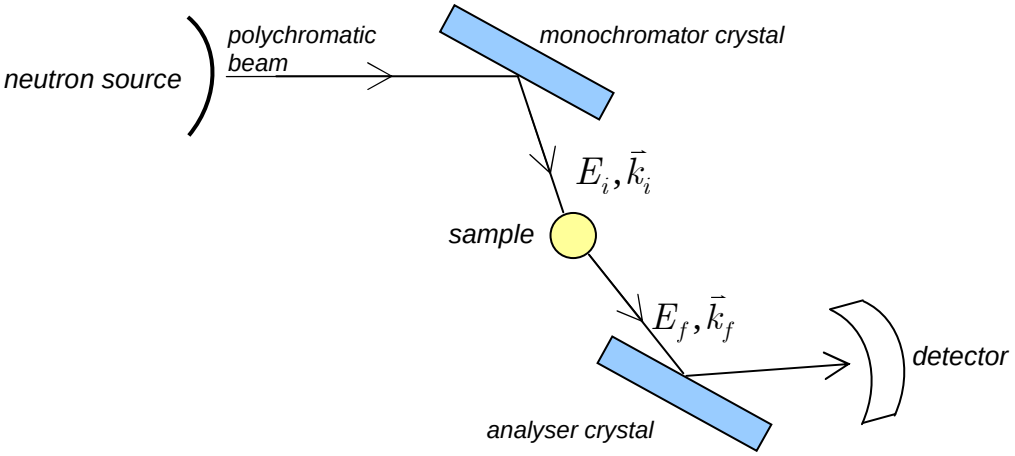
Inelastic Neutron Scattering

In an ***elastic*** neutron scattering event, a momentum $\hbar\vec{Q} = \hbar\left(\vec{k}_f - \vec{k}_i\right)$ is transferred from the neutron to the sample. This leads to a minuscule translation of the entire sample, but the internal state of the sample remains unchanged.

Inelastic scattering of neutrons creates or annihilates an excitation inside the sample, so that both the energy of the neutron and the internal state of the sample (here denoted as λ) is modified. Experimentally, one has to keep track not only of the ***flight direction*** of the scattered neutron but also of its ***energy***.

This can be experimentally accomplished by the following methods:

1 — Triple-axis spectrometer
(commonly used at steady-state neutron sources)



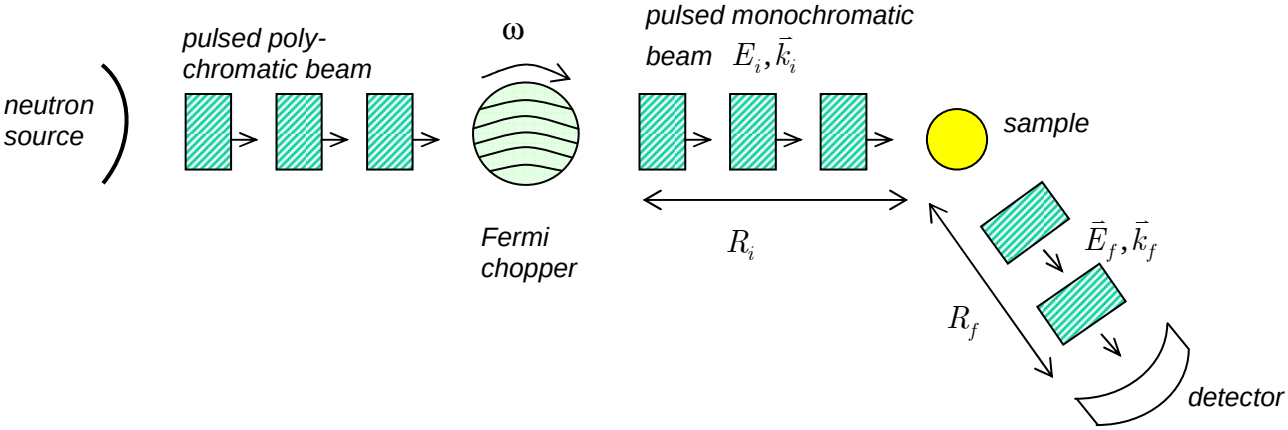
Bragg scattering from analyzer selects scattered neutrons with unique energy and momentum.

