



MAX-PLANCK-GESELLSCHAFT

Magnetic Neutron Scattering and Spin-Polarized Neutrons

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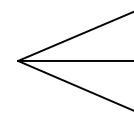


Nuclear Neutron Scattering:

- Elastic Neutron Scattering
Diffraction – Crystal Structure
- Inelastic Neutron Scattering
Excitations - Phonons

Magnetic Neutron Scattering:

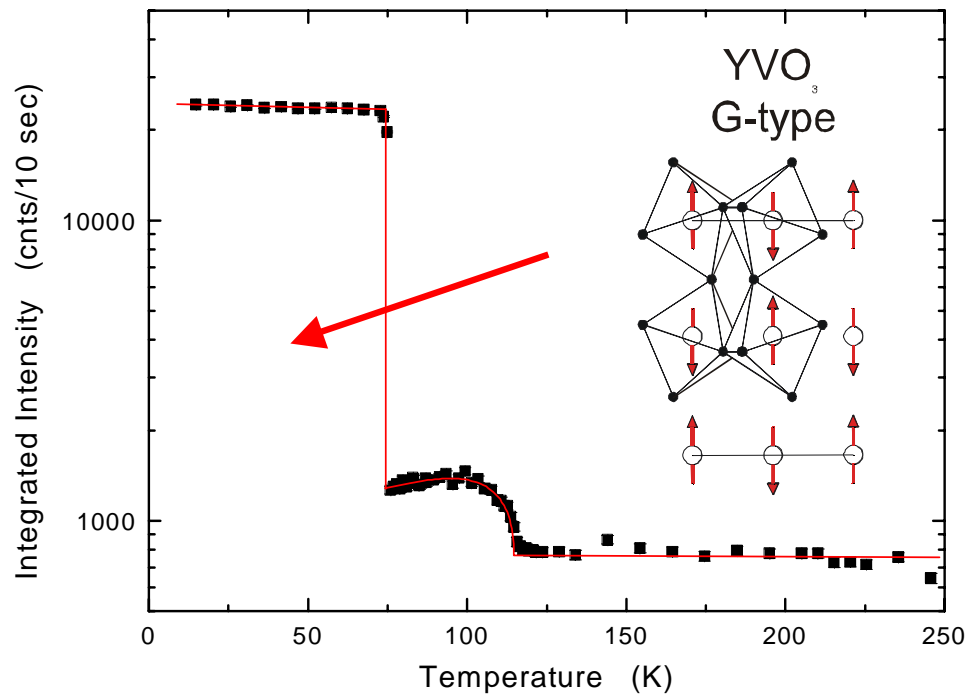
- Elastic Neutron Scattering
Diffraction – Spin Structure
- Inelastic Neutron Scattering
Excitations - Magnons



