Introduction to Neutron Spin Echo Spectroscopy

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1. The neutron scattering event

(in the Fraunhofer approximation)

\[ \vec{Q} = \vec{k}_i - \vec{k}_f \]

\[ \Delta E = E_i - E_f = \hbar \omega = \text{energy transfer} \]

\[ k = \frac{2\pi}{\lambda} \quad \text{neutron momentum} \]

\[ E = \frac{(\hbar k)^2}{2m_n} \quad \text{neutron energy} \]
2. **QENS**: quasielastic neutron scattering

A limiting case of inelastic scattering, centered at $\omega = 0$ characterized by small energy transfers

Energy spectra = structural and dynamical information

$S(Q, \omega) = \text{dynamic structure factor}$

$\Delta E = E_i - E_f$

$\Delta E \ll E_i$

- elastic peak
- quasielastic line
- inelastic peaks

$\omega < 0 \ (\text{gain}) \quad \omega = 0 \quad \omega > 0 \ (\text{loss})$
3. Neutron spectroscopy landscape

(methods to measure dynamics)

XTL, TOF, XTL-TOF, TOF-TOF, XTL-XTL, TOF-XTL, NSE

1meV ~ 8cm\(^{-1}\) ~ 1ps
1µeV ~ 0.008cm\(^{-1}\) ~ 1ns
1neV ~ 8x10\(^{-6}\) cm\(^{-1}\) ~ 1µs
4. Neutron Spin Echo = NSE - two principles

4.1. Larmor precession

- **Larmor Frequency**
  \[ \omega_L = |\gamma B| \]

- **Accumulated phase**
  \[ \phi = \omega_L t = \gamma_L B t = \frac{\gamma B l}{v} \]

- **Neutron Gyromagnetic Ratio**
  \[ |\gamma/2\pi| \approx 30 \text{ MHz/T} \]

Figure by Laura R. Stingaciu, ORNL http://xrayphysics.com/sequences.html

Neutron Spin Echo Spectroscopy by Piotr A. Zolnierczuk, NXS-2017: https://www.youtube.com/watch?v=tIFysL68PhM
Neutron Spin Echo = NSE - two principles

4.2. Hann echo

https://en.wikipedia.org/wiki/Neutron_spin_echo


Marcus Hennig: Dynamics of ... Studied by Neutron Scattering, 2011, Corpus ID: 91625454
Neutron Spin Echo – a quasielastic process
Neutron Spin Echo signal

\[ I \sim \langle \cos \phi \rangle = \langle \cos \omega \tau \rangle \]

\[ I \sim I(Q) \pm \int S(Q, \omega) \cos(\omega \tau) \, d\omega \]

- \( B \sim I(Q) \)
- \( A \sim I(Q, \tau) = \mathcal{F}[S(Q, \omega)] \)

\[ \frac{I(Q, \tau)}{I(Q, 0)} = \frac{2A}{U-D} \]

Fourier time

\[ \tau \approx 0.186 J \lambda^3 \, [\text{ns}] \]

\([J] = \text{Tm}, [\lambda] = \text{Å}\)

Neutron Spin Echo Spectroscopy by Piotr A. Zolnierczuk, NXS-2017: https://www.youtube.com/watch?v=tIFysL66PnM
Coherent and incoherent scattering in NSE

\[ I_{\text{up}} = I_{\text{coh}} + \frac{1}{3} I_{\text{inc}} \]

\[ I_{\text{down}} = \frac{2}{3} I_{\text{inc}} \]

\[ I_{\text{echo}} = I_{\text{coh}} f_{\text{coh}}(\tau) - \frac{1}{3} I_{\text{inc}} f_{\text{inc}}(\tau) \]
Energy and time domain \( \text{QENS} \leftrightarrow \text{NSE} \)

**QENS:** Dynamic Structure Factor

\[
S(Q, \omega) \quad \longrightarrow \quad \text{Fourier Transform} \quad \longrightarrow \quad I(Q, \tau)
\]

**NSE:** Intermediate Scattering Function

\[
I_D(Q, \omega) = S(Q, \omega) \ast R(Q, \omega) \quad \quad \quad I_D(Q, \tau) = I(Q, \tau) \ast R(Q, \tau)
\]

Neutron Spin Echo Spectroscopy by Piotr A. Zolnierczuk, NXS-2017: https://www.youtube.com/watch?v=tIFysL68PhM
SNS-NSE overview

- Ultrahigh resolution spectrometer for characterizing slow dynamics of soft condensed matter at nanoscopic and mesoscopic scale
- Detects neutron velocity changes < $10^{-5}$
- The only NSE spectrometer at a pulse source
- The first NSE spectroscopic design based on superconducting technology
- The only NSE spectrometer with complete magnetic shielding
SNS-NSE, the Neutron Spin Echo spectrometer @ SNS