Fig. 1

Triple Axis Spectrometer

TRISP at FRM-II
http://www.fkf.mpg.de/keimer/groups/frm/index.html

2 — Time-of-flight spectrometer
(commonly used at pulsed neutron sources)



The incident neutron energy is selected by the rotation frequency ω of the Fermi chopper (which consists of curved neutron-absorbing blades) and its phase delay relative to the source emission. The final energy can be computed from the time of arrival of the scattered neutron pulse at the detector.

$$t = \frac{R_i}{v_i} + \frac{R_f}{v_f} \Rightarrow v_f = \frac{R_f}{t - R_i / v_i}$$

$$E_f = \frac{1}{2} m_n {v_f}^2$$

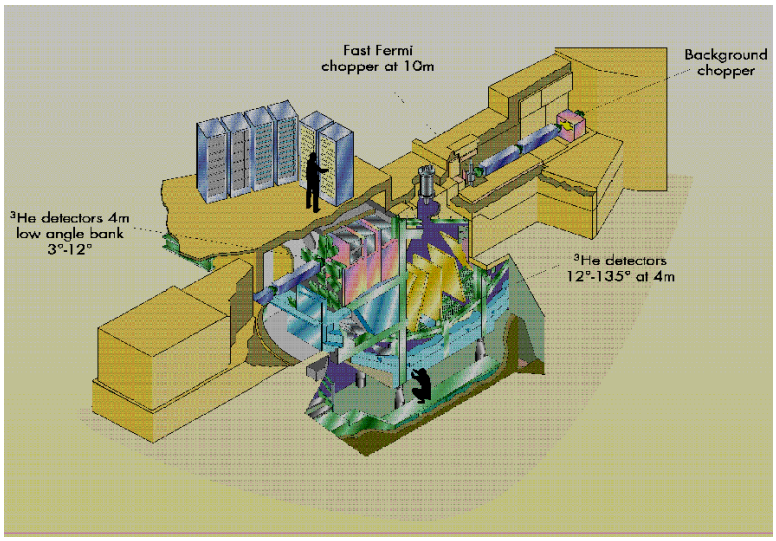
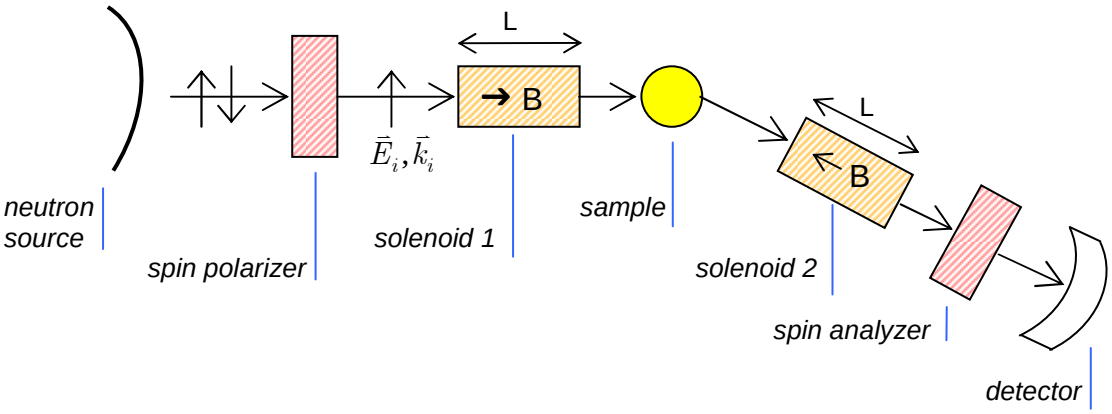


