Neutron Scattering

Basic properties of neutron and electron

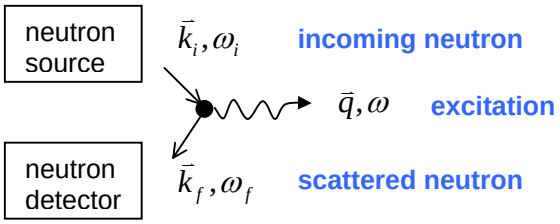
	neutron	electron
mass	$m_n = 1.675 \times 10^{-27}\,kg$	$m_e = 9.109 \times 10^{-31}\,kg$
charge	0	e
spin	$s = ½$	$s = ½$
magnetic dipole moment	$\mu_n = \frac{-e\hbar}{2m_n} g s_n$ with $g_n = 3.826$	$\mu_e = \frac{-e\hbar}{2m_e} g s_e$ with $g_e = 2.0$
energy	$E = \frac{\hbar^2 k^2}{2m_n}$ $\quad k = \frac{2\pi}{\lambda}$ $E[meV] = \frac{81.81}{\lambda^2[Å]}$	$E = \frac{\hbar^2 k^2}{2m_e}$ $E[eV] = \frac{150.26}{\lambda^2[Å]}$
interaction with matter:		
Coulomb interaction	—	✓
strong-force interaction	✓	—
magnetic dipole-dipole interaction	✓	✓

Several salient features are apparent from this table:

– electrons are charged and experience strong, long-range Coulomb interactions in a solid. They therefore typically only penetrate a few atomic layers into the solid. ***Electron scattering*** is therefore a ***surface-sensitive*** probe. Neutrons are uncharged and do not experience Coulomb interaction. The strong-force interaction is naturally strong but very short-range, and the magnetic interaction is long-range but weak. Neutrons therefore penetrate deeply into most materials, so that ***neutron scatterin***g is a ***bulk probe.***

– Electrons with wavelengths comparable to interatomic distances ($\lambda \sim 2Å$) have energies of several tens of electron volts, comparable to energies of plasmons and interband transitions in solids. Electron scattering is therefore well suited as a probe of these ***high-energy excitations***. Neutrons with $\lambda \sim 2Å$ have energies of several tens of meV, comparable to the thermal energies $k_B T$ at room temperature. These so-called "thermal neutrons" are excellent probes of ***low-energy excitations*** such as lattice vibrations and spin waves with energies in the meV range.

Basic idea of neutron scattering experiment:



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

$$\omega = \omega_f - \omega_i = \frac{\hbar}{2m_n}\left(k_f{}^2 - k_i{}^2\right)$$

$\omega = 0$ elastic scattering

$\omega \neq 0$ inelastic scattering

Neutron sources

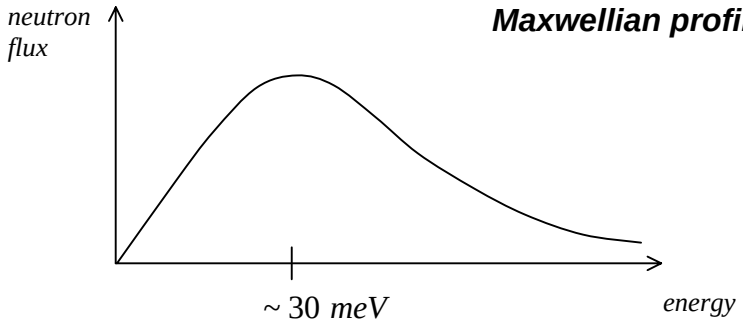
Research reactor

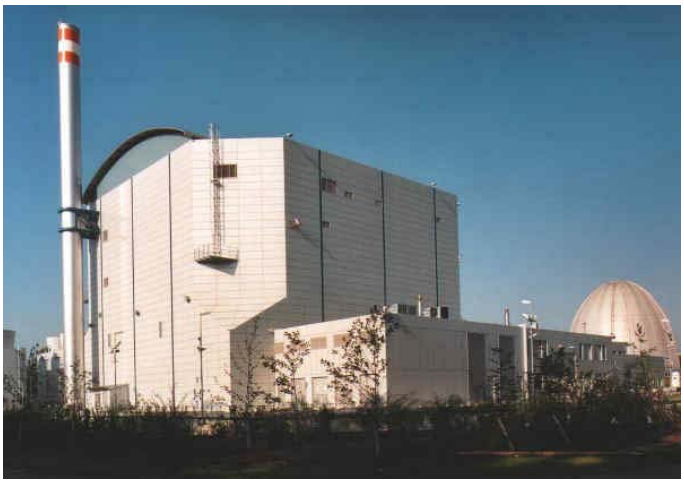
$^{235}U + n \rightarrow A + B + 2.3n$ (A, B: fission fragments)

