Introduction to Neutron Spin Echo Spectroscopy

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Methods and Applications of SANS, NR, and NSE
NCNR, June 18th – 23rd, 2012

Outline

- History
- Dynamic Neutron Scattering (Recap)
- The principles of Neutron Spin Echo
- Instrumental Setup of Neutron Spin Echo
- Conclusion

The beginning

Neutron Spin Echo: A New Concept in Polarized Thermal Neutron Techniques

F. Mezei

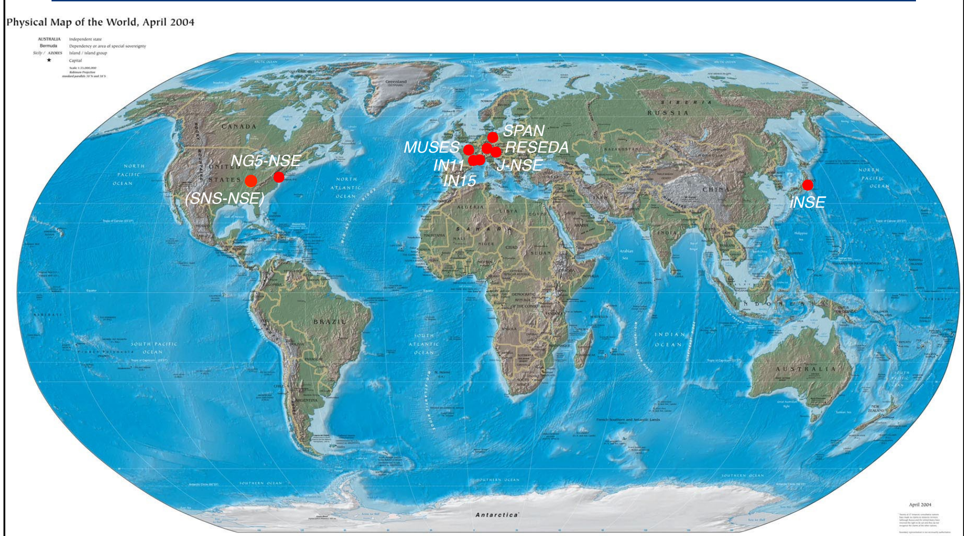
Institut Laue-Langevin, Grenoble, France* and
Central Research Institute for Physics, Budapest, Hungary

Received July 7, 1972

A simple method to change and keep track of neutron beam polarization non-parallel to the magnetic field is described. It makes possible the establishment of a new focusing effect we call neutron spin echo. The technique developed and tested experimentally can be applied in several novel ways, e.g. for neutron spin flipper of superior characteristics, for a very high resolution spectrometer for direct determination of the Fourier transform of the scattering function, for generalised polarization analysis and for the measurement of neutron particle properties with significantly improved precision.

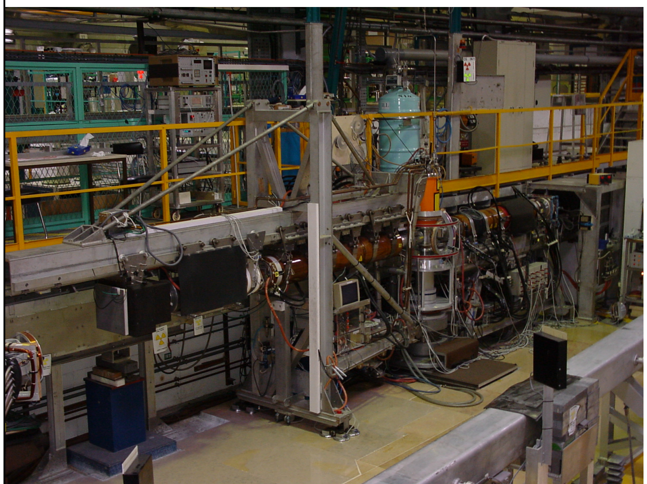
F. Mezei, *Z. Physik*. **255**, 146 (1972).