Fig. 2

Time-of-flight spectrometer

MARI at ISIS
http://www.isis.stfc.ac.uk/instruments/mari/

3 — Spin-echo spectrometer



The magnetic fields B in the two solenoids are equal in magnitude but opposite in direction. In the magnetic field, the neutron spins undergo Larmor precessions with frequency

$$\omega_L = \frac{\gamma \mu_N B}{\hbar}.$$

The phase angle of the neutron spins after the first solenoid is then

$$\phi_i = \frac{\omega_L L}{v_i}$$

If the beam is not perfectly monochromatic (i.e. there is a spread of initial neutron velocities v_i), the beam will be depolarized at the sample position. If the scattering at the sample is perfectly elastic, however, every neutron will reverse its Larmor precessions in the second solenoid, and the beam polarization will be fully restored at the analyzer. If the scattering is inelastic, the final beam polarization is reduced:

$$\Delta\Phi = \Phi_i - \Phi_f = \omega_L L\left(\frac{1}{v_i} - \frac{1}{v_f}\right) = \omega_L L\left(\frac{1}{v_i} - \frac{1}{v_i + \Delta v}\right) \approx \frac{\omega_L L}{{v_i}^2}\Delta v \text{ for } \Delta v \ll v_i$$

energy transfer $\hbar\omega = \frac{m_n}{2}\left({v_f}^2 - {v_i}^2\right) \approx m_n v_i \Delta v \Rightarrow \Delta\Phi \approx \frac{\hbar\omega_L L}{m_n {v_i}^3}\omega \equiv \omega\tau_{SE}$ τ_{SE} = spin echo time

The net beam polarization determined by the analyzer is thus a measure of the inelasticity of the scattering process:
$\langle \cos\Delta\Phi \rangle = \int d\omega S(Q,\omega)\cos\omega\tau_{SE} = S(Q,\tau_{SE})$ where $S(Q,\tau_{SE})$ is the Fourier transform of the scattering probability $S(Q,\omega)$.

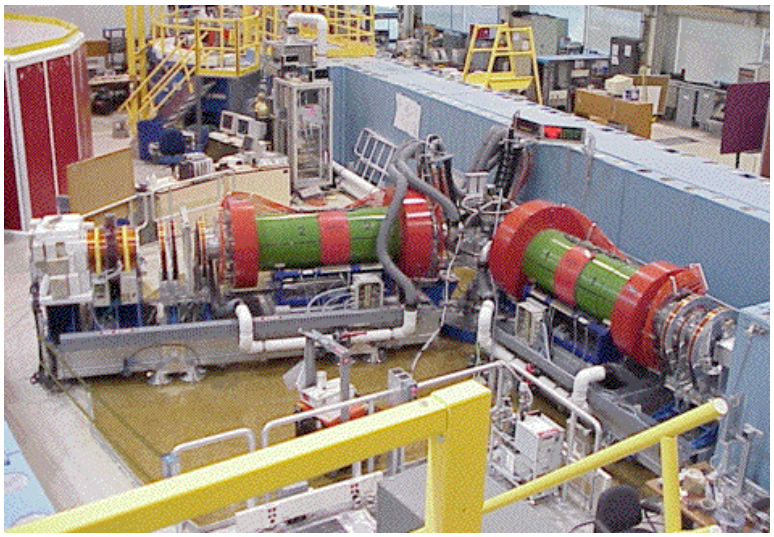


Fig. 3

Neutron spin echo spectrometer

NSE at NIST
http://www.ncnr.nist.gov/instruments/nse/NSE_70deg_20010226.png

For a quantitative description of inelastic neutron scattering, one writes down a ***double-differential*** scattering cross section as follows:

$$\frac{d^2\sigma}{d\Omega\, dE_f} = \frac{k_f}{k_i}\left(\frac{m}{2\pi\hbar^2}\right)^2 \left|\left\langle \vec{k}_f m_f \lambda_f \left| V \right| \vec{k}_i m_i \lambda_i \right\rangle\right|^2 \delta\left(E_{\lambda i} - E_{\lambda f} - \hbar\omega\right)$$

where $\hbar\omega$ is the difference between final and incident neutron energies. The derivation uses Fermi's Golden Rule, as in the case of elastic scattering, and the δ-function guarantees energy conservation in the scattering event. Specializing to nuclear scattering, which does not affect the spin state $|m\rangle$ of the neutron, we obtain

$$\frac{d^2\sigma}{d\Omega\, dE_f} = \frac{k_f}{k_i}\left|\sum_{\vec{R}} b_{\vec{R}} \left\langle \lambda_f \left| e^{i\vec{Q}\cdot\vec{R}} \right| \lambda_i \right\rangle\right|^2 \delta\left(E_{\lambda i} - E_{\lambda f} - \hbar\omega\right) \text{ with}$$

