Neutron Spin Echo Spectroscopy

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uncluding slides and animations from
R. Gähler, R. Cywinski, W. Bouwman
Neutron flux

$$\varphi = \Phi \eta \frac{dE \, d\Omega}{4\pi}$$

definition of the beam : \(Q, E \) and polarisation
Neutron Spin Echo

why?

very high resolution

how?

using the transverse components of beam polarization

Larmor precession
Neutron Spin Echo

I: polarized neutrons - Larmor precession
II: NSE : Larmor precession
III: NSE : semi-classical description
IV: movies
V: quantum mechanical approach
VI: examples
VII: NSE and structure
I: polarized neutrons - Larmor precession
II: NSE: classical description
III: NSE: quantum mechanical description
IV: movies
V: NSE and coherence
VI: exemples
VII: NSE and structure
Polarized Neutrons

- polarizer
- analyzer

magnetic field (guide - precession)

Neutron source

magnetic field

polarizer \( \vec{B} \parallel \vec{P} \) analyzer

flipper

sample

Neutron detector
Longitudinal polarization analysis

Why longitudinal ????

because we apply a magnetic field and measure the projection of the polarization vector along this field

after F. Tasset
Larmor Precession

Motion of the polarization of a neutron beam in a magnetic field

\[ \frac{d\vec{\mu}}{dt} = \gamma \cdot (\vec{\mu} \times \vec{B}) = \vec{\mu} \times \vec{\omega}_L \]

"gyromagnetic ratio" of neutrons

\[ \gamma = 2.9 \text{ kHz}/G \]
Larmor Precession

\[ \frac{d\vec{S}}{dt} = \gamma \cdot (\vec{S} \times \vec{B}) \]

- \( d\vec{S} \perp \vec{B} \) \( \Rightarrow \) precession around \( \vec{B} \)
- \( d\vec{S} \perp \vec{S} \) \( \Rightarrow \) precession frequency is constant;

in both cases \( d\vec{S} \propto S \sin \theta \)
during \( dt \), the angular change of \( S \sin \theta \) around \( \vec{B} \) is constant:

\[ \Rightarrow \] the precession ‘Larmor’ frequency \( \omega_L \) does not depend on \( \theta \)
Why Precession

relation spin - magnetic moment

nucleons

\[ \mu_N = \frac{e\hbar}{2m_p} \]

electrons

\[ \mu_B = \frac{e\hbar}{2m_e} \]

e is the elementary charge, \( \hbar \) is the reduced Planck's constant,

\( m_p \) is the proton rest mass

\( m_e \) is the electron rest mass

The values of nuclear magneton

\begin{align*}
\text{SI} & \quad 5.050 \times 10^{-27} \text{ J} \cdot \text{T}^{-1} \\
\text{CGS} & \quad 5.050 \times 10^{-24} \text{ erg} \cdot \text{G}^{-1}
\end{align*}

The values of Bohr magneton

\begin{align*}
\text{SI} & \quad 9.274 \times 10^{-24} \text{ J} \cdot \text{T}^{-1} \\
\text{CGS} & \quad 9.274 \times 10^{-21} \text{ erg} \cdot \text{G}^{-1}
\end{align*}

ratio \quad \sim 1800
Larmor Precession

NMR spin echo
Erwin Hahn 1950

\[ \frac{d\vec{\mu}}{dt} = -\gamma \vec{\mu} \times \vec{H} = \vec{\mu} \times \vec{\omega}_L \]
NMR spin echo
Erwin Hahn 1950

\[ \frac{d\vec{\mu}}{dt} = -\gamma \vec{\mu} \times \vec{H} = \vec{\mu} \times \vec{\omega}_L \]
Larmor Precession

NMR spin echo
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Neutron spin echo
Ferenc Mezei 1972
Larmor Precession

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Neutron Spin Echo

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Larmor precession and flippers

Precession angle
\[ \varphi = \omega_L t = \gamma B t = \gamma B d / v \]

\[ \varphi = 2916.4 \text{ Hz/G} \times 2\pi \times B \text{ [G]} \times d \text{ [cm]} \times \lambda \text{ [Å]} / 395600 \]