NSE Today in the World



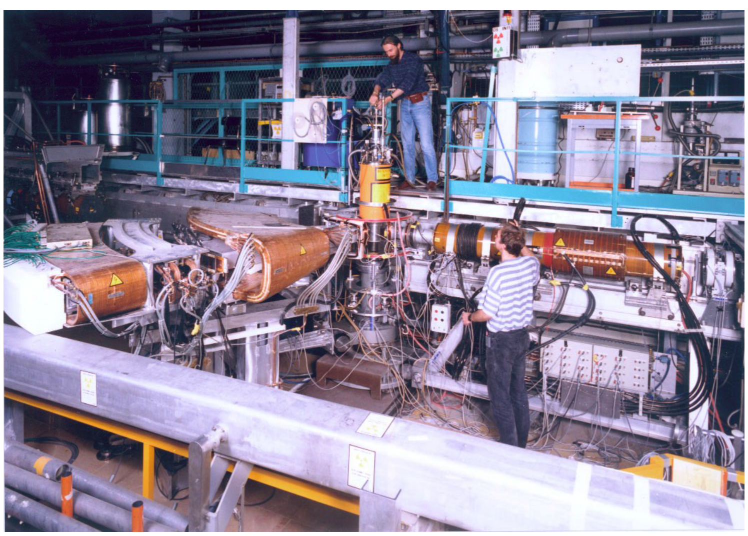
+ NSE option for triple-axis spectrometers
+ developing ones

What does a NSE Spectrometer Look Like?