ferro / antiferromagnetic
magnetic moment
direction of the spins

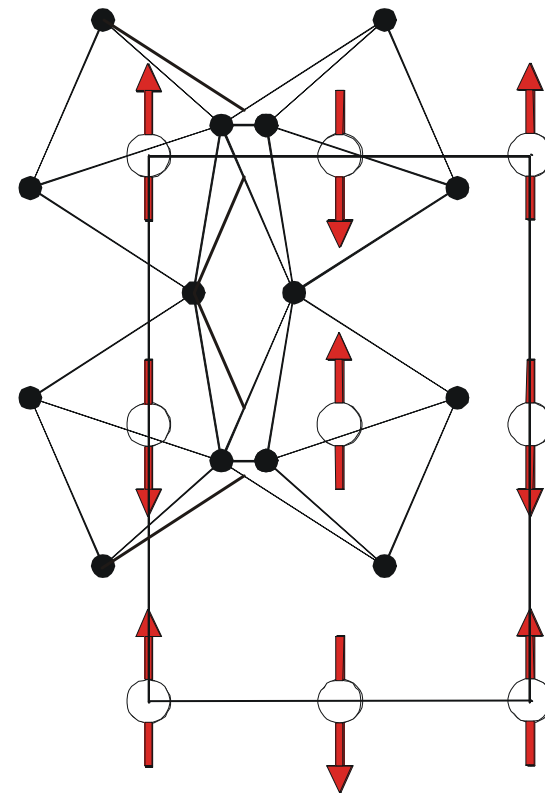


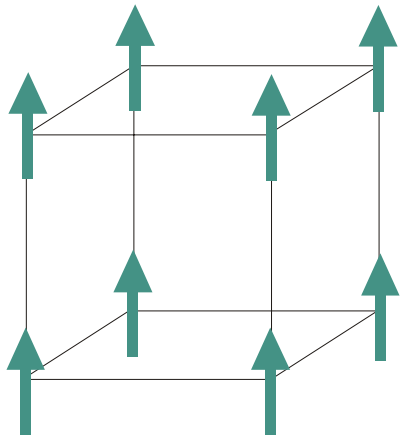
$(1/2, 1/2, 1/2)$ - Bragg Reflection



$$\mu_0 = 1.72 \mu_B / \text{V}^{3+}\text{-ion}$$

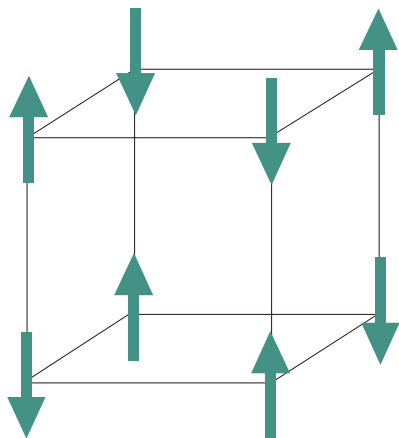
$T < 77 \text{ K}$
G-type



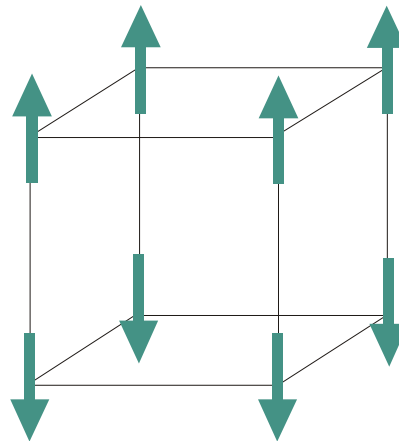


TYPE:
Ferromagnet

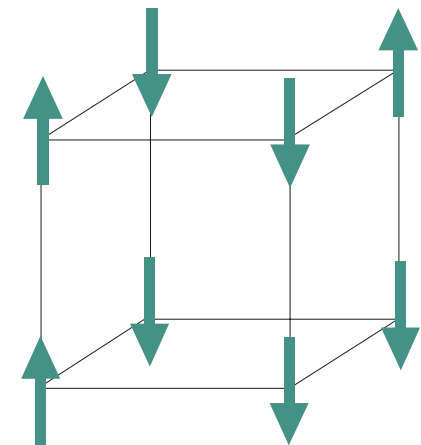
Antiferromagnet



G-type



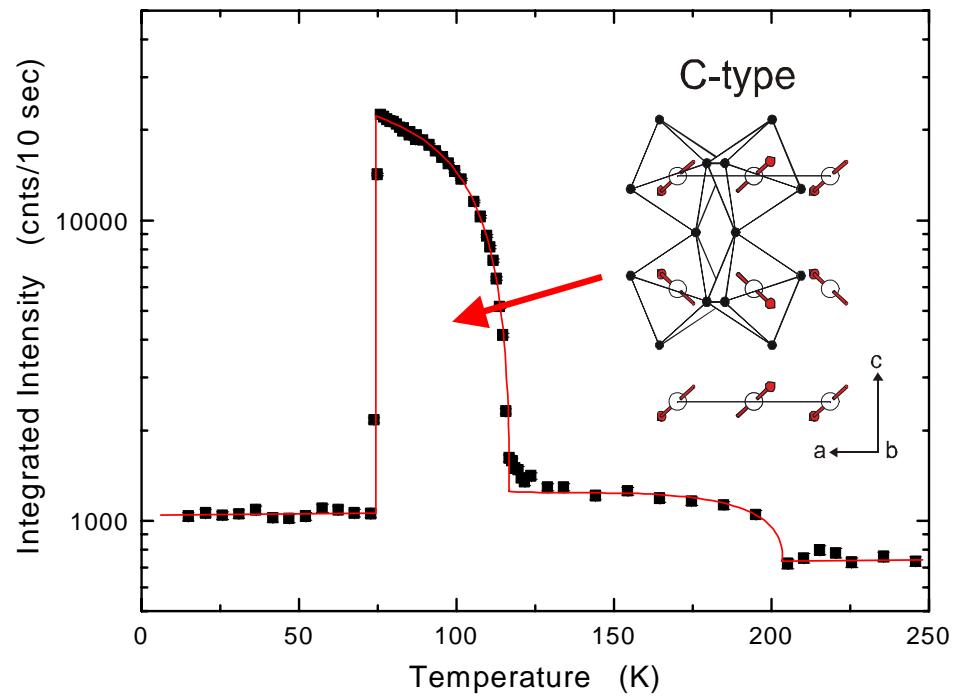
A-type



C-type

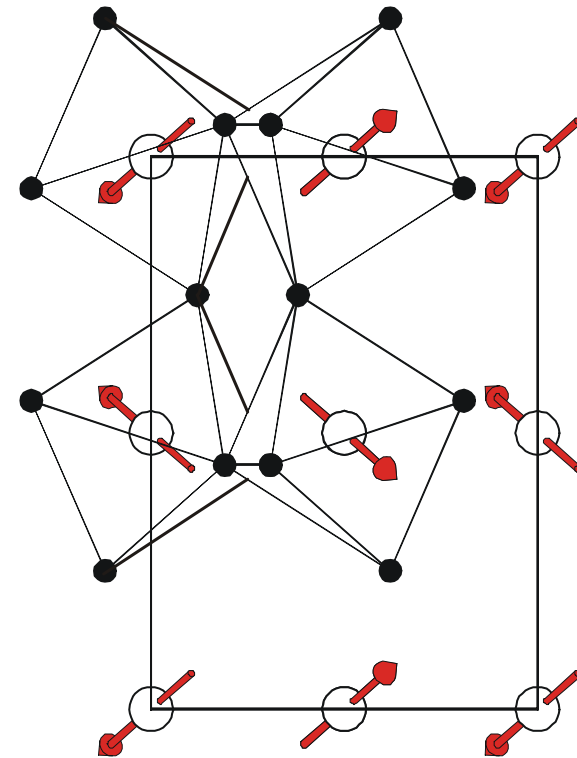


$(1/2, 1/2, 0)$ - Bragg Reflection



$$\mu_0 = 1.05 \mu_B / V^{3+}\text{-ion}$$

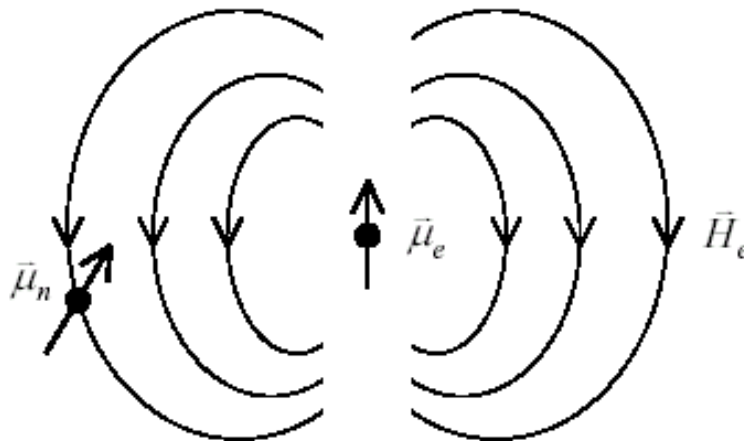
$77 \text{ K} < T < 116 \text{ K}$
C-type





Magnetic Neutron Scattering and Spin-Polarized Neutrons

Physical origin: potential of magnetic dipole moment of the neutron in magnetic field generated by electron spins and orbital moments in the solid.



Specialize to contribution of electron **spin** to \vec{H}_e :

$$\vec{\mu}_e = -2\mu_B \vec{s}_e \text{ with } \mu_B = \frac{e\hbar}{2m_e}$$

$$\vec{\mu}_n = -g_n \mu_N \vec{s}_n \equiv -\gamma \mu_N \vec{\sigma} \text{ with } \mu_N = \frac{e\hbar}{2m_n} \text{ and } \gamma = \frac{g_n}{2} = 1.913$$

\vec{s}_e and \vec{s}_n are spin- $\frac{1}{2}$ operators, $\vec{\sigma} = 2\vec{s}_n$ is the neutron Pauli matrix whose eigenvalues are ± 1 .



Differential neutron scattering cross section for one electron at rest at origin of coordinate system:

$$\frac{d\sigma}{d\Omega} = \left(\frac{m_n}{2\pi\hbar^2} \right)^2 \left| \langle \vec{k}_f, m_f | H_{\text{int}} | \vec{k}_i, m_i \rangle \right|^2 \quad \text{with } H_{\text{int}} = -\vec{\mu}_n \cdot \vec{H}_e$$

This follows from Fermi's Golden Rule, as in the case of nuclear neutron scattering. The only qualitative difference is that we have to keep track of the neutron spin state $|m\rangle = |\pm 1\rangle$ in addition to the spatial part of its wave function labelled by the quantum number \vec{k} .