For \( \varphi = 2\pi \)

\[ B d = 135.7 \text{ G.cm/Å} = 1.357 \times 10^{-3} \text{ T.m/Å} \]
Beam Polarization

\[ |\vec{P}| = \frac{I_+ - I_-}{I_+ + I_-} \]

\[ \Delta \phi \propto \Delta \lambda \]
neutron spin echo

Polarised neutron beam

$\pi/2$ flipper

$B_1$

$\pi$ flipper

$B_2$

$\pi/2$ flipper

$P \perp H$

Sample

$P \parallel H$

Analyser

$P_{NSE}^o$

$P_{NSE}^{scat.}$

$1.357 \times 10^{-3} \, T.m/\lambda [nm]$

$P_{\parallel B} = \langle \cos(\gamma B \ell/v) \rangle = \int f(v) \cos(\gamma B \ell/v) dv$
neutron spin echo

\[ \vec{P} \parallel \vec{B} \quad \vec{P} \perp \vec{B} \]

\[ \left\{ \begin{array}{c}
\pi \text{ flipper} \\
\pi/2 \text{ flipper}
\end{array} \right. \]

\[ B_1 \quad \ell_1 \quad B_2 \quad \ell_2 \]

polarized neutron beam

echo condition does not depend on the wavelength

\[ B_1 \ell_1 = B_2 \ell_2 \]
The basic equations for this scenario are:

\[ \phi_1 = \omega_L \cdot \frac{l}{v_1} \]
\[ \phi_2 = \omega_L \cdot l \left( \frac{1}{v_1} - \frac{1}{v_2} \right) \]

For \( v_1 = v \) and \( v_2 = v + \Delta v \) it follows:

\[ \phi_2 = \omega_L \cdot l \left[ \frac{1}{v} - \frac{1}{v + \Delta v} \right] \approx \omega_L \cdot l \cdot \frac{\Delta v}{v^2} = \omega_L \cdot t \cdot \frac{\Delta v}{v} \]
basic equations

\[ h \omega = m/2 \cdot (v_1^2 - v_2^2) = m/2 \cdot (v_1 + v_2)(v_1 - v_2) \approx m \cdot v \cdot \Delta v \]

[Diagram showing scattering theory: \( \Delta v \rightarrow \omega \)]

\[ \phi_2 \rightarrow \phi_2 \omega \rightarrow \ell \left[ \frac{1}{1/v \cdot t_{NSE}} \right] \approx \omega_L \ell \Delta v/v^2 = \omega_L t \Delta v/v \]

and \( t_{NSE} = \omega_L \cdot \ell \cdot h/(mv^3) = \omega_L \cdot t/(2\omega_o) \)
Polarised neutron beam

$\pi/2$ flipper

Sample

$\pi$ flipper

$\pi/2$ flipper

analyser

$P_{NSE}^0$

$P_{NSE}^\text{scat.}$

$P_{NSE}$

$P_{\text{scat.}}$

$P_{s}$

$\omega_o = 0$

\[ P_{NSE} = P_s \langle \cos(\phi - \langle \phi \rangle) \rangle = P_s \frac{\int S(Q, \omega) \cos[t(\omega - \omega_o)] \, d\omega}{\int S(Q, \omega) \, d\omega} \]

for quasi-elastic scattering $\omega_o = 0$

\[ \frac{P_{\text{scat.}}}{P_s} = \Re \left[ \frac{S(Q, t)}{S(Q)} \right] = I(Q, t) \]

most generally $\phi - \langle \phi \rangle = f(\vec{q}, \omega) \propto S((\vec{Q}), t)$

locally
paramagnetic scattering:

\[ P' = -\hat{Q} (\hat{Q} \cdot \vec{P}) \]

the $\pi$ flipper is the sample
measuring principle

$S(Q, \omega)$: probability for a momentum change $Q$ and an energy change $\omega$ upon scattering

**Typical $S(Q, \omega)$**

**Polar diagrams**

$t_{NSE} = t_1$

$t_{NSE} = t_2$

Distribution of spins at analyzer

$\frac{dI}{d\phi}$

Small $t_{NSE}$

Large $t_{NSE}$
Let $S(Q, \omega)$ be the scattering function and let $R(Q, \omega)$ be the resolution function.

