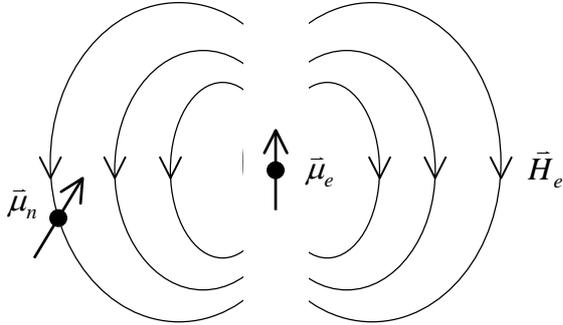


Magnetic Neutron Scattering and Spin-Polarized Neutrons

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-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:

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

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):

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

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

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

Use trick to simplify this expression:

$$\int \frac{d\vec{p}}{|\vec{p}|^2} e^{i\vec{p}\cdot\vec{r}} = 2\pi \int_0^\infty d|\vec{p}| \int_{-1}^1 e^{i|\vec{p}||\vec{r}|\cos\Theta} d(\cos\Theta)$$

$$= 2\pi \int_0^\infty d|\vec{p}| \frac{\sin|\vec{p}||\vec{r}|}{|\vec{p}||\vec{r}|} = \frac{2\pi^2}{|\vec{r}|}$$

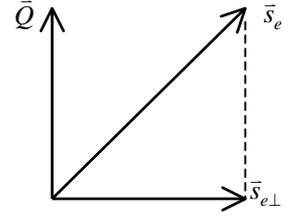
Here, \vec{p} is an auxiliary variable without physical meaning.

$$\nabla \times \left(\vec{s}_e \times \nabla \frac{1}{|\vec{r}|} \right) = \frac{1}{2\pi^2} \int \frac{d\vec{p}}{|\vec{p}|^2} \vec{\nabla} \times (\vec{s}_e \times \vec{\nabla}) e^{i\vec{p}\cdot\vec{r}}$$

$$= \frac{1}{2\pi^2} \int \hat{p} \times (\vec{s}_e \times \hat{p}) e^{i\vec{p}\cdot\vec{r}} d\vec{p}$$

$$\langle \vec{k}_f | \nabla \times \left(\vec{s}_e \times \nabla \frac{1}{|\vec{r}|} \right) | \vec{k}_i \rangle = \frac{1}{2\pi^2} \int d\vec{r} e^{-i\vec{Q}\cdot\vec{r}} \int d\vec{p} \hat{p} \times (\vec{s}_e \times \hat{p}) e^{i\vec{p}\cdot\vec{r}}$$

$$= 4\pi \underbrace{\hat{Q} \times (\vec{s}_e \times \hat{Q})}_{\equiv \vec{s}_{e\perp}}$$



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

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

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

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \left| \langle m_f | \vec{\sigma} \cdot \vec{s}_{e\perp} | m_i \rangle \right|^2$$

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

$$\langle m_f | \vec{\sigma} \cdot \vec{s}_{e\perp} | m_i \rangle = s_{e\perp} \langle m_f | \sigma_z | m_i \rangle$$

$$= \begin{cases} s_{e\perp} & \text{if } m_f = m_i \\ 0 & \text{otherwise} \end{cases}$$

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

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

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

$\bar{\mathcal{M}}(\bar{r}) = \mathcal{M}(\bar{r}) \hat{\eta}$ 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:

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

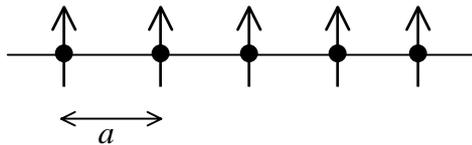
where \bar{K}_M are the magnetic reciprocal lattice vectors and