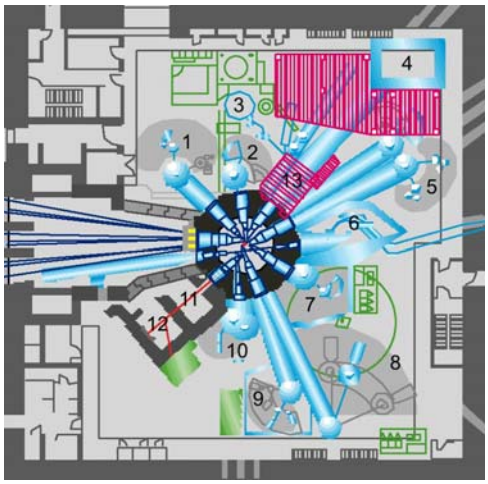
$\Rightarrow$ chain reaction, keeps going by itself until "fuel" (uranium enriched by ^{235}U) is exhausted $\Rightarrow$ source of both ***energy*** (nuclear power reactors) and ***neutrons*** (research reactors)

research reactors optimized for neutron flux $\Rightarrow$ low power

fission reaction most favorable for thermal neutrons $\Rightarrow$ fast neutrons slowed down by "moderator" (H_2O, D_2O)







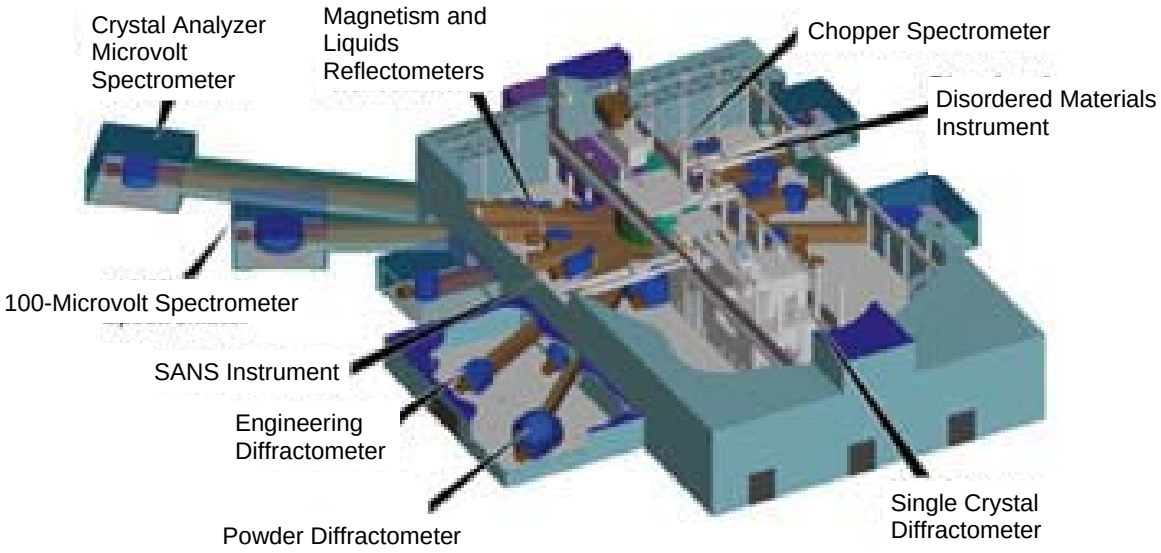
Research reactor FRM-II in Munich, Germany
Outside view of the building (left) and layout of the experimental hall (right). The neutron beam tubes (blue) tap into the flux emitted from the reactor core (center) and guide the neutrons to various neutron scattering instruments. http:///www.frm2.tu-muenchen.de

Spallation source

$p + Hg \rightarrow A + B + xn$

Fragmentation of target atoms induced by high energy proton beam from accelerator.

Typical energy of spallation neutrons >> thermal energy $\Rightarrow$ need to use moderator for scattering experiments.



Layout of the Spallation Neutron Source (SNS) at Oak Ridge National Lab (USA)
Protons are accelerated in the linear accelerator, and various neutron scattering instruments are grouped around the target, where the neutrons are produced in a spallation reaction.
http://www.sns.gov

Neutron detectors

Since neutrons are electrically neutral, they are difficult to detect directly. One therefore converts them into charged particles via a nuclear reaction such as:

$n + {}^3He \rightarrow {}^3H + p$

The protons are collected by a high electric field and converted into electric current.