If a spectrometer is set to $\omega'$, then the norm. countrate is:

$$I(Q,\omega') = \int S(Q,\omega) R(Q,\omega-\omega') \, d\omega = S \otimes R; \text{ convolution;}$$

The function $R(Q,\omega-\omega')$ should be the same over the range, where $S(Q,\omega)$ is significant;

If, like in spin echo, the Fourier Transform of $I$ is the signal, then the convolution of $S$ and $R$ can be written as

$$\text{FT}(I) = \text{FT}\{S \otimes R\} = \text{FT}\{S\} \cdot \text{FT}\{R\}$$

For NSE: $\text{FT}(I) = \text{FT}\left\{ \frac{\gamma^2}{\gamma^2 + \omega^2} \otimes \frac{\gamma_o^2}{\gamma_o^2 + \omega^2} \right\} = e^{-\gamma t} e^{-\gamma_o t} = e^{-(\gamma + \gamma_o) t}$
Subtleties of NSE

- Fields are mostly longitudinal;
- Adiabatic transitions at ends;
- ‘π/2 coils’ to start or end precession;
- ‘π coils’ to reverse effective field direction; (Hahn’s echoes; NMR-imaging);
- ‘Fresnel coils’ to compensate field inhomogeneities
- Adiabaticity parameter; at sample; at coils;
- How to measure polarization?
- Spin flip due to spin-incoherent (2/3) or paramagnetic scattering (depends on O(P, Q))
- Spin echo for ferromagnetic samples;

**Fresnel coils:**

**Standard cylindrical coil:**
\[ \int B \, dl \approx B_0 \, L + B_0 \, r^2/2D; \]
Correction by current loops:
\[ \int B_F \, df \sim I; \]
Current around loop

Density of loops increases with \( r^2 \);
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Plane wave entering a static B-field

\( \Psi_0 = e^{i(k_0y - \omega_0 t)} \)

\( y = 0 \)

The 2 states \( \Psi_+ \) and \( \Psi_- \) have different kinetic energies \( E_0 \pm \mu \cdot B \)

Static case \([dB/dt = 0]\): no change in total energy \((\omega = \omega_0)\) but change in \( k \)

\[
\frac{\hbar^2 k_\pm^2}{2m} = \frac{\hbar^2 k_0^2}{2m} \pm \mu B; \quad \Rightarrow \quad \frac{\hbar^2}{2m} (k_\pm^2 - k_0^2) = \pm \mu B;
\]

\[
\mu \cdot B, \quad E_{\text{kin}} \quad \Rightarrow \quad (k_\pm^2 - k_0^2) \approx (k_\pm - k_0) 2k_0 = \Delta k_\pm \cdot 2k_0; \quad \Delta k_\pm = \frac{2m \mu B}{\hbar^2 2k_0} = \frac{\mu B}{\hbar \times v}
\]

\( v \) is the classical neutron velocity
Both states have equal amplitudes, as the initial polarization is perpendicular to the axis of quantization (z-axis);
These amplitudes are set to 1 here.

Energy diagram:

\[
H = \begin{bmatrix} 1 \\ 1 \end{bmatrix} H_0
\]

\[
H_+ = H_0 \cdot e^{iy\Delta k}
\]

\[
H_- = H_0 \cdot e^{-iy\Delta k}
\]

\[
\Delta k = \mu \cdot B/\hbar \nu
\]
in a magnetic field

$$E_{\text{kin}} \quad [-E_{\text{pot}}]$$

$$E_0$$

$$\Psi = \begin{bmatrix} 1 \\ 1 \end{bmatrix} \Psi_0$$

$$\Psi = \begin{bmatrix} \Psi^+ \\ \Psi^- \end{bmatrix} = \begin{bmatrix} e^{i\Delta k \cdot y_0} \\ e^{-i\Delta k \cdot y_0} \end{bmatrix} \Psi_0$$

$$\Delta k = \mu \cdot B / \hbar v$$

$$t_0 = y_0 / v$$

Setting the polarizer to x-direction:

$$I_x = \frac{1}{\sqrt{2}} (\Psi^+ + \Psi^-) \times \mathbf{CC} = \frac{1}{2} (e^{i\Delta k \cdot y_0} + e^{-i\Delta k \cdot y_0})(e^{-i\Delta k \cdot y_0} + e^{i\Delta k \cdot y_0})$$

$$I_x = \frac{1}{2} \left(1 + e^{2i\Delta k \cdot y_0} + e^{-2i\Delta k \cdot y_0}\right) = 1 + \cos(2\Delta k \cdot y_0) = 1 + \cos \left(\frac{2\mu B}{\hbar v} \cdot y_0\right)$$

$$I_x = 1 + \cos (\omega_L t_0); \quad [I_y = 1 + \sin (\omega_L t_0)]; \quad \omega_L = \frac{2\mu B}{\hbar}; \quad \text{Larmor precession!}$$
wavepacket (bandwidth $\Delta \lambda$) of length $\Delta y$ and lateral width $\Delta x = \Delta z$;
$\Delta y \approx \lambda^2 / \Delta \lambda$; $\Delta x \approx \lambda / (2\pi \theta)$; $\theta =$ beam divergence; typ. values: $\Delta x, \Delta y \approx 100 \text{ Å}$