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

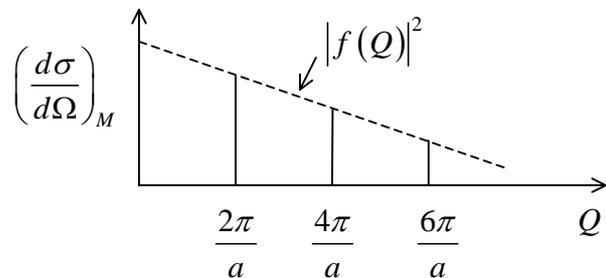
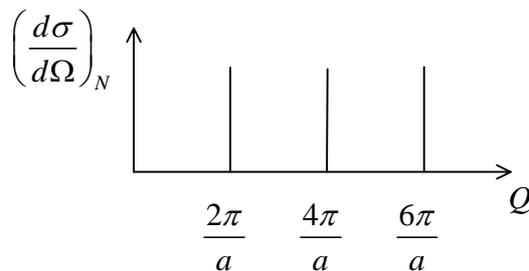
The + or - sign is chosen depending on whether the magnetic moment at lattice site \bar{d} is parallel or antiparallel to $\hat{\eta}$.

Examples:

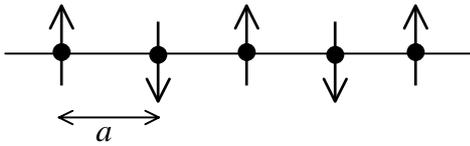
1 One-dimensional ferromagnet



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

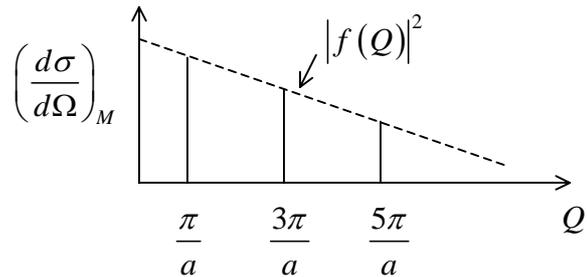
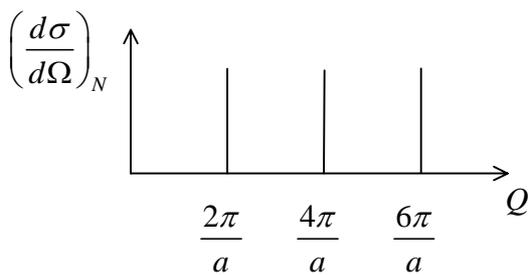


2 One-dimensional antiferromagnet



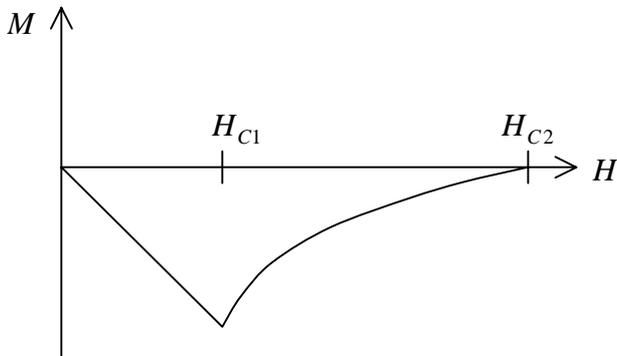
magnetic unit cell twice as large as nuclear unit cell $\Rightarrow K_M = \frac{\pi}{a}n \neq K_N = \frac{2\pi}{a}n$