Vector potential of dipole field (from classical electromagnetism):

$$\vec{A}_e = \frac{\mu_0}{4\pi} \frac{\vec{\mu}_e \times \vec{r}}{|\vec{r}|^3} = \frac{\mu_0}{4\pi} \vec{\mu}_e \times \vec{\nabla} \frac{1}{|\vec{r}|}$$

$$\vec{H}_e = \vec{\nabla} \times \vec{A}_e = \frac{\mu_0}{4\pi} \vec{\nabla} \times \left(\vec{\mu}_e \times \nabla \frac{1}{|\vec{r}|} \right)$$

$$\frac{d\sigma}{d\Omega} = \left(\frac{m_n}{2\pi\hbar^2} \right)^2 (2\gamma\mu_N\mu_B)^2 \left| \langle \vec{k}_f, m_f | \underbrace{\vec{\sigma}_n \cdot \vec{\nabla} \times \left(\vec{s}_e \times \nabla \frac{1}{|\vec{r}|} \right)}_{\text{magnetic field}} | \vec{k}_i, m_i \rangle \right|^2$$

neutron
spin

magnetic
field



Use trick to simplify this expression:

$$\begin{aligned} \int \frac{d\bar{p}}{|\bar{p}|^2} e^{i\bar{p}\cdot\bar{r}} &= 2\pi \int_0^\infty d|\bar{p}| \int_{-1}^1 e^{i|\bar{p}||\bar{r}|\cos\Theta} d(\cos\Theta) \\ &= 2\pi \int_0^\infty d|\bar{p}| \frac{\sin|\bar{p}||\bar{r}|}{|\bar{p}||\bar{r}|} = \frac{2\pi^2}{|\bar{r}|} \end{aligned}$$

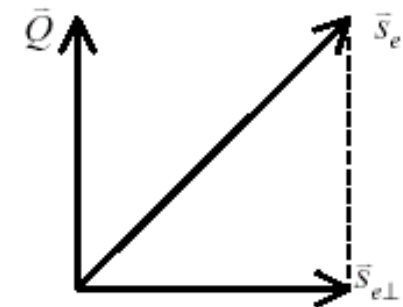
Here, \bar{p} is an auxiliary variable without physical meaning.

$$\begin{aligned} \nabla \times \left(\bar{s}_e \times \nabla \frac{1}{|\bar{r}|} \right) &= \frac{1}{2\pi^2} \int \frac{d\bar{p}}{|\bar{p}|^2} \bar{\nabla} \times (\bar{s}_e \times \bar{\nabla}) e^{i\bar{p}\cdot\bar{r}} \\ &= \frac{1}{2\pi^2} \int \hat{p} \times (\bar{s}_e \times \hat{p}) e^{i\bar{p}\cdot\bar{r}} d\bar{p} \end{aligned}$$

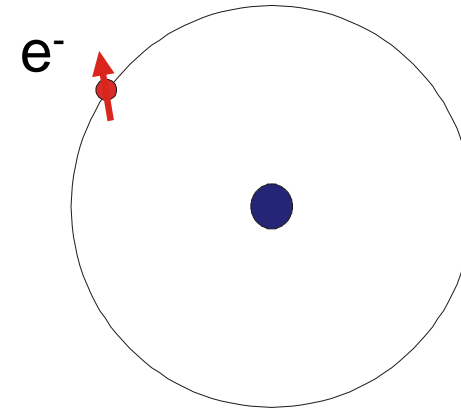
$$\begin{aligned} \langle \bar{k}_f | \nabla \times \left(\bar{s}_e \times \nabla \frac{1}{|\bar{r}|} \right) | \bar{k}_i \rangle &= \frac{1}{2\pi^2} \int d\bar{r} e^{-i\bar{Q}\cdot\bar{r}} \int d\bar{p} \hat{p} \times (\bar{s}_e \times \hat{p}) e^{i\bar{p}\cdot\bar{r}} \\ &= 4\pi \underbrace{\hat{Q} \times (\bar{s}_e \times \hat{Q})}_{\equiv \bar{s}_{e\perp}} \end{aligned}$$

The last line follows by doing the \bar{r} -integration first and using

$$\frac{1}{(2\pi)^3} \int d\bar{r} e^{i(\bar{p}-\bar{Q})\cdot\bar{r}} = \delta(\bar{p}-\bar{Q})$$



$$Q = k_f k_i$$

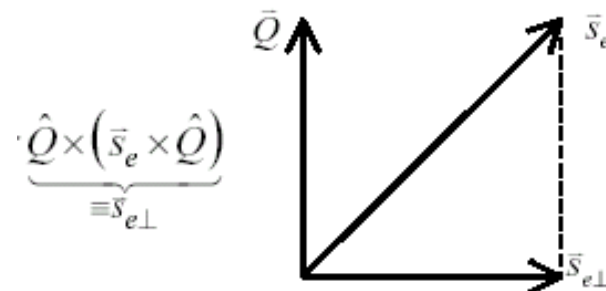


collect all prefactors:

$$\left(\frac{m_n}{2\pi\hbar^2}\right)^2 (2\gamma\mu_N\mu_B)^2 \left(\frac{\mu_0}{4\pi}\right)^2 (4\pi)^2 = (\gamma r_0)^2$$

where $r_0 = 2.8 \times 10^{-15} \text{ m}$ is the “classical electron radius” that also appeared in the Thompson cross section for x-ray scattering.

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \underbrace{\left| \langle m_f | \vec{\sigma} \cdot \vec{s}_{e\perp} | m_i \rangle \right|^2}_{\left[1 - (\hat{n} \cdot \hat{Q})^2 \right]}$$





For an **unpolarized** neutron beam, one has to average over the neutron spin states $|m\rangle$. For convenience, take the neutron spin quantization axis \hat{z} to be parallel to $\vec{s}_{e\perp}$. Then

$$\begin{aligned}\langle m_f | \vec{\sigma} \cdot \vec{s}_{e\perp} | m_i \rangle &= s_{e\perp} \langle m_f | \sigma_z | m_i \rangle \\ &= \begin{cases} s_{e\perp} & \text{if } m_f = m_i \\ 0 & \text{otherwise} \end{cases}\end{aligned}$$

The cross section for a single electron at rest for an unpolarized neutron beam is therefore

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 |\vec{s}_{e\perp}|^2$$

with $\vec{s}_{e\perp}$ the projection of the electron spin perpendicular to \vec{Q} .