$\left|\vec{k}_i\right\rangle = e^{i\vec{k}_i\cdot\vec{r}}$ and $\left|\vec{k}_f\right\rangle = e^{i\vec{k}_f\cdot\vec{r}}$

We first evaluate this expression for a gas of free nuclei of mass M and initial wave vector $\vec{\xi}_i$, so that

$\left|\lambda_i\right\rangle = \frac{1}{\sqrt{V}}\, e^{i\vec{\xi}_i\cdot\vec{R}}$ and $E_{\lambda i} = \frac{\hbar^2 \xi_i^{\,2}}{2M}$ where V is the volume of the sample.

The matrix element becomes $\left\langle \lambda_f \left| e^{i\vec{Q}\cdot\vec{R}} \right| \lambda_i \right\rangle = \frac{1}{V}\int d\vec{R}\, e^{i\left(\vec{\xi}_i - \vec{\xi}_f + \vec{Q}\right)\cdot\vec{R}}$

This integral is zero unless $\vec{Q} = \vec{k}_f - \vec{k}_i = \vec{\xi}_f - \vec{\xi}_i$ (momentum conservation).

Plugging this expression into the condition for energy conservation, one obtains

$$E_{\lambda f} - E_{\lambda i} = \frac{\hbar^2}{2M}\left(\vec{\xi}_f^{\,2} - \vec{\xi}_i^{\,2}\right) = \frac{\hbar^2}{2M}\left(\vec{Q}^2 + 2\vec{Q}\cdot\vec{\xi}_i\right).$$

We need to sum the matrix elements in the double-differential cross section over all final states consistent with energy and momentum conservation, then thermally average over initial states. The sum over final states can be written as

$$\sum_{\lambda_f}\left|\sum_{\vec{R}}\left\langle \lambda_f \left| e^{i\vec{Q}\cdot\vec{R}} \right| \lambda_i \right\rangle\right|^2 = \sum_{\lambda_f}\sum_{\vec{R}\vec{R}'}\left\langle \lambda_i \left| e^{i\vec{Q}\cdot\vec{R}} \right| \lambda_f \right\rangle\left\langle \lambda_f \left| e^{-i\vec{Q}\cdot\vec{R}'} \right| \lambda_i \right\rangle$$

$$= \sum_{\bar{R}\bar{R}'} \left\langle \lambda_f \left| e^{i\bar{Q}(\bar{R}-\bar{R}')} \right| \lambda_i \right\rangle \quad \text{for complete set of states } |\lambda_f\rangle$$

$$= \sum_{\bar{R}\bar{R}'} \delta_{\bar{R}\bar{R}'} \text{ for independent nuclei } \bar{R},\ \bar{R}'$$

$$= N$$

If one takes an average over an ensemble of nuclei with a thermal distribution of initial wave vectors, one obtains the cross section for neutron scattering from an ***ideal gas:***

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f N}{k_i} b^2 \underbrace{\left\langle \delta\left[\hbar\omega - \frac{\hbar^2}{2M}\left(Q^2 + 2\bar{Q}\cdot\bar{\xi}_i\right)\right]\right\rangle}_{\equiv S(\bar{Q},\omega)}$$

"dynamical structure factor"