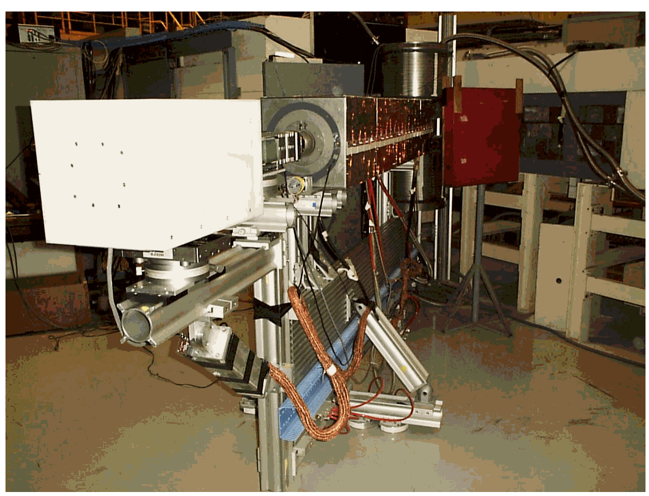
IN-11 @ ILL
The first

IN-11c @ ILL; Wide Detector Option

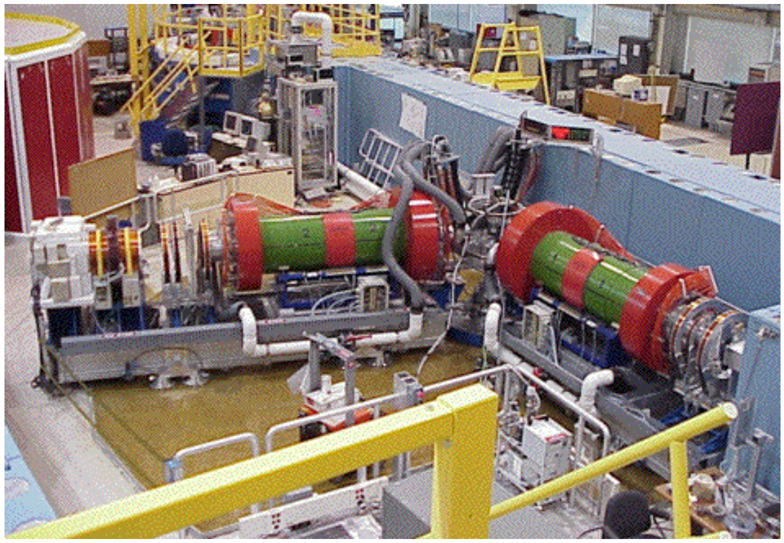


IN-15 @ ILL

MUSES, Resonance Spin Echo @ LLB

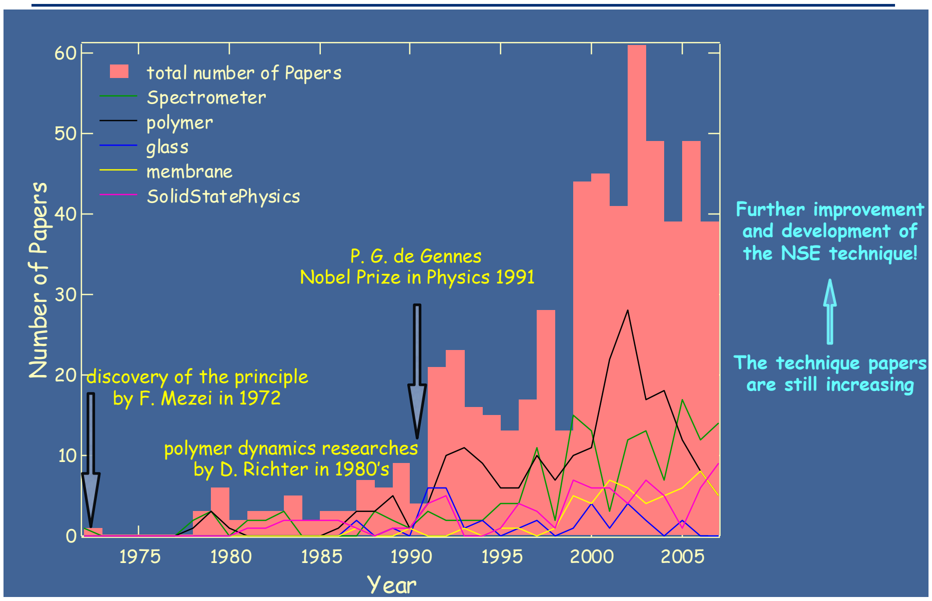


NSE @ NCNR



NSE @ SNS

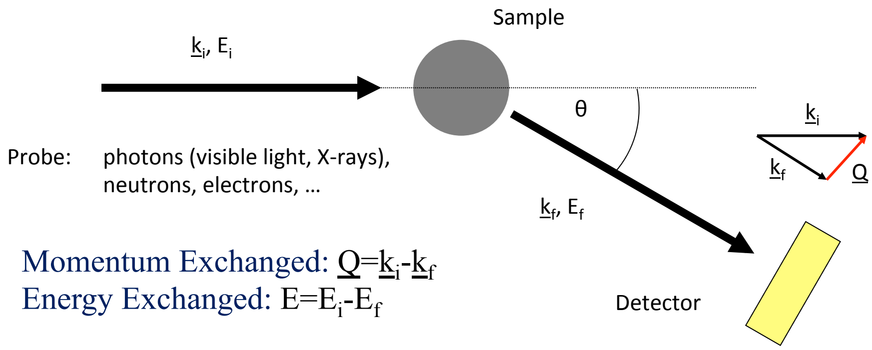
NSE Publications



publication record; searched using web of science: keyword=neutron spin echo

Dynamic Neutron Scattering

Static and Dynamic Scattering



Momentum Exchanged: $\underline{Q}=\underline{k}_i-\underline{k}_f$
Energy Exchanged: $E=E_i-E_f$

StaticScattering: $\frac{d\sigma}{d\Omega} \approx S(Q)$ Dynamic Scattering: $\frac{d^2\sigma}{d\Omega dE} \approx S(Q,E)$

Static and Dynamic Scattering

$$S(Q) = FT_S\left\{\left\langle \exp[-i\underline{Q}(\underline{r}_i - \underline{r}_j)] \right\rangle\right\}$$

