Neutron Spin Echo Spectroscopy

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including slides and animations from
R. Gähler, R. Cywinski, W. Bouwman
from the source to the detector

Neutron flux

\[ \varphi = \Phi \eta \frac{dE}{4\pi} \]

source flux distribution

intensity losses

field of neutron instrumentation

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

why ?

very high resolution without intensity losses

how ?

use the transverse components of beam polarization

Larmor precession
Neutron Spin Echo

I: polarized neutrons - Larmor precession
II: NSE : classical
III: NSE : semi-classical
IV: movies
V: quantum mechanical description ??
VI: examples
VII: NSE and structure
Neutron Spin Echo

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)
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

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

\[ d\vec{S} \perp \vec{B} \quad \Rightarrow \text{precession around } \vec{B} \]
\[ d\vec{S} \perp \vec{S} \quad \Rightarrow \text{precession frequency is constant;} \]

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

\[ \Rightarrow \quad \text{the precession ‘Larmor’ frequency } \omega_L \text{ does not depend on } \theta \]

“gyromagnetic ratio” of neutrons \( \gamma = 2.9 \text{ kHz/G} \)
**???? Precession ????**

**relation spin - magnetic moment**

**nucleons**

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

*e* is the elementary charge,
\( \hbar \) is the reduced Planck's constant,
\( m_p \) is the proton rest mass

**electrons**

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

*e* is the elementary charge,
\( \hbar \) is the reduced Planck's constant,
\( m_e \) is the electron rest mass

**The values of nuclear magneton**

- **SI** 5.050 × 10⁻²⁷ J·T⁻¹
- **CGS** 5.050 × 10⁻²⁴ Erg·Oe⁻¹

**The values of Bohr magneton**

- **SI** 9.274 × 10⁻²⁴ J·T⁻¹
- **CGS** 9.274 × 10⁻²¹ Erg·Oe⁻¹

**ratio** ~ 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

Neutron spin echo
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Larmor precession and flippers

Polarized neutron beam

Precession angle

\[ \phi = \omega_L t = -\gamma B t = -\gamma B \frac{d}{v} \]

\[ \phi = 2916.4 \text{ Hz/G} \times 2\pi \times B \text{ [G]} \times d \text{ [cm]} \times \lambda \text{ [Å]} \times \frac{1}{395600} \]

For \( \phi = 2\pi \)

\[ B d = 135.7 \text{ G.cm/ Å} = 1.357 \times 10^{-3} \text{ T.m/ nm} \]
$\Delta \phi \propto \Delta \lambda$

Beam Polarization

$$|\vec{P}| = \frac{I_+ - I_-}{I_+ + I_-}$$
Polarised neutron beam

$P \perp H$

$\pi/2$ flipper

Sample

$B_1$

$P_{\text{NSE}}^o$

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

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

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

\[ B_1 \ell_1 = B_2 \ell_2 \]

echo condition does not depend on the wavelength
basic equations

\[ \phi_1 = \omega_L \cdot \ell / v_1 \]

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

for \( v_1 = v \) and \( v_2 = v + \Delta v \) \( \Rightarrow \)

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

\[ \phi_2 = \phi_1 = t_{NSE} \cdot \omega \]

scattering theory: \( \Delta v \rightarrow \omega \)

\[ \hbar \omega = \frac{m}{2} \cdot (v_1^2 - v_2^2) = \frac{m}{2} \cdot (v_1 + v_2)(v_1 - v_2) \approx \frac{m \cdot v \cdot \Delta v}{2} \]

\[ \phi_2 - \phi_1 = \frac{1}{\hbar} \cdot t_{NSE} \cdot \ell \cdot \sqrt{\frac{m}{\hbar} \cdot \ell} = \frac{(\Delta \nu/v)}{v} \approx \omega_L \cdot \Delta v / v^2 = \omega_L \cdot t \cdot \Delta v / v \]

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

\( \pi /2 \) flipper

Sample

\( \pi \) flipper

\( \pi /2 \) flipper

Analyser

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

for quasi-elastic scattering \( \omega_o = 0 \)

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

most generally

\[ \phi - \langle \phi \rangle = f(\vec{q}, \omega) \propto S(\vec{Q}, t) \text{ locally} \]
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

\[ \phi = t_{NSE} \cdot \omega \]

Polar diagrams:

- \( t_{NSE} = t_1 \)
- \( t_{NSE} = t_2 \)

Distribution of spins at analyzer:

- 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;
- ‘$\pi/2$ coils’ to start or end precession;
- ‘$\pi$ 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:

$$\int B\text{d}l \approx B_0 L + B_0 \frac{r^2}{2D};$$
Correction by current loops:

$$\int B_F \text{d}f \sim I;$$

Current around loop

Fresnel coil density of loops increases with $r^2$;
Is the echo amplitude a polarization ?