Another type of neutron detector is based on a gas of $^{10}BF_3$. ^{10}B has a high neutron capture cross section via the nuclear reaction

$n + {}^{10}B \rightarrow {}^7Li + {}^4He$.

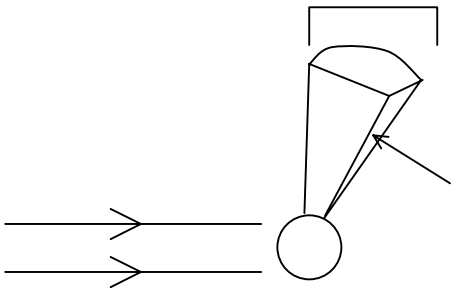
The energetic nuclei produced in this reaction ionize gas molecules, which are again collected by a high electric field.

Interaction of neutrons with matter

	elastic scattering	**inelastic scattering**
strong-force interaction ("nuclear scattering")	position of nuclei in solid (lattice structure)	lattice vibrations (phonons)
magnetic interaction	position and orientation of electronic magnetic moments in solids (ferromagnetism, antiferromagnetism)	spin excitations (magnons, spin waves)

Elastic nuclear scattering

basic quantity: differential cross-section $\frac{d\sigma}{d\Omega}$



solid angle element covered by detector

incident neutron beam, flux $\phi = \frac{\#\text{ of neutrons}}{\text{area} \cdot \text{time}}$

$\frac{d\sigma}{d\Omega}$ = # of neutrons scattered into solid angle element $d\Omega$ per unit time, normalized to incident flux. Material-specific quantity, experimental conditions (size of detector, incident flux) normalized out.

dimensions: $\left[\frac{d\sigma}{d\Omega}\right] = \frac{1}{[\Delta\Omega]\,[t]\,[\phi]} =$ area

dimensionless

calculation of $\frac{d\sigma}{d\Omega}$ through Fermi's Golden Rule:

transition rate $\left(\#\text{ of transitions per unit time}\right): W = \frac{2\pi}{\hbar}\left|\left\langle \vec{k}_f \left| V \right| \vec{k}_i \right\rangle\right|^2 \underbrace{\rho_f\left(E\right)}_{\text{Density of final states}}$

$$\left.\begin{aligned} |k_i\rangle &= \frac{1}{\sqrt{L^3}} e^{i\vec{k}_i\cdot\vec{r}} \\ |k_f\rangle &= \frac{1}{\sqrt{L^3}} e^{i\vec{k}_f\cdot\vec{r}} \end{aligned}\right\}$$ plane waves, normalized to sample size L

$$\rho_f\left(E\right) = \underbrace{\left(\frac{L}{2\pi}\right)^3}_{\text{density of states in } k\text{-space}} \frac{d\vec{k}_f}{dE}$$

$$d\vec{k}_f = k_f{}^2 dk_f d\Omega$$

$$\rho_f\left(E\right) = \left(\frac{L}{2\pi}\right)^3 k_f{}^2 \frac{dk_f}{dE} d\Omega = \left(\frac{L}{2\pi}\right)^3 \frac{m_n k_f}{\hbar^2} d\Omega \qquad \text{with } \frac{dE}{dk_f} = \frac{\hbar^2 k_f}{m_n}$$

incident neutron flux: $\frac{\text{velocity}}{L^3} = \frac{\hbar k_i}{m_n L^3}$

$k_i = k_f$ for elastic scattering

$$\Rightarrow \frac{d\sigma}{d\Omega} = \frac{W}{\text{incident flux}} = \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left|\int V e^{i\left(\vec{k}_i - \vec{k}_f\right)\cdot\vec{r}} d\vec{r}\right|^2$$

$$= \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left|\int V\left(\vec{r}\right) e^{-i\vec{Q}\cdot\vec{r}} d\vec{r}\right|^2 \qquad \text{"Born approximation"}$$

For short range strong force, use approximate interaction potential

$$V\left(\vec{r}\right) = \frac{2\pi\hbar^2}{m_n} b\, \delta\left(\vec{r} - \vec{R}\right)$$