Fourier Transform of the Space Correlation Function

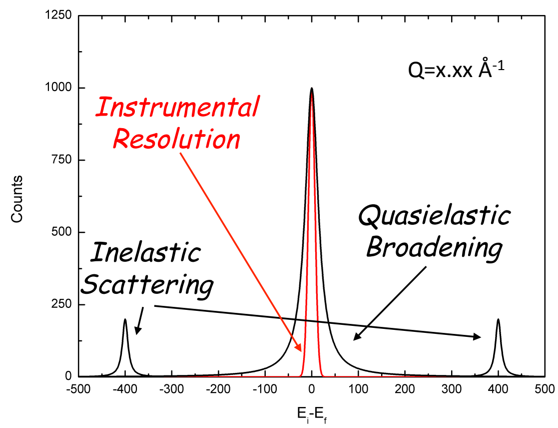
$$S(Q,E) = FT_T\left\{\left\langle \exp[-i\underline{Q}(\underline{r}(t) - \underline{r}(0))] \right\rangle\right\} = FT_T\{I(Q,t)\} = FT_{ST}\{G(r,t)\}$$

Fourier Transform of the Space-Time Correlation Function

I(Q,t) Intermediate Scattering Function (ISF)

G(r,t) van Hove Correlation Function

S(Q,E)



Thermally Activated Motions Takes Place in the System.

Inelastic Scattering

excitation: neutrons exchange energy with an oscillatory motion which has a finite energy transfer

Ex: phonon, magnon, ...

Quasielastic Scattering

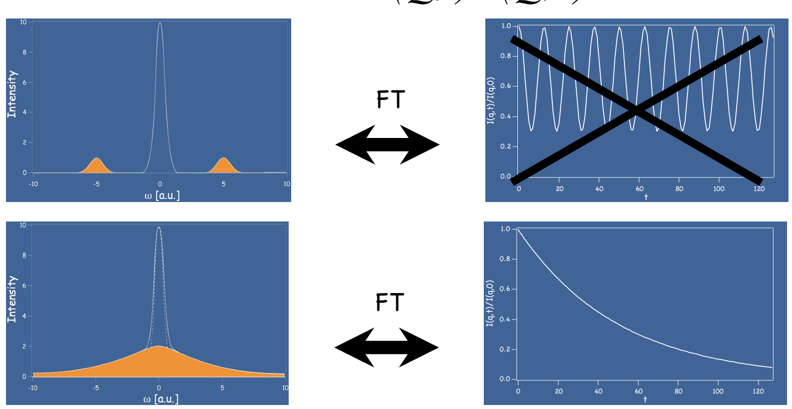
relaxation: neutrons exchange energy with random motion which makes another new equilibrium state (no typical finite energy transfer exists)

Ex.: rotation, vibration, diffusion…

Energy and Time Domain

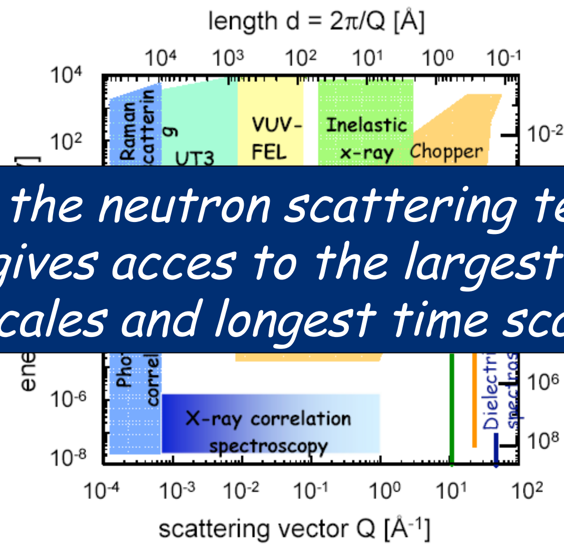
NSE works in the TIME domain

NSE measures the Normalized Intermediate Scattering Function: *I(Q,t)/I(Q,0)*



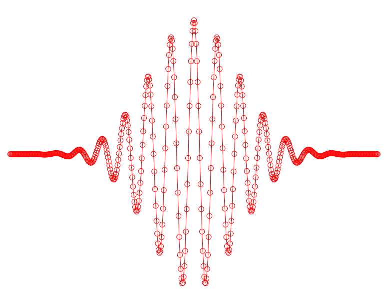
NSE Deals Almost Exclusively with Quasielastic Scattering