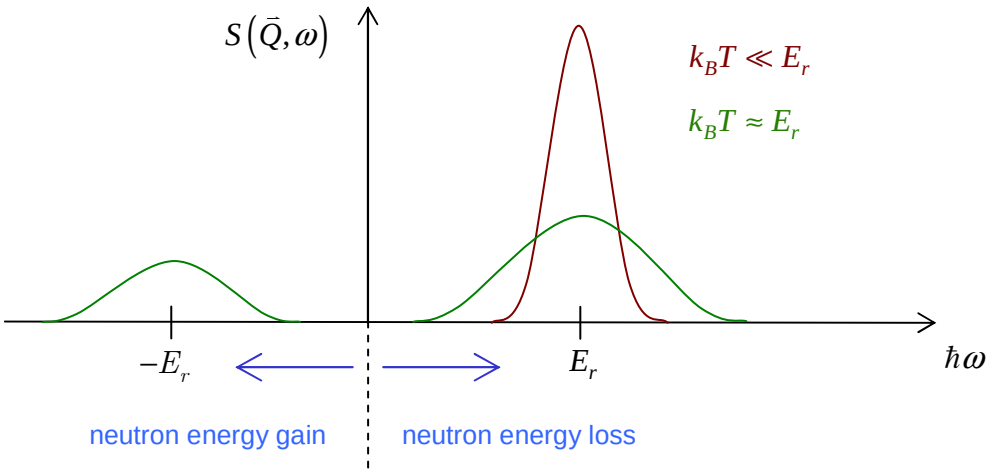
$$S(\bar{Q},\omega) = \frac{\int d\bar{\xi}_i\ e^{-\hbar^2\bar{\xi}_i^2/2Mk_BT}\,\delta\left[\hbar\omega - \frac{\hbar^2}{2M}\left(Q^2 + 2\bar{Q}\cdot\bar{\xi}_i\right)\right]}{\int d\bar{\xi}_i\ e^{-\hbar^2\bar{\xi}^2/2Mk_BT}}$$

$$= \frac{\int d\xi_x\ e^{-\hbar^2\xi_x^{\,2}/2Mk_BT}\,\delta\left[\hbar\omega - \frac{\hbar^2}{2M}\left(Q^2 + 2Q_x\cdot\xi_{ix}\right)\right]}{\int d\xi_x\ e^{-\hbar^2\xi_x^{\,2}/2Mk_BT}}$$

where the x-direction was chosen to coincide with $\bar{Q}$. Using the identities

$\delta(ax) = \frac{1}{|a|}\delta(x)$ and $\int e^{-ax^2} = \sqrt{\frac{\pi}{a}}$ one obtains

$$S(\bar{Q},\omega) = \frac{1}{\sqrt{4\pi k_B T E_r}} \exp\left[-\frac{(\hbar\omega - E_r)^2}{4k_B T E_r}\right]$$ where $E_r = \frac{\hbar^2 Q^2}{2M}$ is the average ***recoil energy*** transferred to the nuclei in the scattering event.



At low temperatures, the nuclei move slowly, and most of them are sped up upon colliding with a neutron, leading to a ***loss*** of neutron energy in the scattering events. At high temperatures, a significant number of nuclei move towards the neutron before the scattering event, so that they are slowed down and, conversely, the neutron ***gains*** energy upon scattering. The dynamical

structure factors for neutron energy gain and loss are related through the so-called “principle of detailed balance”:

$$S\left(-\vec{Q},-\omega\right)=e^{-\hbar\omega/k_BT}S\left(+\vec{Q},+\omega\right)$$

which can easily be verified for the case of the ideal gas, but holds generally for inelastic neutron scattering. A consequence of this relationship is that the cross section for neutron energy gain vanishes at $T=0$, because in this limit there are no thermally generated excitations present in the sample.

The scattering function (for an ideal gas) discussed above also applies to “deep inelastic scattering” of neutrons from nuclei in condensed matter, involving energy transfers well above the atomic binding energies. An example is a recent time-of-flight spectroscopy study in hydrogen atoms in a polymer, with energy transfers up to about $\hbar\omega=100eV$.