(b: "scattering length"; $\vec{R}$: position of nucleus)

b depends on the details of the nuclear structure and varies greatly (and almost randomly if you don't know a lot about nuclear physics) among elements/isotopes. A table of scattering lengths can be found on the web under http://www.ncnr.nist.gov/resources/n-lengths/

for single nucleus: $\frac{d\sigma}{d\Omega} = |b|^2$

total cross section: $\sigma = \int \frac{d\sigma}{d\Omega} d\Omega = 4\pi b^2$

lattice of nuclei: $V(\vec{r}) = \frac{2\pi\hbar^2}{m_n} \sum_{\vec{R}} b_{\vec{R}}\, \delta(\vec{r} - \vec{R})$ $b_{\vec{R}}$: scattering length of nucleus at lattice site $\vec{R}$

$$\frac{d\sigma}{d\Omega} = \left| \int d\vec{r} \sum_R b_R\, \delta(\vec{r} - \vec{R}) e^{i\vec{Q}\cdot\vec{r}} \right|^2$$

$$= \left| \sum_R b_R\, e^{i\vec{Q}\cdot\vec{R}} \right|^2$$

For most $\vec{Q}$, matrix elements are very small because phase factors contributed by different nuclei cancel out. Matrix element is large only for $\vec{Q} = \vec{K}$ (reciprocal lattice vector, defined as $e^{i\vec{K}\cdot\vec{R}} = 1$).
If this condition is satisfied, the constructive interference leads to a huge enhancement ("Bragg peak") of the scattering rate.

Use relation $\left| \sum_{\vec{k}} e^{i\vec{Q}\cdot\vec{k}} \right|^2 = N \frac{(2\pi)^3}{v_0} \sum_{\vec{k}} \delta(\vec{Q} - \vec{K})$ derived on problem set.

$\vec{K}$: reciprocal lattice vector
v_0: volume of unit cell
N: total # of unit cells

If all nuclei are identical:

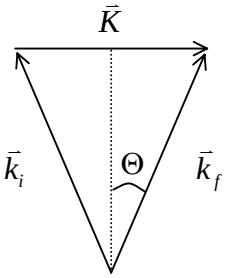
$$\frac{d\sigma}{d\Omega} = b^2 \frac{N(2\pi)^3}{v_o} \sum_{\vec{K}} \delta(\vec{Q} - \vec{K})$$

for unit cell with several atoms, basis vector $\vec{d}$

$$\frac{d\sigma}{d\Omega} = N \frac{(2\pi)^3}{v_0} \sum_{\vec{K}} \delta(\vec{Q} - \vec{K}) \left| F_N(\vec{K}) \right|^2$$

$$F_N(\vec{K}) = \sum_{\vec{d}} e^{i\vec{Q}\cdot\vec{d}} b_{\vec{d}}$$ "nuclear structure factor"

Relationship between reciprocal lattice vector and scattering angle Θ at Bragg peak:



$$\sin\Theta = \frac{K}{2k} = \frac{1}{2} \frac{2\pi n}{d} \frac{\lambda}{2\pi} \Rightarrow 2d \sin\Theta = n\lambda$$ "Bragg's law"

n = integer
d = lattice constant

Collect intensities of all Bragg reflections ⇒ determine lattice structure (crystallography).

This calculation is strictly valid only for a single isotope with nuclear spin $I = 0$. However, almost all elements are found naturally with a distribution of isotopes, most of which have $I \neq 0$. The scattering length b depends on details of the nuclear structure and can vary strongly from one isotope to the other. It also depends on the combined spin of the nucleus and the neutron which can take the values $I + ½$ or $I - ½$. The nuclear spins are randomly oriented in a solid, except at extremely low temperatures. For a given isotope, one therefore has to average over the scattering lengths b_+ for $I + ½$ and b_- for $I - ½$; e.g. for hydrogen ($I = ½$) $b_+ = 10.85\ fm$ and $b_- = -47.50\ fm$, and for deuterium ($I = 1$) $b_+ = 9.53\ fm$ and $b_- = 0.98\ fm$.

The number of states for these two situations is:

$2\left(I + ½\right) = 2I + 2$ for $I + ½$

$2\left(I - ½\right) = 2I$ for $I - ½$

Further, the different naturally occurring isotopes of each element are incorporated randomly in a solid, with no correlations between different lattice positions. Taking these complications into account, the differential cross section for elastic nuclear neutron scattering from a solid containing a single element per unit cell becomes

$$\frac{d\sigma}{d\Omega} = \left\langle \left| \sum_{\vec{R}} b_{\vec{r}} e^{i\vec{Q}\cdot\vec{R}} \right|^2 \right\rangle = \sum_{\vec{R}\vec{R}'} \left\langle b_{\vec{R}}\, b_{\vec{R}'} \right\rangle e^{i\vec{Q}\cdot\left(\vec{R}-\vec{R}'\right)}$$

where $\langle ... \rangle$ denotes the average over isotopes and nuclear spin states. Since there are no correlations between b-values for different nuclei, we have