Dynamic Neutron Scattering



NSE is the neutron scattering techniques that gives acces to the largest length-scales and longest time scales

Neutron Spin Echo Principle of Operation



The Idea of Neutron Spin Echo

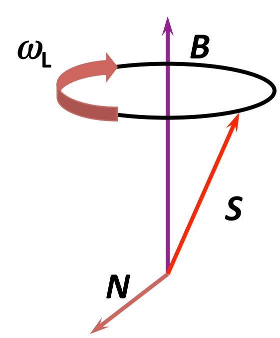
NSE Breaks the Relationship between Intensity & Resolution

- Traditional Instruments – define *both* incident & scattered wavevectors in order to define *E* and *Q* accurately
- Traditional Instruments – use collimators, monochromators, choppers etc to define both $\underline{k}_i$ and $\underline{k}_f$
- NSE – measure the *difference* between appropriate components of $\underline{k}_i$ and $\underline{k}_f$ (original use: measure $\underline{k}_i - \underline{k}_f$ i.e. energy change)
- NSE – use the neutron's spin polarization to encode the difference between components of $\underline{k}_i$ and $\underline{k}_f$
- NSE – can use large beam divergence &/or poor monochromatization to increase signal intensity, while maintaining very good resolution

Neutron Precession

Neutron Properties

- Mass, $m_n = 1.675\times10^{-27}$ kg
- Spin, $S = 1/2$ [in units of $h/(2\pi)$]
- Gyromagnetic ratio $\gamma = g_n\mu_n/[h/(2\pi)] = 1.832\times10^{8}\ s^{-1}T^{-1}$ (29.164 MHz T^{-1})

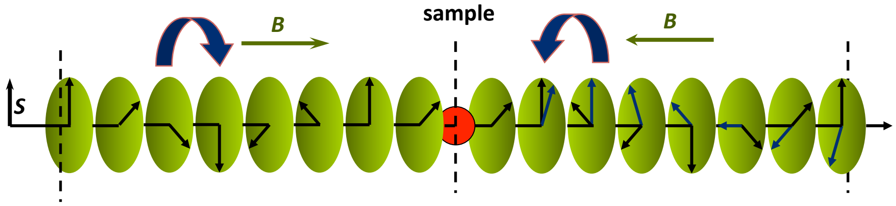


In a Magnetic Field

- The neutron experiences a torque from a magnetic field *B* perpendicular to its spin direction. ⟹ $N = S \times B$
- Precession with the Larmor frequency: ⟹ $\omega_L = \gamma B$

Scattering Event: Single Neutron

• elastic scattering • inelastic scattering



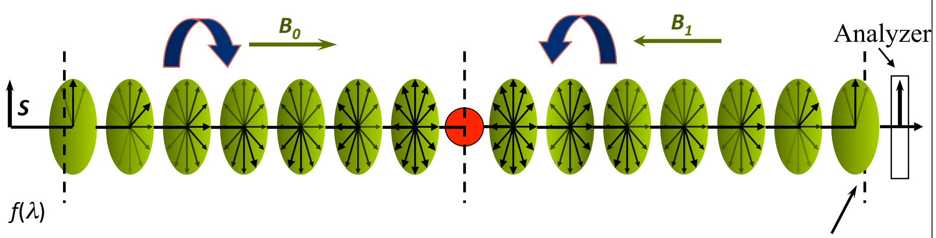
$$\varphi = \gamma \frac{\int B dl}{v} \qquad \Delta\varphi = \gamma\left(\frac{1}{v} - \frac{1}{v'}\right)\int B dl = \frac{\gamma \Delta v}{v^2}\int B dl = \frac{m\lambda}{h}\frac{\gamma \Delta v}{v}\int B dl$$

$$NT(\lambda) = \frac{1}{2\pi}\int \frac{\gamma B m \lambda}{h} dl = \frac{\gamma m \lambda}{2\pi h}\int B dl = 7370 \times J[T \cdot m] \times \lambda[\mathring{A}]$$

$$J = \int B dl$$

J field integral. At NCNR: J_{max} = 0.5 T.m
NT (λ=8Å) ≈ 3×10^5

Scattering Event: Neutron Beam



Again:
The measured quantity is the Polarization, i.e. the spin component along x: $P_x = \langle \cos\varphi(\lambda) \rangle$:

Elastic scattering

$$\overline{\varphi} = \left\langle \gamma \frac{\int B_0 dl}{v} - \gamma \frac{\int B_1 dl}{v} \right\rangle_{f(\lambda)}$$

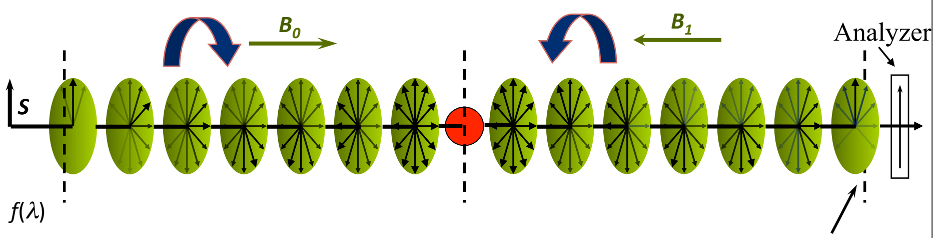
Echo Condition: $J_0 = J_1$

$P_x = 1$ $\qquad \overline{\varphi} = 0$

Note:

The requirement that φ=0 can be in some cases released. This treatment is valid for the most common case of Quasi-Elastic Scattering

Scattering Event: Neutron Beam



Quasi-Elastic scattering

Again:
The measured quantity is the Polarization, i.e. the spin component along x: $P_x=\langle\cos\varphi(\lambda)\rangle$:

$$\varphi = \left\langle \gamma \frac{\int B_0 dl}{v(\lambda)} - \gamma \frac{\int B_1 dl}{v(\lambda)+\delta v} \right\rangle$$

Series Expansion in δλ and δJ

$$\varphi \approx \gamma \frac{m}{h} J_0 \delta\lambda + \gamma \frac{m}{h}(J_0 - J_1)\lambda$$

phase

$$\hbar\omega = \Delta E = \frac{h^2}{2m}\left[\frac{1}{\lambda^2} - \frac{1}{(\lambda+\delta\lambda)^2}\right] \approx \frac{h^2}{m}\frac{\delta\lambda}{\lambda^3}$$

$$\delta\lambda = \frac{\omega}{2\pi h} m\lambda^3$$

$$\varphi = \gamma \frac{m^2\lambda^3}{2\pi h^2} J_0\omega + \gamma \frac{m}{h}(J_0 - J_1)\lambda$$

0 at the echo condition

The Basic Equations of NSE

$$P_x = \langle\cos(\varphi)\rangle = \iint f(\lambda) S(Q,\omega) \cos\left[\gamma \frac{m^2\lambda^3}{2\pi h^2} J_0\omega + \gamma \frac{m}{h}(J_0 - J_1)\lambda\right] d\lambda d\omega$$

Even Function

$$P_x = \langle\cos(\varphi)\rangle = \int f(\lambda)\cos\left[\gamma \frac{m}{h}(J_0 - J_1)\lambda\right] d\lambda \times \int S(Q,\omega)\cos\left[\gamma \frac{m^2\lambda^3}{2\pi h^2} J_0\omega\right] d\omega$$

FT of the wavelength distribution

FT of the Dynamic Structure Factor

Fourier time $t = \gamma \frac{m^2}{2\pi h^2} J_0 \lambda^3$

$$P_x(\Delta J^{ph_i}, Q, t) = P_s(\Delta J^{ph_i}) \int S(Q,\omega)\cos[\omega t] d\omega = P_s(\Delta J^{ph_i}) I(Q,t)$$