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

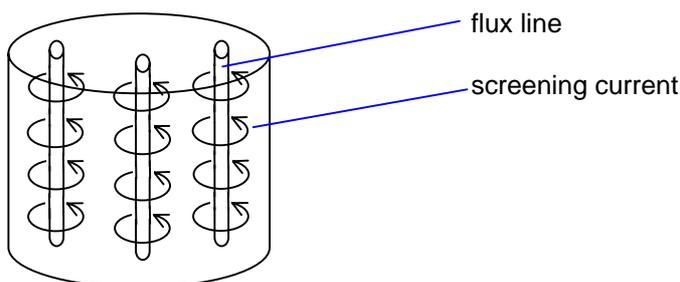


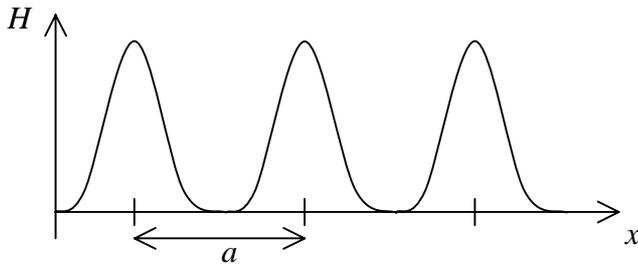
3 Flux-line lattice in type-II superconductor

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")

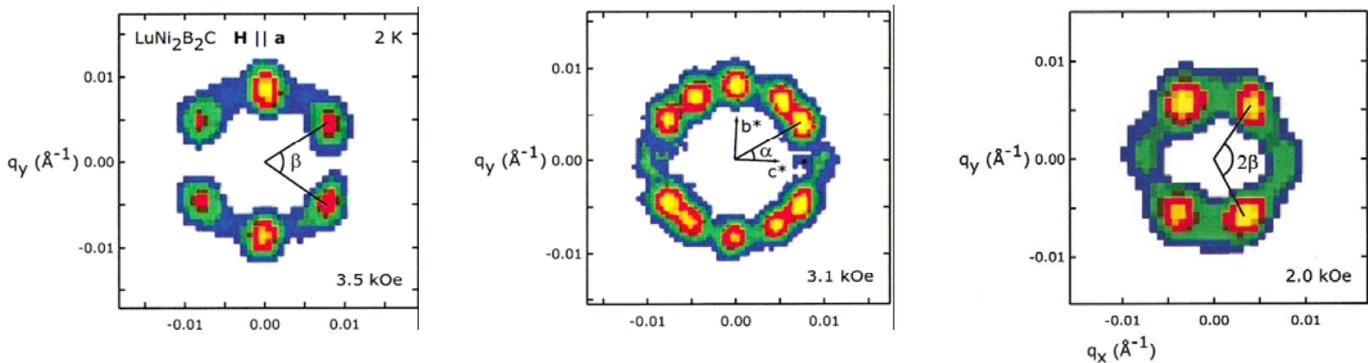




$$a \sim \sqrt{\frac{\Phi_0}{H}} \text{ where } \Phi_0 = \frac{h}{2e} \text{ is the magnetic flux quantum}$$

$$a \sim 500 \text{ \AA} \text{ for } H = 1T$$

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))

As we have seen, nuclear and magnetic neutron scattering have comparable strengths. They can be distinguished experimentally through three different methods:

- 1 The magnetic form factor reduces the intensity of magnetic Bragg reflections with large $|\vec{k}|$, whereas there is no form factor for nuclear scattering. Strong reflections with large $|\vec{k}|$ must therefore be nuclear in origin.
- 2 Magnetic Bragg peaks vanish at the magnetic ordering temperature (the Curie temperature T_C for ferromagnets, or the Néel temperature T_N for antiferromagnets). Nuclear Bragg peaks vanish at the melting temperature, which is typically larger than T_N or T_C .
- 3 The neutron spin operator does not appear in the cross section for coherent nuclear scattering. The neutron spin state is therefore unaffected by nuclear scattering. By contrast, magnetic neutron scattering can be (but does not have to be) associated with a spin-flip of the neutron.