$$\left\langle b_{\vec{R}}\, b_{\vec{R}'} \right\rangle = \left\langle b \right\rangle^2 = \sum_{\xi} c_\xi \left[\frac{2I_\xi + 2}{4I_\xi + 2}\, b_\xi^+ + \frac{2I_\xi}{4I_\xi + 2}\, b_\xi^- \right] \quad \text{for } \vec{R} = \vec{R}'$$

$$\left\langle b_{\vec{R}}\, b_{\vec{R}'} \right\rangle = \left\langle b^2 \right\rangle = \sum_{\xi} c_\xi \left[\frac{2I_\xi + 2}{4I_\xi + 2} \left(b_\xi^+\right)^2 + \frac{2I_\xi}{4I_\xi + 2} \left(b_\xi^-\right)^2 \right] \quad \text{for } \vec{R} \neq \vec{R}'$$

where $\langle ... \rangle$ denotes the different naturally occurring isotopes and c_ξ their relative abundance.

Splitting the sum into two terms, we have

$$\frac{d\sigma}{d\Omega} = \sum_{\substack{\vec{R},\vec{R}' \\ \vec{R}\neq\vec{R}'}} \langle b \rangle^2 e^{i\vec{Q}\cdot\left(\vec{R}-\vec{R}'\right)} + \sum_{\vec{R}} \left\langle b^2 \right\rangle$$

$$= \sum_{\vec{R},\vec{R}'} \langle b \rangle^2 e^{i\vec{Q}\cdot\left(\vec{R}-\vec{R}'\right)} + \sum_{\vec{R}} \left[\left\langle b^2 \right\rangle - \langle b \rangle^2 \right]$$

where $\sum_{\vec{R}} \langle b \rangle^2$ has been added to the first term and subtracted from the second term in the sum. With the restriction $\vec{R} \neq \vec{R}'$ thus removed, the first term is identical to the result obtained above for the case of a single element per unit cell, except that b^2 was replaced by $\langle b \rangle^2$.

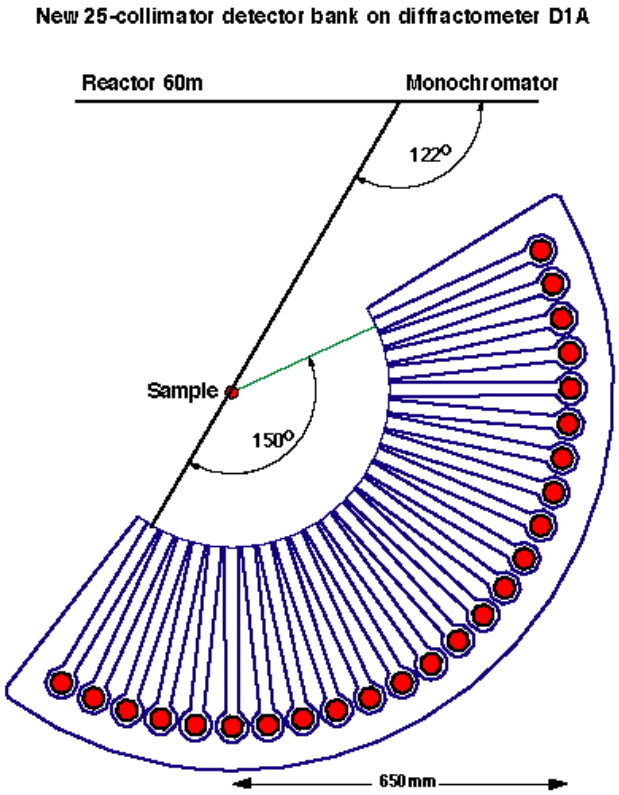
Therefore $\frac{d\sigma}{d\Omega} = \underbrace{N\frac{(2\pi)^3}{V_0}\langle b\rangle^2 \sum_{\vec{K}} \delta\left(\vec{Q}-\vec{K}\right)}_{\text{"coherent" scattering}} + \underbrace{N\left[\langle b^2\rangle - \langle b\rangle^2\right]}_{\text{"incoherent" scattering}}$

Again, the chemical properties of the elements are dictated entirely by the number of protons and electrons and completely independent of the number of neutrons per nucleus (isotope) or the nuclear spin. The latter quantities are therefore distributed completely randomly in the solid, and the corresponding "incoherent" term in the scattering cross section has no $\vec{Q}$-dependence.

