathbf{Q} .

> > T. J. Hicks, Magnetism in Disorder, 1995, Clarendon Press, Oxford

Diffuse scattering from a paramagnet, MnF₂

For a paramagnet,
$$z(\mathbf{r}) = \delta(r)$$
, $\frac{\mathrm{d}\sigma^{\pm\mp}}{\mathrm{d}\Omega} \propto \frac{2}{3}f^2(Q)S(S+1)$

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R. M. Moon, T. Riste and W. K. Koehler, Phys. Rev. 181 (1969) 920



nuclear scattering

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magnetic scattering



A. P. Murani *et al.*, Physica B **267-268** (1999) 131 T. J. Hicks, *Magnetism in Disorder*, 1995, Clarendon Press, Oxford J. R. Stewart *et al.*, J. Appl. Phys. **87** (2000) 5425

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Form-factor like background, frozen moments

A. P. Murani et al., Physica B 267-268 (1999) 131

- T. J. Hicks, Magnetism in Disorder, 1995, Clarendon Press, Oxford
- J. R. Stewart et al., J. Appl. Phys. 87 (2000) 5425



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Diffuse scattering from magnetic frustration, Gd₂Ti₂O₇

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 $\frac{\mathrm{d}\sigma^{\pm\mp}}{\mathrm{d}\Omega} \propto \frac{2}{3} f^2(Q) \sum \langle \mathbf{S}_o \mathbf{S}_r \rangle \frac{\sin(Qr)}{Or}$



J. R. Stewart et al., J. Phys.: Condens. Matter 16 (2004) L321

Diffuse scattering in metallic glasses, Fe_{80-x}Ru_xB₂₀

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$$\frac{\mathrm{d}\sigma^{\pm\pm}}{\mathrm{d}\Omega} = \left| \int (b(\mathbf{r}) \mp M_{\perp z}(\mathbf{r})) e^{i\mathbf{Q}\cdot\mathbf{r}} d\mathbf{r} \right|^2$$

 $M(\mathbf{r})$ is directional - its sign can change as a function of \mathbf{r} .



N. Cowlam et al., J. Phys.: Condens. Matter 17 (2005) 3585

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Inelastic scattering Reflectivity and dynamical scattering Neutron polarimetry

After coffee...