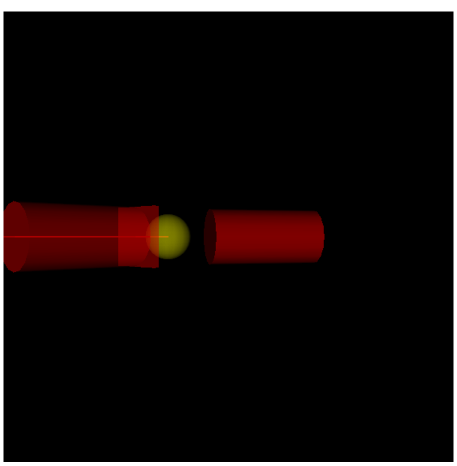
NSE measures the Fourier Transform of the Dynamic Structure Factor

NSE principles: summary

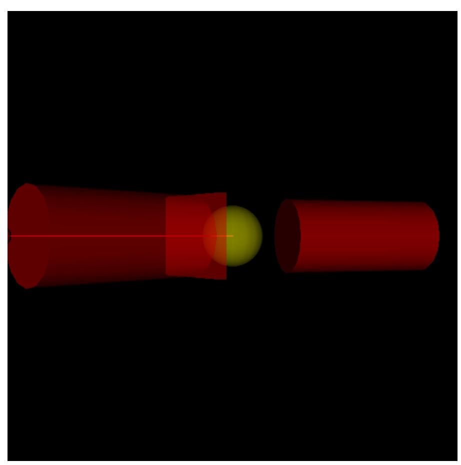
- Neutron Spin Echo Instruments are "machinery" which measure the Fourier transform of the Dynamic Structure Factor, $S(Q,\omega)$, i.e. the (normalized) Intermediate Scattering Function (ISF), $I(Q,t)/I(Q,0)$.
- The neutrons' velocity is encoded into their spin state.
  - The neutron beam goes through two equals homogeneous magnetic field paths before and after the sample.
  - Within the magnetic fields each neutrons' spin performs a precession at a speed determined by the field intensity.
  - If the scattering event does not change the neutrons' speed (elastic scattering), the initial polarization of the beam is recovered.
- The polarization of the scattered neutron beam is proportional to the ISF at a specific value of t determined by the strength of the precession field (and by the incident neutron wavelength).