Both elastic nuclear neutron scattering (which is sensitive to the positions of the nuclei) and x-ray diffraction (which is sensitive to the positions of the electrons) are powerful probes of the ***lattice structure*** of materials. The overall magnitudes of the scattering cross sections for x-rays and neutrons are similar, because $r_0 \sim 10^{-15} m \sim b_{\xi}$. But there are also substantial differences in detail, so that the two techniques often yield complementary information:

– The scattering length b for nuclear neutron scattering varies erratically from one element to the other. By contrast, the analogue for x-rays increases systematically with the number of electrons, Z. The x-ray cross section is therefore dominated by heavy elements. Neutrons have a greater sensitivity for light elements such as hydrogen and oxygen. The differences in the scattering lengths of different isotopes (particularly hydrogen and deuterium) can also be exploited in neutron experiments with isotopic ***labelling*** (see below).
– The nucleus is much smaller than typical neutron wave lengths, but the dimensions of electron clouds of atoms are comparable to typical x-ray wavelengths. Interference effects between x-rays scattered from different regions inside the same atom can therefore not be neglected. The x-ray scattering cross section thus contains a "form factor" $f\left(\vec{Q}\right) = \int d\vec{r}\,\rho\left(\vec{r}\right)e^{i\vec{Q}\cdot\vec{r}}$ that depends on the electron charge distribution $\rho\left(\vec{r}\right)$ inside each atom. A similar form factor appears in the cross section for magnetic neutron scattering to be discussed later.
– Incoherent scattering due to randomly distributed isotopes and nuclear spins does not appear in the x-ray cross section. Conversely, thermal diffuse scattering associated with the creation and annihilation of lattice vibrations, which invariably appears in x-ray scattering experiments at nonzero temperature, can be eliminated easily from elastic nuclear neutron scattering experiments (see below).
– Neutron scattering lengths can be both negative and positive, depending on the phase shifts the neutron experiences upon scattering from the nucleus. The corresponding quantity for x-rays is always positive. This has important consequences for the reflection of neutrons from surfaces, as discussed later.

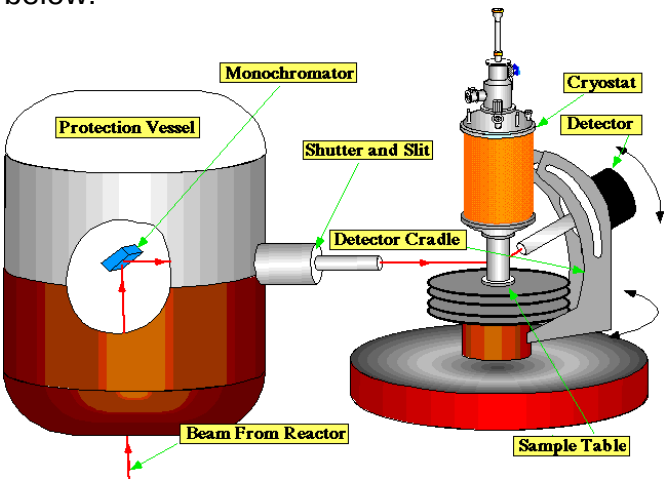
The details of elastic nuclear neutron scattering experiments depend on whether the sample is a powder consisting of many randomly oriented, typically micron-sized crystallites, of a single crystal. A typical ***powder diffractometer*** is shown below:



Layout of the powder diffractometer D1A at the Institut Laue-Langevin in Grenoble, France
http://www.ill.fr/

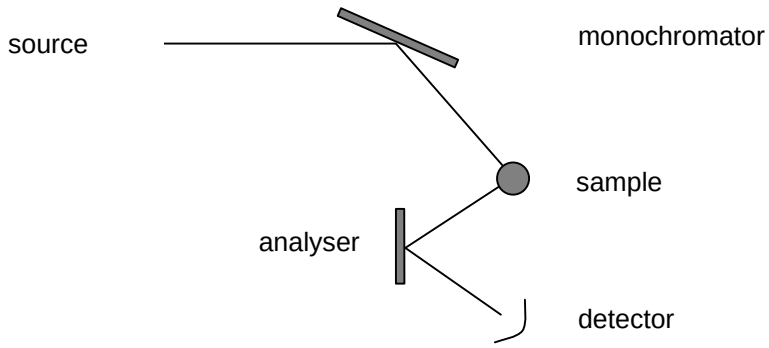
A monochromator selects a single energy E_i from the thermal spectrum emitted by the neutron source. One typically uses a Bragg reflection of a single crystal whose orientation determines E_i. A common monochromator is the (002) reflection of pyrolytic graphite, a material in which the graphite planes are well aligned along the c-axis perpendicular to the planes, but randomly oriented within the planes. A relatively small “mosaic spread” of the orientation along the c-axis determines the spread of wavelengths transmitted by the monochromator, ΔE_i, and hence the instrumental resolution of the wave vector transfer, ΔQ.