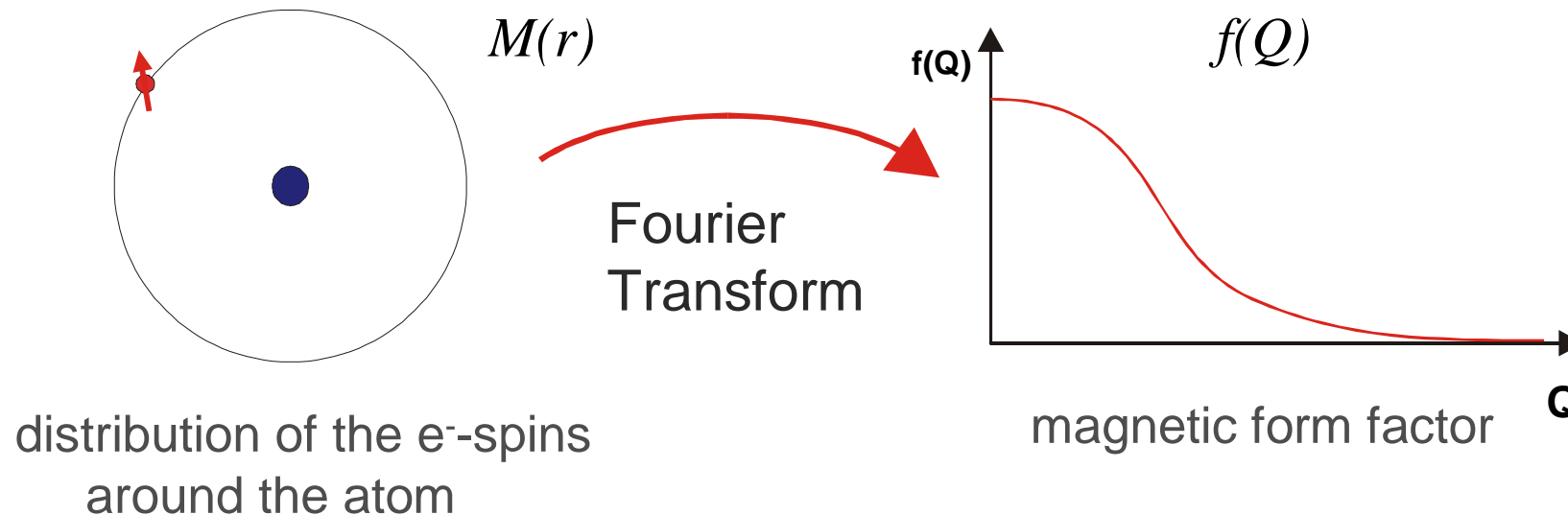


Generalization for an atom

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 |\hat{n}_\perp|^2 |f(\bar{Q})|^2 = (\gamma r_0)^2 [1 - (\hat{n} \cdot \hat{Q})^2] |f(\bar{Q})|^2$$

where $f(\bar{Q}) = \frac{1}{2\mu_B} \int \bar{\mathcal{M}}(\bar{r}) e^{-i\bar{Q} \cdot \bar{r}}$ is the magnetic form factor and

$\bar{\mathcal{M}}(\bar{r}) = \mathcal{M}(\bar{r}) \hat{n}$ is the magnetic dipole moment density contributed by the spin and orbital motion of all unpaired electrons in the atom.





Generalization for collinear magnets

For collinear magnets, that is, magnetically ordered solids where all electron magnetic dipole moments point either parallel or antiparallel to a single direction $\hat{\eta}$, this expression can be generalized in analogy to the differential cross section for x-ray or nuclear neutron scattering:

$$\begin{aligned}\frac{d\sigma}{d\Omega} &= (\gamma r_0)^2 \left[1 - (\hat{\eta} \cdot \hat{Q})^2 \right] \left| \sum_{\vec{R}} (\pm) f_{\vec{R}}(\vec{Q}) e^{i\vec{Q} \cdot \vec{R}} \right|^2 \\ &= (\gamma r_0)^2 \left[1 - (\hat{\eta} \cdot \hat{Q})^2 \right] N \frac{(2\pi)^3}{V_0} \sum_{\vec{K}_M} |F_M(\vec{K}_M)|^2 \delta(\vec{Q} - \vec{K}_M)\end{aligned}$$

where \vec{K}_M are the magnetic reciprocal lattice vectors and

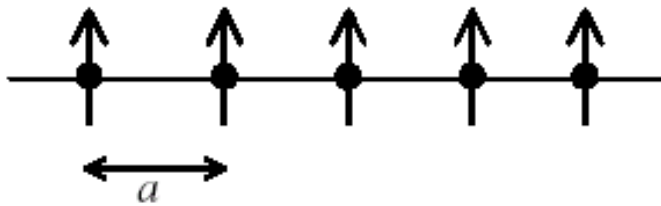
$$F_M(\vec{Q}) = \sum_{\vec{d}} (\pm) f_{\vec{d}}(\vec{Q}) e^{i\vec{Q} \cdot \vec{d}} \quad \leftarrow$$

The + or – sign is chosen depending on whether the magnetic moment at lattice site \vec{d} is parallel or antiparallel to $\hat{\eta}$.

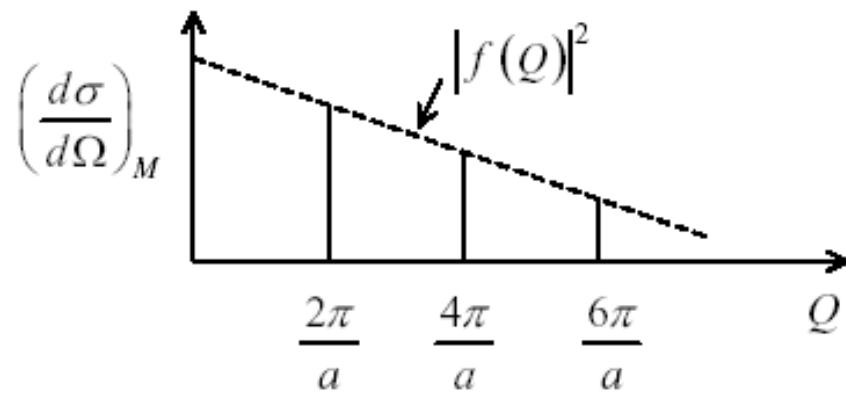
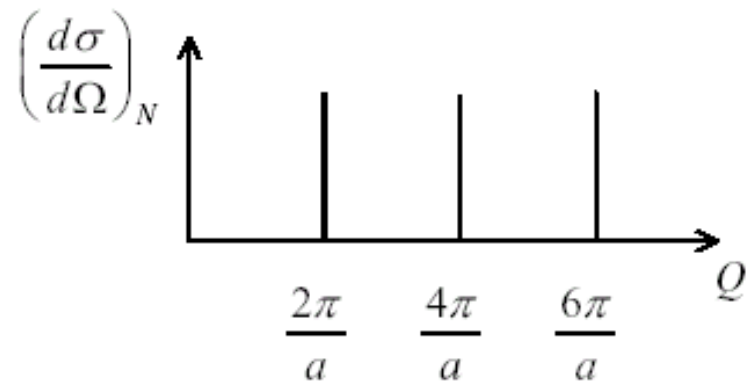


Examples:

1 One-dimensional ferromagnet

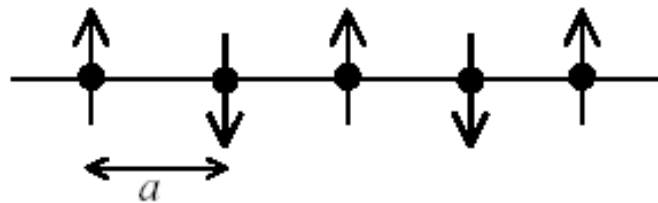


nuclear and magnetic unit cells identical $\Rightarrow K_M = K_N = \frac{2\pi}{a}n$, n integer.