Magnetic Scattering of Spin-Polarized Neutrons

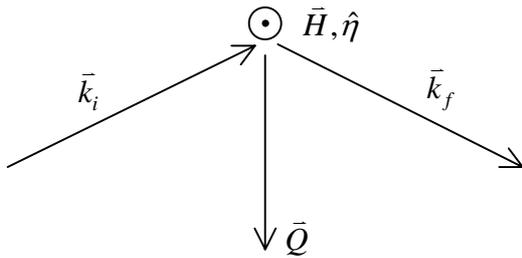
The most unambiguous method to discriminate between nuclear and magnetic neutron scattering is to keep track of the spin state of the neutron before and after the scattering event. In this case, the magnetic neutron scattering cross section for a collinear magnet can be written as

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

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} , $\hat{\eta}$, and \vec{Q} , the scattering process does or does not cause a spin flip of the neutron.

Examples:

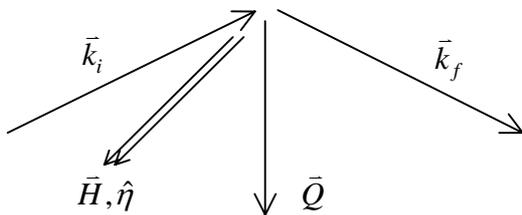
- 1 For a **ferromagnet**, the applied field determines both the neutron spin quantization axis and the electron spin direction $\hat{\eta}$, so that $\hat{\eta} \parallel \vec{H} \parallel \hat{z}$. First, consider a situation in which the field is applied perpendicular to the scattering plane:



$$\begin{aligned} \hat{\eta}_\perp &= \hat{\eta} = \hat{z} \\ \Rightarrow \langle m_f | \vec{\sigma} \cdot \hat{\eta}_\perp | m_i \rangle &= \langle m_f | \vec{\sigma}_z | m_i \rangle \\ &= \begin{cases} +1 & \text{if } m_f = m_i = +1 \\ -1 & \text{if } m_f = m_i = -1 \\ 0 & \text{if } m_f \neq m_i \end{cases} \end{aligned}$$

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

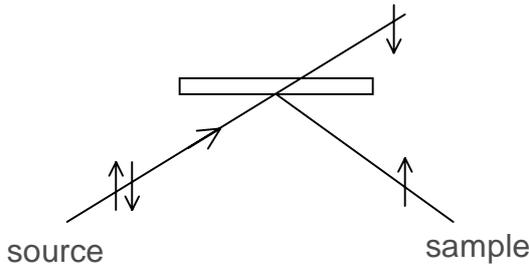
In order to apply this method, one needs to produce a beam of neutrons in a single spin state. This can be accomplished by two fundamentally different methods.

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:

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

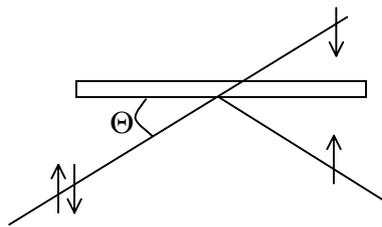
The scattering cross section is thus **different** for the two different spin states of the neutron. This difference can be exploited to generate a spin-polarized neutron beam. For the (100) reflection of the ferromagnet Cu_2MnAl ("Heusler alloy"), $b \approx r_0 \gamma f(\vec{Q})$ so that **only** spin-up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



The beam incident upon the sample is therefore almost perfectly spin-polarized. A variant of this method relies on total external reflection from a ferromagnetic mirror. By analogy to the above argument, the critical angle for a ferromagnetic film can be written as

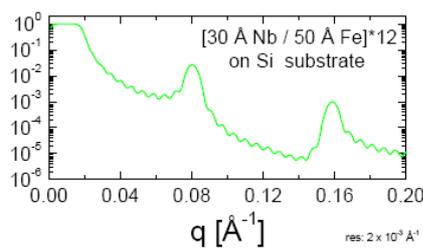
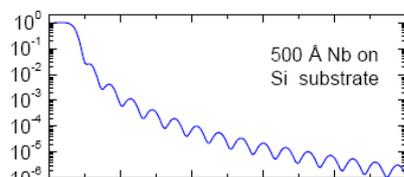
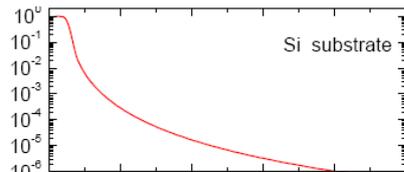
$$\Theta_{c\uparrow\downarrow} \sim \sqrt{2\delta_{\uparrow\downarrow}} = \frac{\sqrt{4\pi p(b \pm \gamma r_0 f)}}{k}$$