A multidetector allows the simultaneous detection of multiple Bragg reflections. Neutron powder diffraction is further discussed in the exercise session. If the sample is a single crystal, the sample orientation has to be adjustable in order to satisfy the Bragg condition, as shown in the ***single-crystal diffractometer*** below:



Single-crystal diffractometer D15 at the Institut Laue-Langevin in Grenoble, France
http://www.ill.fr

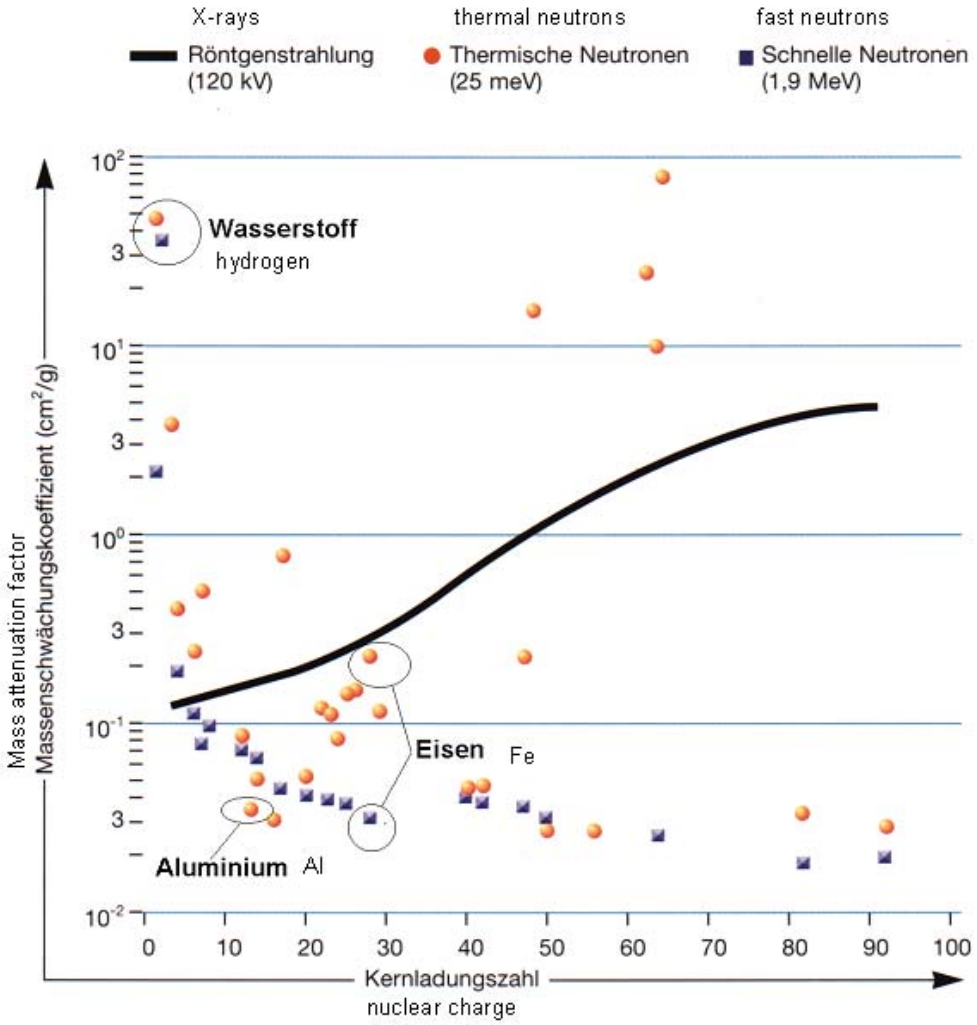
In both powder and single-crystal diffractometers, the detector collects both elastically and inelastically scattered neutrons. This is acceptable for most elastic neutron scattering experiments, because the background from inelastically scattered neutrons is not very large. To suppress this background, one can use an ***analyzer crystal*** set for an energy equal to that of the incident beam:



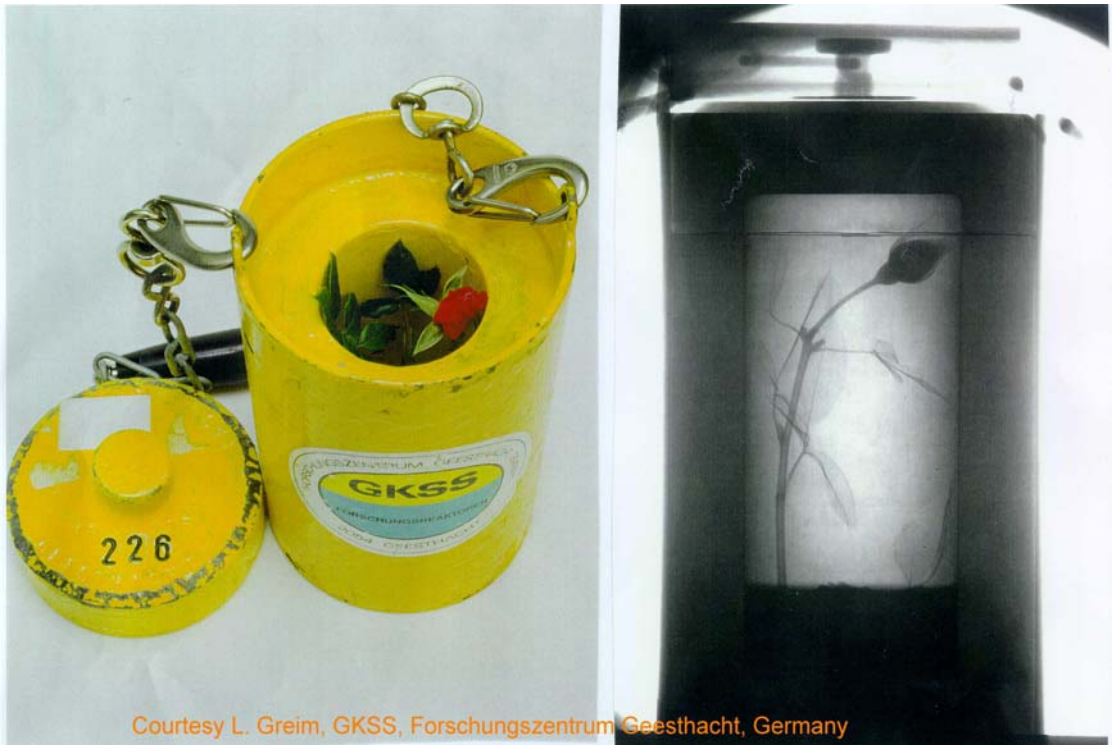
The energy spread ΔE_f transmitted by the analyser crystal is typically around $0.1E_f \sim 1meV$, that is, much smaller than typical phonon energies. The analyser crystal therefore eliminates thermal diffuse scattering very efficiently. This is much more difficult for x-ray diffraction, because typical x-ray photon energies are of order $10keV$, much larger than typical phonon energies.

Neutron Radiography

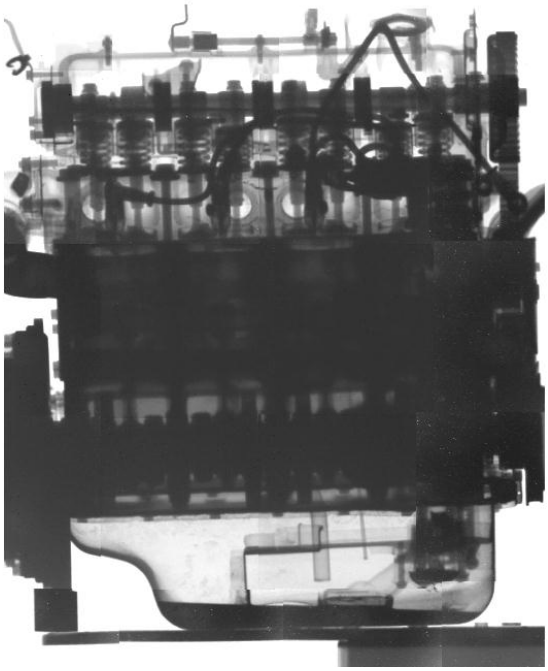
Coherent and incoherent scattering of neutrons also lead to an attenuation of the direct beam, which can be measured and analysed. Another factor contributing to the attenuation of the neutron beam in the sample is ***neutron absorption*** due to the capture of neutrons by certain isotopes. Both effects are combined in the attenuation coefficient per unit mass, μ / ρ:



Some elements such as boron $(Z = 5)$ cadmium $(Z = 48)$, or gadolinium $(Z = 64)$ have large attenuation coefficients due to absorption. They can be used as radiation shielding at neutron sources. Note that hydrogen also has a very large attenuation coefficient due to its large incoherent scattering cross section. This is demonstrated in the following radiograph of a rose in a lead container:



Note that the x-ray attenuation coefficient of the rose is negligible compared to that of the container. The high sensitivity of neutron radiography to hydrogen can be exploited in unique technological applications, e.g. by visualizing the flow of oil in a running motor:



Neutron radiograph of a running car engine
http://www1.physik.tu-muenchen.de/antares/