Polarised neutron beam

$P \perp B$

$B_1$

$N_2$

$P \parallel B$

$\pi/2$ flipper

$\pi$ flipper

$\ell_1$

$\ell_2$

$P_{NSE}^o$

$P_{NSE}^{scat.}$

What is the polarization of the beam ???
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Plane wave entering a static B-field

\[ \Psi_0 = e^{i(k_0 y - \omega_0 t)} \]

\[ \sigma_+ \quad \sigma_+ \]

\[ \sigma_- \quad \sigma_- \]

\[ H? \quad B = B_z \]

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 \(k\)

\[ \frac{\hbar^2 k^2_\pm}{2m} = \frac{\hbar^2 k^2_0}{2m} \pm \mu B; \quad \Rightarrow \quad \frac{\hbar^2}{2m} \left( k^2_\pm - k^2_0 \right) = \pm \mu B; \]

\[ \mu \cdot B, E_{\text{kin}} \quad \Rightarrow \quad \left( k^2_\pm - k^2_0 \right) \approx (k_\pm - k_0)2k_0 = \Delta k_\pm \cdot 2k_0; \quad \Delta k_\pm = \frac{2m \mu B}{\hbar^2 2k_0} = \frac{\mu B}{\hbar \times v} \]

\( v \) 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:
in a magnetic field

\[ E_{\text{kin}} \to [-E_{\text{pot}}] \]

\[ \Psi = \begin{bmatrix} 1 \\ 1 \end{bmatrix} \Psi_0 \]

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

\[ \Delta k = \mu \cdot B / \hbar v \]

\[ t_0 = y_0 / v \]

Setting the polarizer to x-direction:

\[ I_x = \frac{1}{\sqrt{2}} (\Psi^+ + \Psi^-) \times \mathbf{C} \mathbf{C} = \frac{1}{2} \left( e^{i\Delta k \cdot y_0} + e^{-i\Delta k \cdot y_0} \right) \left( e^{-i\Delta k \cdot y_0} + e^{i\Delta k \cdot y_0} \right) \]

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

\[ I_x = 1 + \cos (\omega_L t_0); \quad \left[ I_y = 1 + \sin (\omega_L t_0) \right]; \quad \omega_L = 2\mu B / \hbar; \quad \text{Larmor precession!} \]
wavepackets instead of plane waves

wavepacket (bandwidth $\Delta \lambda$) of length $\Delta y$ and lateral width $\Delta x = \Delta z$;
$\Delta y \approx \lambda^2 / \Delta \lambda$; $\Delta x \approx \lambda / (2\pi\theta)$; $\theta = \text{beam divergence}$; 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 $y_0$:

$$
\frac{dE}{E} = \frac{2t}{t_0} \Rightarrow t = \frac{y_o}{2V} \cdot \frac{2\mu B}{m \sqrt{2}} = \frac{2\mu B \cdot y_o}{m \sqrt{3}} = \frac{\omega_L \cdot t}{2\omega_o}
$$

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
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 $d = \frac{2\pi}{Q}$ [Å]

source: ESS
Neutron Spin Echo

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Complete separation of the two packets implies that no coherent superposition of both states exists any more.

\[ \Delta y \approx \frac{\lambda^2}{\Delta \lambda}; \]

Larmor precession

Directions of the individual spins of the polychromatic beam after passage through B fields of different lengths

What is the real life - what is fantasy?
what is the real life - what is fantasy?

what is a spin?

what is the coherent superposition of states?

quantum mechanics = plane waves
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
is the coherent superposition of states the “sum” of the two or not?

\[ |+\rangle + |-\rangle \]

spin the the plane classical
quantum mechanical

the spin is not a vector (classical)
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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 45 Å.

Neutron Spin Echo

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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

Polarisation, $P(z)$

slope $\sim \lambda^2(\Delta\rho_p)^2\phi(1-\phi)$

$e^{-\Sigma_t}$

$\Sigma_t = \int_0^\infty \frac{\xi}{(\Delta\rho_p)^2\phi(1-\phi)}$ d$\xi$

spin-echo length, $z$

$\Sigma_t$
SESANS measures directly shape ridges
Realisation SESANS

monochromator
polariser
magnet 1
field stepper
guide field
analyser
detector
polariser
polariser

sample

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

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

TS2 ISIS

TU Delft