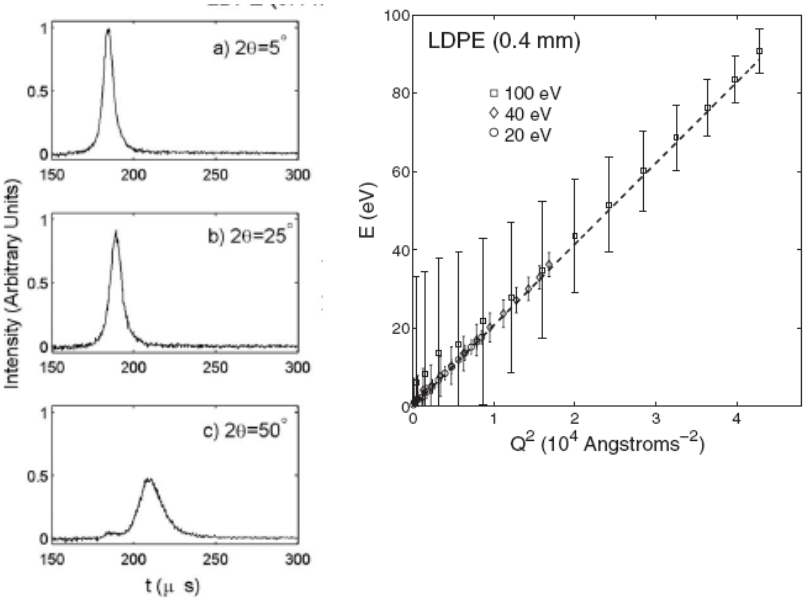


Fig. 4

Deep inelastic neutron scattering from hydrogen in a polymer

C. Stock et al.,
Phys. Rev. B 81, 024303 (2010)

For $\hbar\omega$ in the meV regime, the recoil of the atoms can be neglected. In the limit $\hbar\omega\gg E_r$, the Fourier transform of the scattering function for an ideal gas, which can be measured by spin-echo spectroscopy, can be written as

$$S\left(Q,t\right)=\int d\omega S\left(\vec{Q},\omega\right)e^{i\omega t}=e^{-\frac{Q^2t^2}{2\mu_\beta}}$$

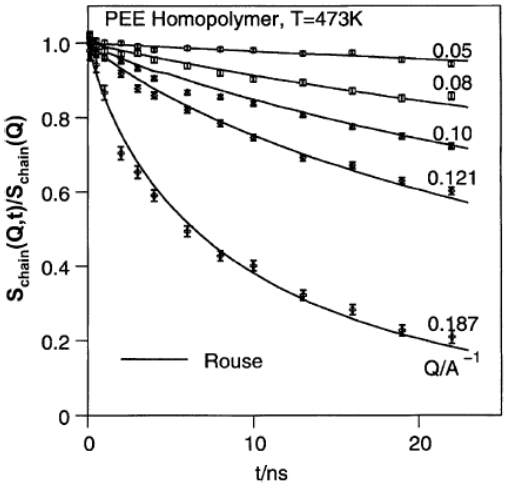
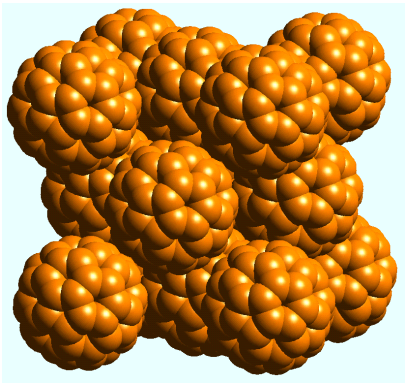


Fig. 5

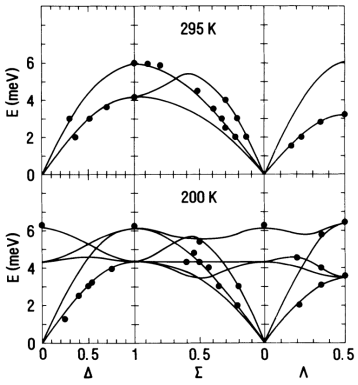
Neutron spin-echo spectroscopy of a polymer melt

D. Richter, Journal of the Physical Society of Japan 75, 111004 (2006)

Fig. 5 shows neutron spin-echo data on a polymer melt at high temperatures, which indicate a relaxation rate that decreases strongly with increasing Q, in qualitative agreement with this expression. The formula does not provide a quantitative description, however, because the polymer molecules diffuse through the melt, rather than moving ballistically as in an ideal gas. Neutron spin-echo spectroscopy has provided a lot of insight into the collective dynamics of polymers.



a)



b)

Fig. 6

Lattice structure and phonon dispersions of C_{60}

a) http://www.godunov.com/bucky/c60-gold.gif

b) L. Pintschovius et al., PRL 69, 2662 (1992)

(to be continued)