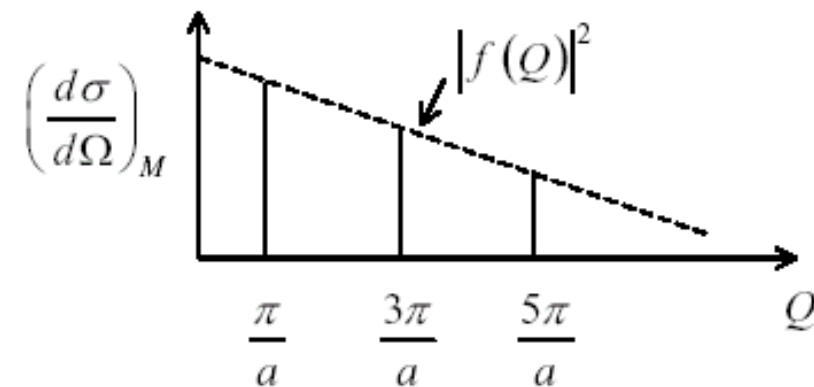
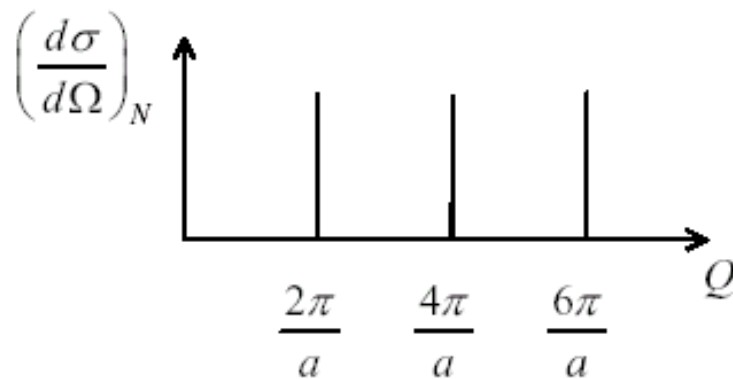


2 One-dimensional antiferromagnet



magnetic unit cell twice as large as nuclear unit cell $\Rightarrow K_M = \frac{\pi}{a}n \neq K_N = \frac{2\pi}{a}n$

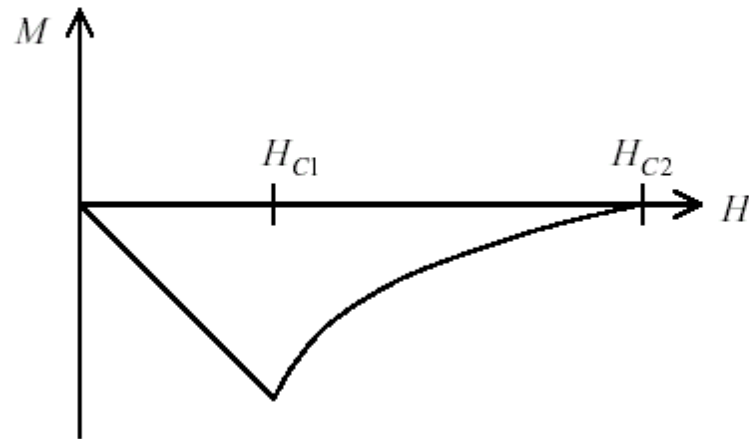
$$|F_M|^2 = |f(Q)|^2 |1 - e^{iQa}|^2 = 4|f(Q)|^2 \sin^2 \frac{Qa}{2} = \begin{cases} 4|f(Q)|^2 & \text{for } n \text{ odd} \\ 0 & \text{for } n \text{ even} \end{cases}$$



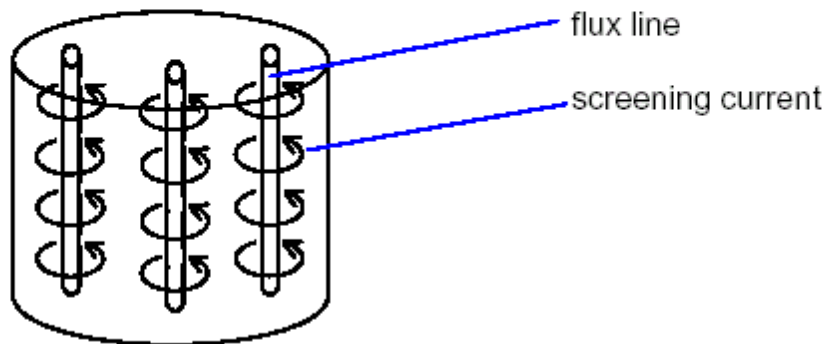


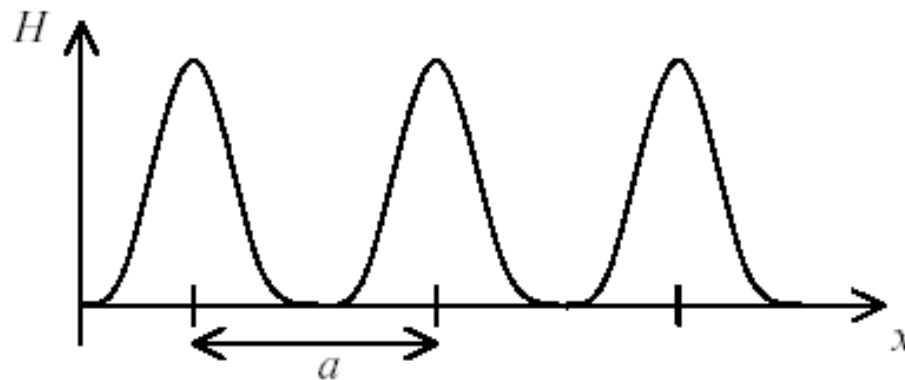
3 Flux-line lattice in type-II superconductor