$\Delta k = \mu \cdot B / \hbar v$;

$\delta = v \cdot t$

Time splitting $dt = t$ of the two wave packets, separated by the propagation through the field of length $y_0$:

$$\frac{dE}{E} = \frac{2t}{t_o} \Rightarrow t = \frac{y_o}{2V} \cdot \frac{2\mu B}{\frac{1}{2}mV^2} = \frac{2\mu B \cdot y_o}{mV^3} = \frac{\omega_L \cdot t}{2\omega_o}$$

For fields of typ. 1 kG and length of m, $t$ is in the ns range for cold neutrons; In Neutron Spin echo spectroscopy, $t$ is the ‘spin echo time’;
The first field splits the wavepacket into two, the second one overlaps them again; The analyser superposes both packets; Complete overlap of scattered wave packets.
Neutron Spin Echo

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energy $E = \hbar \nu$ [meV]

scattering vector $Q$ [Å$^{-1}$]

length $d = 2\pi/Q$ [Å]

Inelastic Neutron Scattering

Spin Echo

X-ray correlation spectroscopy

Brillouin scattering

Raman scattering

Photon correlation

Backscattering

Chopper

Multi-Chopper

Inelastic x-ray

UT3

VUV-FEL

source: ESS

time $t$ [ps]
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Complete separation of the two packets implies that no coherent superposition of both states exists any more.
what is the real life - what is fantasy?

what is a spin?

what is the coherent superposition of states?

quantum mechanics = plane waves
is a coherent superposition of the two states their "sum"?

$|+\rangle + |-\rangle$ vs. the spin in the plane

the spin is not a vector (classical view)

what is classical what is quantum mechanical
coherent superposition of states

the spin is not a vector (classical)

\[ s = \pm \frac{1}{2} \]

\[ s_z = \begin{cases} +\frac{1}{2} \\ -\frac{1}{2} \end{cases} \]
coherent superposition of states

spin 1 case gives a hint: two \( s_z=0 \) states

singlet \( s_z=0 \) of the \( S=1 \) state
coherent superposition of states

Basics of QM:
2 slit experiment
wave - particle duality

The classical - “Copenhangen” - description of QM

The Problem of Measurement
E. Wigner 1963
coherent superposition of states

Basics of QM:
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- The classical - “Copenhagen” - description of QM

The Problem of Measurement
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Neutron spin echo spectrometers-

above: Mezei’s first spin echo precession coils

below: IN11

right: $\pi/2$ flipper coils

HZ Berlin
Examples of neutron spin echo studies

Reptation in polyethylene

The dynamics of dense polymeric systems are dominated by entanglement effects which reduce the degrees of freedom of each chain.

de Gennes formulated the reptation hypothesis in which a chain is confined within a “tube” constraining lateral diffusion – although several other models have also been proposed.

The measurements on IN15 are in agreement with the reptation model. Fits to the model can be made with one free parameter, the tube diameter, which is estimated to be 45Å.

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Larmor precession codes scattering angle

Unscattered beam gives spin echo at $\varphi = 0$
Independent of height and angle

Scattered wave vector transfer $Q_z$
results in precession $\phi$
$\phi \equiv zQ_z$
Proportional to the spin echo length $z$

Measure polarisation: $P(z) = \cos(zQ_z)$
Fourier transform scattering cross-section:
real space density correlation function $G(z)$

Direct information in measurement

Ordering
Compactness
Correlation lengths
Fractal dimensions
Specific surfaces
Etc…
SESANS measures directly shape ridges
Realisation SESANS

- monochromator
- polariser
- magnet 1
- field stepper
- guide field
- analyser
- detector
- polariser
- sample
Spin-echo reflectometer OffSPEC

- Off-specular to measure in-plane scattering
- Specular reflectivity of bent surfaces high-resolution
- Separation specular and off-specular

ISIS 2\textsuperscript{nd} target station
Off specular reflectometer Spin-echo components for High resolution without collimation
Thank you!