- DENEX detector
- Analyzer
- Main precession = SC NbTi
- Sample = 3 x 3 cm²
- Flippers
- mu metal shielding

neutrons.ornl.gov/nse
SNS-NSE Sample Environments

ThermoJET Temperature Forcing System (TFS), 275K - 400K

Janis cryostat, 5K - 700K
SNS-NSE Data Reduction For Spin Echo Experiments

DrSPINE

DrSPINE is a unified reactor - pulse source NSE data reduction software

Research papers

Efficient data extraction from neutron time-of-flight spin-echo raw data

P. A. Zolnierczuk, O. Holderer, S. Pasini, T. Kozielewski, L. R. Stingaciu and M. Monkenbusch

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Neutron spin-echo spectrometers with a position-sensitive detector and operating with extended time-of-flight-tagged wavelength frames are able to collect a comprehensive set of data covering a large range of wavevector and Fourier time space with only a few instrumental settings in a quasi-continuous way. Extracting all the information contained in the raw data and mapping them to a suitable physical space in the most efficient way is a challenge. This article reports algorithms employed in dedicated software, DrSpace (data reduction for spin echo), that achieves this goal and yields reliable representations of the intermediate scattering function $S(Q,t)$ independent of the selected “binning”.

J-NSE Phoenix courtesy of Olaf Holderer, FRMII
SNS-NSE and TOF

- $\Delta \lambda = (5 - 8) \ \text{Å}$
- $2\theta = 3.6^\circ \ Q = 0.02 - 0.15 \ \text{Å}^{-1}$
- $2\theta = 37^\circ \ Q = 0.45 - 0.85 \ \text{Å}^{-1}$
SNS-NSE research highlights

dynamics of soft matter at nano- to meso-scale

Coherent dynamics in polymers, glassy systems, bio-macromolecules

Magnetic Dynamics, Spin glasses & spin fluids

Domain and allosteric motions in proteins

Interaction of solvent molecules with surfaces

Hydrogen jump diffusion

Diffusion processes

Shape fluctuations

Short range translational, rotational and tumbling diffusive motions
SDS micelles in aqueous solution


\[
D_0 = kT / 6\pi \eta R
\
D_{\text{eff}} = D_0 H(Q)/S(Q)
\]

\[
I(Q,t) = I(Q,0) e^{-D_{\text{eff}}(Q)Q^2 t}
\]
Motion in entangled polymer melts


\[ \frac{S(Q, \tau)}{S(Q, 0)} = [1 - F(Q)]S^{loc} + F(Q)S^{esc} \]

- NSE Spin-Incoherent scattering measures proton self-correlation function
- Labeled long linear PEP polymer chain
- Segmental dynamics
Dynamics of phospholipid membranes


- Friction at the interface water - liposomes plays a minor role
- The center of mass diffusion of liposomes and the transient trapping of lipids define the range in which ZG model can be applied

Proposed mechanism of transient trapping and mean square displacement $\langle \Delta r(t)^2 \rangle_N$ as a function of Fourier time
Proteins domains dynamics

L.R. Stingaciu et al., Sci Rep 6, 22148 (2016) https://doi.org/10.1038/srep22148

Three protein fragments in harmonic potential connected by flexible linkers
Spin dynamics in frustrated magnets


Magnetic charges of $\pm Q$ unit and $\pm 3Q$ accumulate on the vertices of the honeycomb lattice.

The charge defect relaxes between nearest neighbors.
Neutron Spin Echo Variations
Paramagnetic NSE

Sample is the $\pi$-flipper

Resonance, MIEZE, and SANS - spin echo

NRSE (Neutron Resonance Spin Echo)
T-NRSE or L-NRSE

RF field instead of solenoid
• compact design
• shorter Fourier times

http://larmor.weblog.tudelft.nl/files/2013/07/S_Rogers_SANS_SESANS1.pdf

SESANS
Spin Echo SANS

• tilted magnetic field
• angle encoded in spin precession

Wide angle spin echo

- Large angular coverage
- Higher Q (up to $3\text{Å}^{-1}$)

https://www.ill.eu/users/instruments/instruments-list/wasp:description/instrument-layout
Summary - NSE in a nutshell

- NSE measures very small velocity changes using neutron spin precession in magnetic field
- Broad $\Delta \lambda/\lambda$ and high resolution
- Intermediate Scattering Function: $I(Q,\tau)$
- Complementary to SANS/SAXS
- Counting intensive and large samples
Reference reading

- R. Pynn, Neutron Scattering, Neutron Spin Echo  
  http://www.iub.edu/~neutron/notes/20061204_Pynn.pdf


- M. Monkenbusch, D. Richter, High Resolution Neutron Spectroscopy  
  http://doi.org/10.1016/j.crhy.2007.10.001


- B. Farago, Basics of Neutron Spin-Echo, ILL Neutron Data Booklet, 
Questions about NSE?

https://neutrons.ornl.gov/instruments

Neutron Spin Echo Spectrometer | Neutron Science at ORNL
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