Magnetization in type-II superconductor



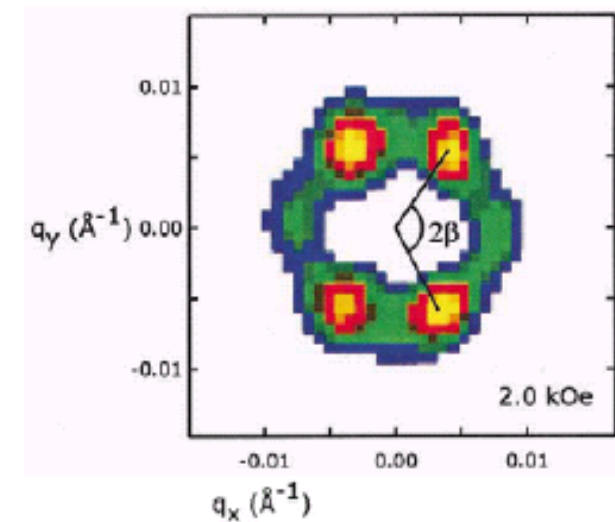
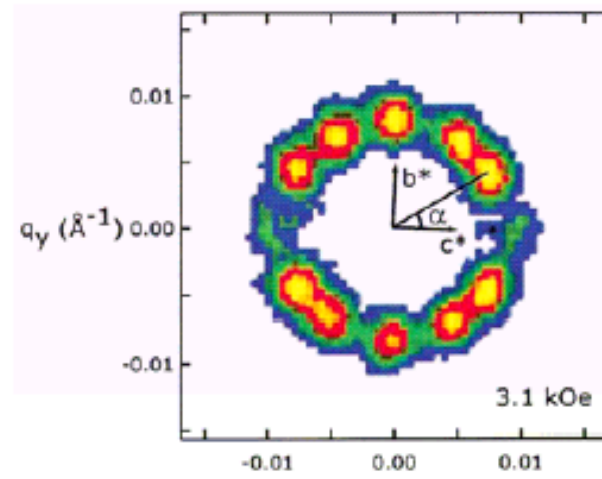
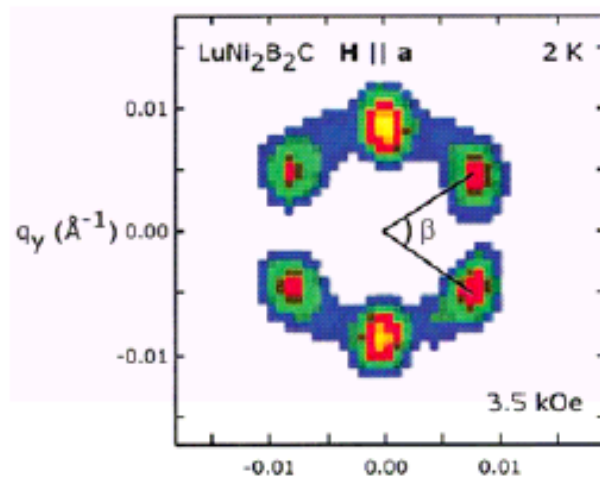
- $0 < H < H_{C1}$: magnetic field completely screened out of superconductor by shielding currents flowing around the perimeter ("Meissner effect")
- $H_{C1} < H < H_{C2}$: magnetic field penetrates partially into superconductor in the form of "flux lines", which form a regular array ("flux line lattice")





$$a \sim \sqrt{\frac{\Phi_0}{H}} \text{ where } \Phi_0 = \frac{h}{2e} \text{ is the magnetic flux quantum}$$
$$a \sim 500 \text{ \AA} \text{ for } H = 1T$$

The periodic magnetic field distribution generated by the flux line lattice inside the superconductor can be revealed by magnetic neutron scattering. Because the lattice constant a is large, the scattering angle $\Theta \sim \frac{\lambda}{2a}$ is small. Even for cold neutrons ($\lambda \sim 5 \text{ \AA}$) and relatively high magnetic fields ($H \sim 1T$), Θ is less than 1° . In order to obtain the resolution required to separate the Bragg reflections from the unscattered beam, one uses a dedicated Small Angle Neutron Scattering (SANS) diffractometer with a long distance ($>10 \text{ m}$) between sample and detector:



Small angle neutron diffraction patterns from flux lines lattice in a type-II superconductor

Note that the structure of the flux line lattice changes as a function of magnetic field

(M.R. Eskildsen et al., PRL **86**, 320 (2001))



How to distinguish between magnetic and nuclear Neutron Scattering?

- temperature dependence
magnetic Bragg peaks vanish above the magnetic ordering temperature
- magnetic form factor decreases with Q
- polarized neutrons



Magnetic Scattering of Spin-Polarized Neutrons

The most unambiguous method to discriminate between nuclear and magnetic neutron scattering is to keep track of the spin state of the neutron before and after the scattering event. In this case, the magnetic neutron scattering cross section for a collinear magnet can be written as

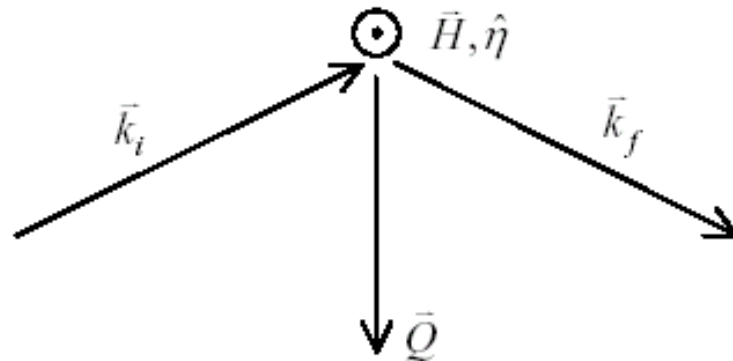
$$\left(\frac{d\sigma}{d\Omega}\right)_M = (\gamma r_0)^2 \left| \langle m_f | \vec{\sigma} \cdot \hat{n}_\perp | m_i \rangle \right|^2 \sum_{\vec{K}_M} \left| F_M(\vec{Q}) \right|^2 \delta(\vec{Q} - \vec{K}_M)$$

The neutron spin quantization axis \hat{z} can be determined by applying a magnetic field \vec{H} at the sample position. Depending on the relative orientation of the vectors \vec{H} , \hat{n} , and \vec{Q} , the scattering process does or does not cause a spin flip of the neutron.



Examples:

- 1 For a **ferromagnet**, the applied field determines both the neutron spin quantization axis and the electron spin direction $\hat{\eta}$, so that $\hat{\eta} \parallel \vec{H} \parallel \hat{z}$. First, consider a situation in which the field is applied perpendicular to the scattering plane:



$$\hat{\eta}_{\perp} = \hat{\eta} = \hat{z}$$

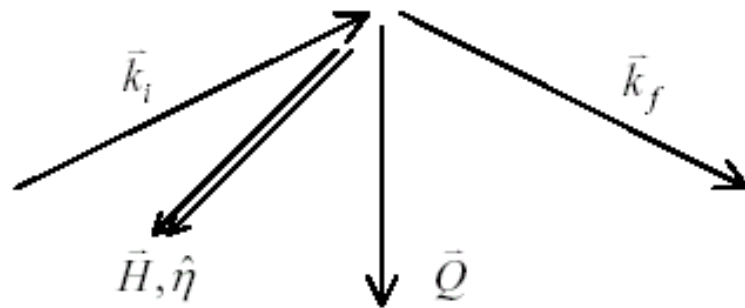
$$\Rightarrow \langle m_f | \vec{\sigma} \cdot \hat{\eta}_{\perp} | m_i \rangle = \langle m_f | \sigma_z | m_i \rangle$$

$$= \begin{cases} +1 & \text{if } m_f = m_i = +1 \\ -1 & \text{if } m_f = m_i = -1 \\ 0 & \text{if } m_f \neq m_i \end{cases}$$

because $|m\rangle$ is an eigenstate of σ_z



The scattering is therefore entirely non-spin-flip. However if the field is applied in a general direction in the scattering plane, both spin-flip and non-spin-flip contributions can be observed:



$$\vec{\sigma} \cdot \hat{\eta}_{\perp} = \sigma_x \eta_{\perp x} + \sigma_y \eta_{\perp y} + \sigma_z \eta_{\perp z}$$