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

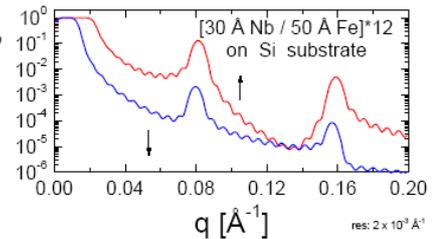
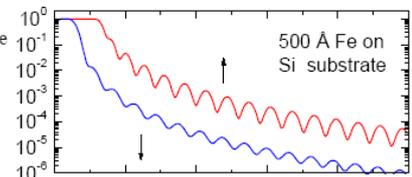
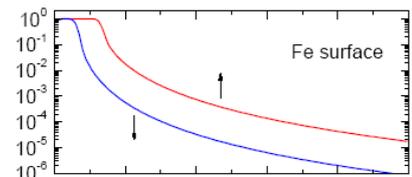


ferromagnetic mirror

nonmagnetic films



magnetic films



Neutron Reflectivity

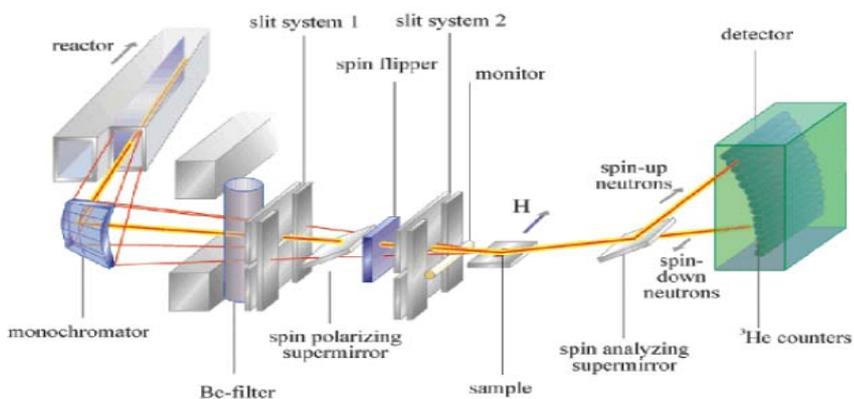
$q [\text{Å}^{-1}]$ res: $2 \times 10^3 \text{ Å}^{-1}$

$q [\text{Å}^{-1}]$ res: $2 \times 10^3 \text{ Å}^{-1}$

Neutron reflectivity from nonmagnetic and magnetic films

<http://www.orau.org/council/02presentations/klose.pdf>

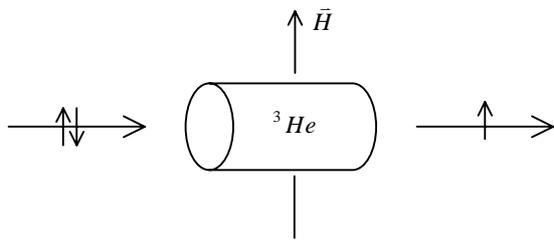
Ferromagnetic “supermirrors” with larger critical angles (see chapter on neutron reflectivity) are commonly used to produce spin-polarized neutrons in modern neutron scattering instruments:



Polarized-beam reflectometer

<http://www.orau.org/council/02presentations/klose.pdf>

2 A method that has been developed in the past few years is the **neutron spin filter**. A spin-polarized ${}^3\text{He}$ 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 ${}^3\text{He}$ nuclei is therefore about three orders of magnitude larger than if neutron and ${}^3\text{He}$ spins are parallel.