Because σ_x and σ_y can be written as linear superpositions of raising and lowering operators:

$$\sigma_x = \frac{1}{2}(\sigma_+ + \sigma_-)$$

$$\sigma_y = \frac{1}{2i}(\sigma_+ - \sigma_-)$$

the x - and y -components of $\vec{\sigma} \cdot \hat{\eta}_{\perp}$ induce spin flips of the neutron



- 2 For an antiferromagnet, the external field does not affect $\hat{\eta}$, and the neutron spin quantization axis is not necessarily parallel to $\hat{\eta}$. In particular, if \vec{H} is applied parallel to \vec{Q} , the product $\vec{\sigma} \cdot \hat{\eta}_{\perp}$ has no z -component, so that the scattering is entirely spin-flip.



Production of Spin-Polarized Neutrons

In order to apply this method, one needs to produce a beam of neutrons in a single spin state. This can be accomplished by two fundamentally different methods.

1 *Nuclear-magnetic interference effect*

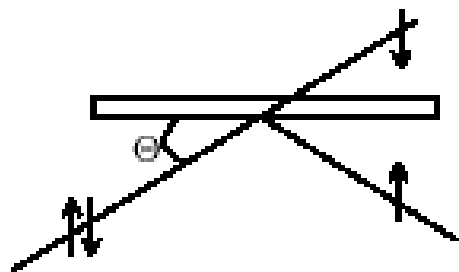
If $\vec{K}_M = \vec{K}_N$, both magnetic and nuclear scattering contribute to a Bragg reflection. Take for example a ferromagnet with one atom per unit cell; with a field applied perpendicular to \vec{Q} as in the example above:

$$\begin{aligned}
 \frac{d\sigma}{d\Omega} &= \sum_{\vec{K}} \left| b + r_0 \gamma f(\vec{Q}) \underbrace{\langle m_f | \vec{\sigma} \cdot \hat{\eta}_{\perp} | m_i \rangle}_{\substack{=+1 \text{ for spin up} \\ =-1 \text{ for spin down}}} \right|^2 \delta(\vec{Q} - \vec{K}) \\
 &= \sum_{\vec{K}} \left(b^2 + |r_0 \gamma f(\vec{Q})|^2 \pm b \gamma r_0 f(\vec{Q}) \right) \delta(\vec{Q} - \vec{K}) \\
 &= \sum_{\vec{K}} \left| b \pm \gamma r_0 f(\vec{Q}) \right|^2 \delta(\vec{Q} - \vec{K})
 \end{aligned}$$

The scattering cross section is thus **different** for the two different spin states of the neutron. This difference can be exploited to generate a spin-polarized neutron beam. For the (100) reflection of the ferromagnet Cu_2MnAl (“Heusler alloy”), $b \approx r_0 \gamma f(\vec{Q})$ so that **only** spin-up neutrons are Bragg reflected:

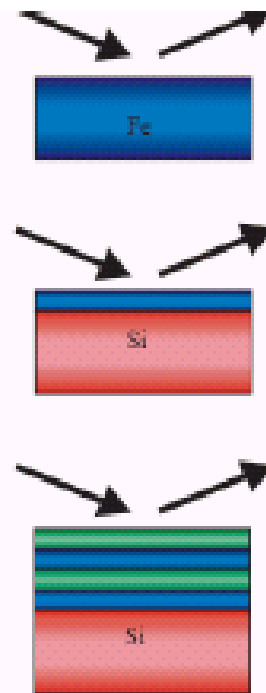
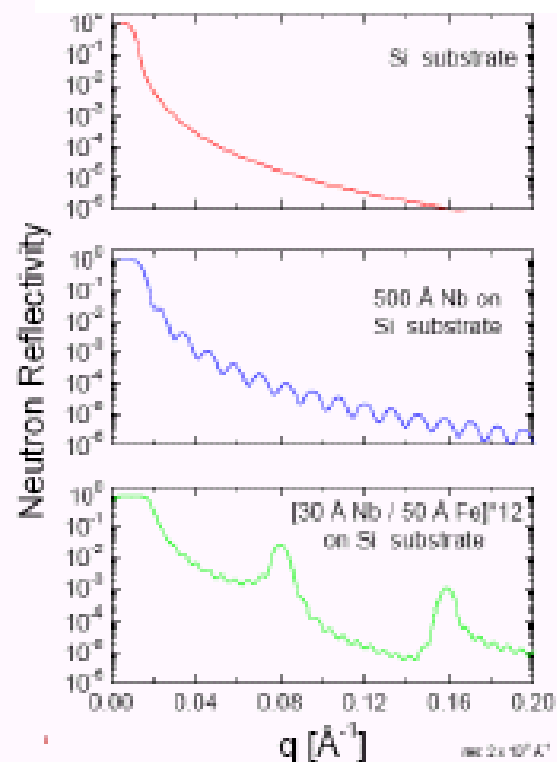


For $\Theta_{C\downarrow} < \Theta < \Theta_{C\uparrow}$, only spin-up neutrons are reflected:

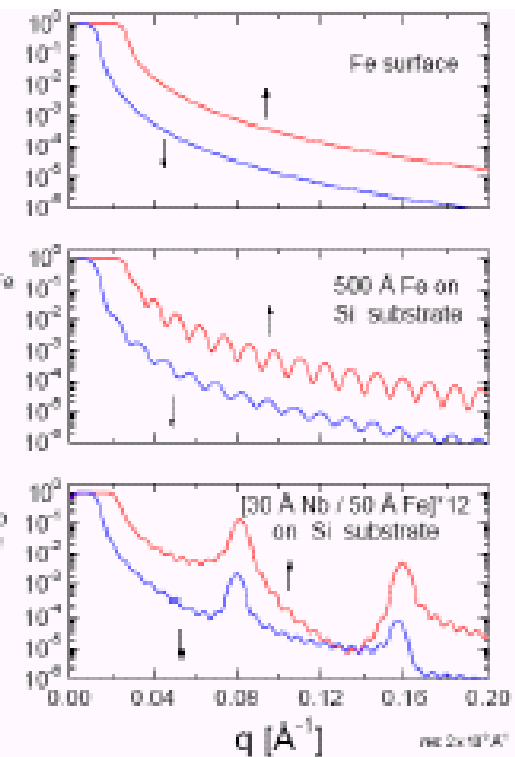


ferromagnetic mirror

nonmagnetic films



magnetic films

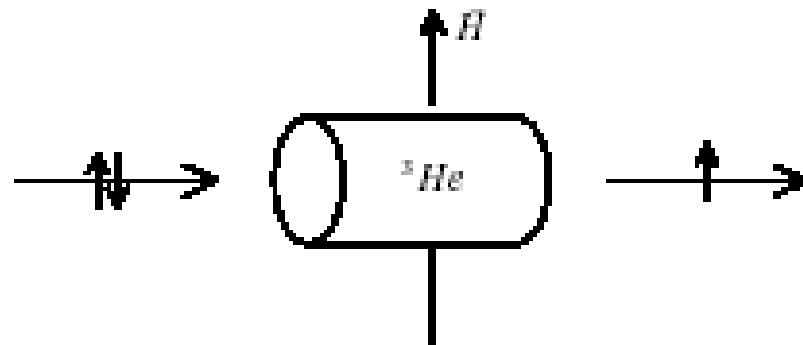


Neutron reflectivity from nonmagnetic and magnetic films

<http://www.ornl.gov/council/02presentations/klose.pdf>



2 A method that has been developed in the past few years is the **neutron spin filter**. A spin-polarized ^3He gas absorbs neutrons through the reaction $n + ^3\text{He} \rightarrow ^4\text{He} (S = 0) \rightarrow p + ^3\text{H}$.



Because the reaction proceeds via an intermediate state with $S = 0$, the absorption cross section for neutrons with spins antiparallel to those of the ^3He nuclei is therefore about three orders of magnitude larger than if neutron and ^3He spins are parallel.



Inelastic Neutron Scattering

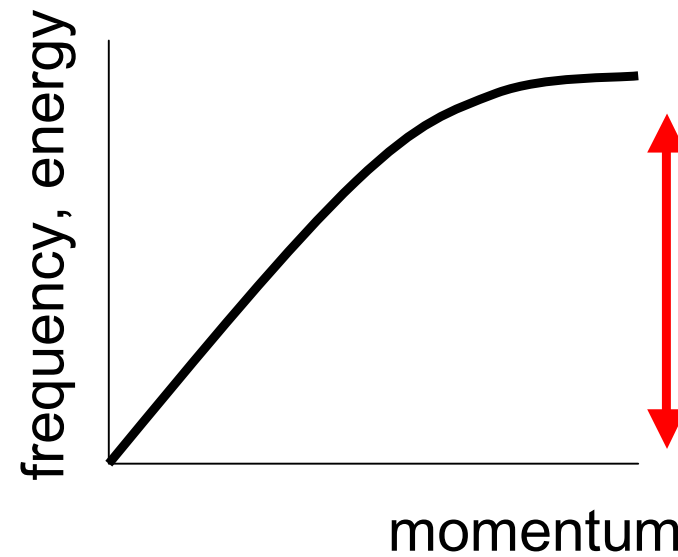


Heisenberg Hamiltonian:

$$H = \sum_{ij} J \mathbf{S}_i \cdot \mathbf{S}_j$$

J = exchange interaction

magnon dispersion



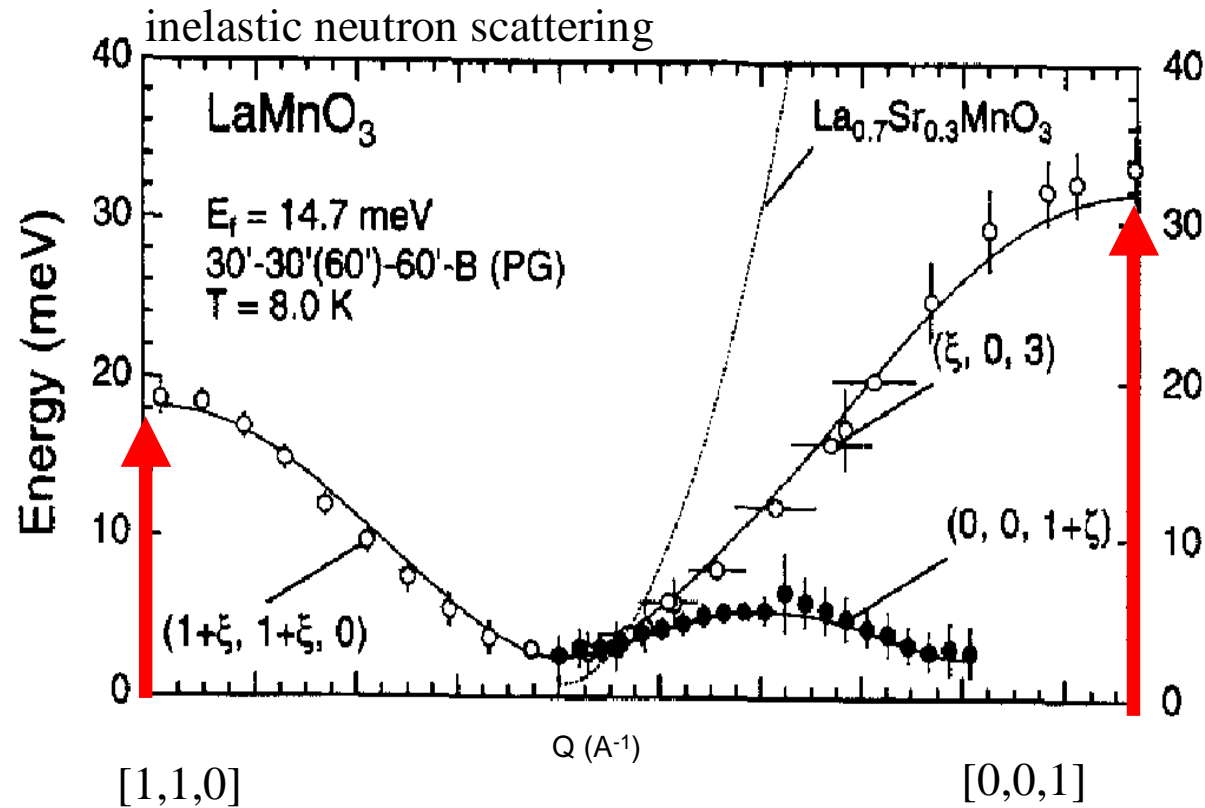
band width $\propto J$



Magnon Dispersion Relation in LaMnO_3



MAX-PLANCK-GESELLSCHAFT



K. Hirota *et al.*, Physica B **237**, 36 (1997)



TRISP Spectrometer at FRM-II



MAX-PLANCK-GESELLSCHAFT