NSE Effect

Elastic Scattering

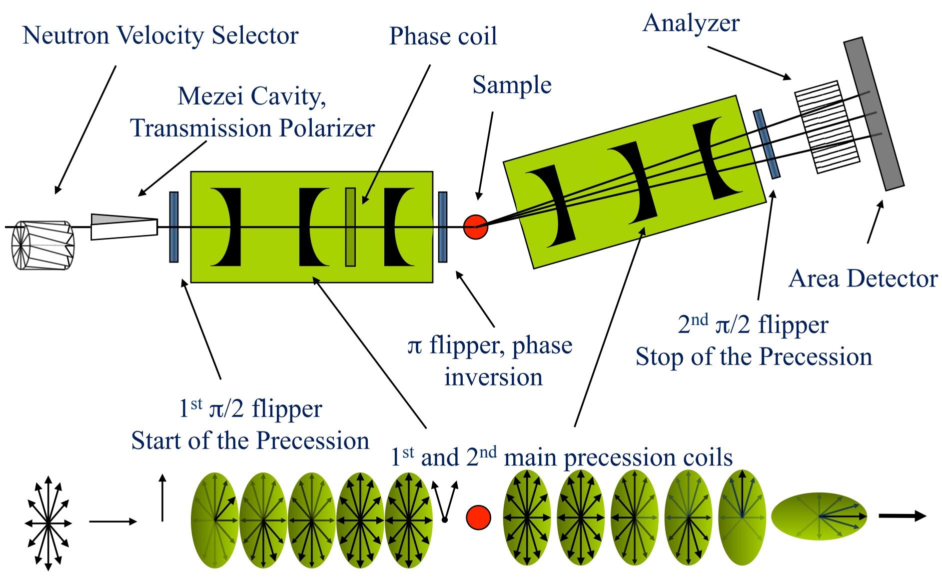


QuasiElastic Scattering

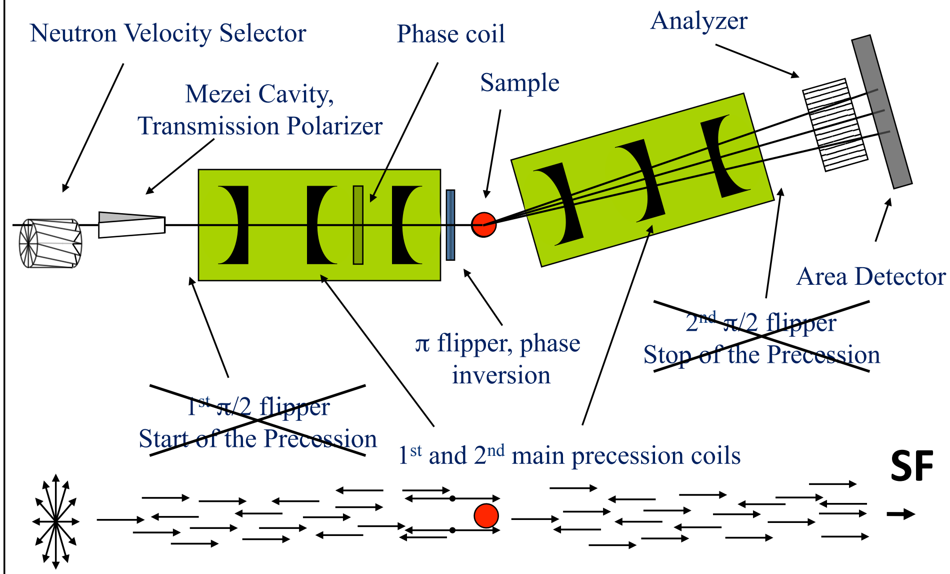


Instrument Setup

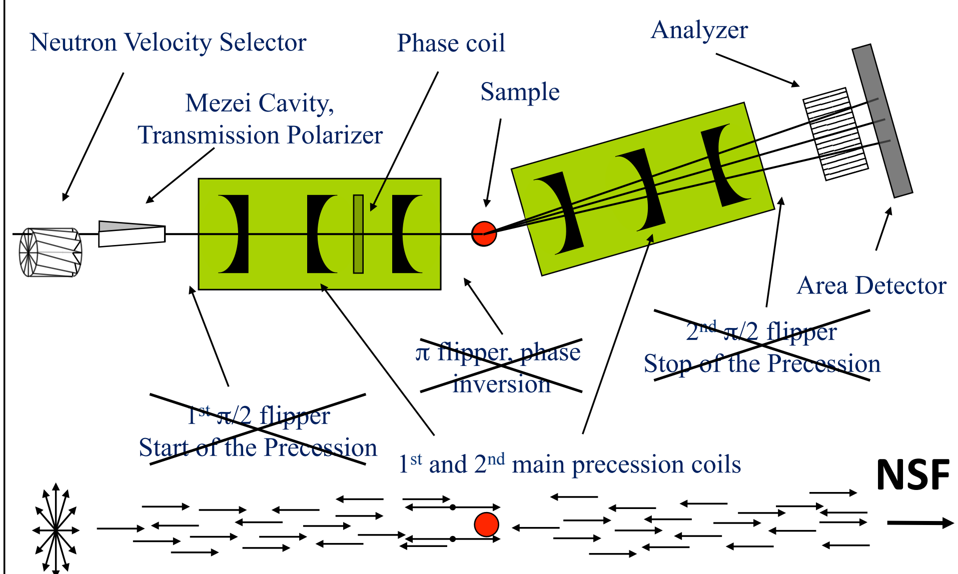
NSE: Instrumental Setup



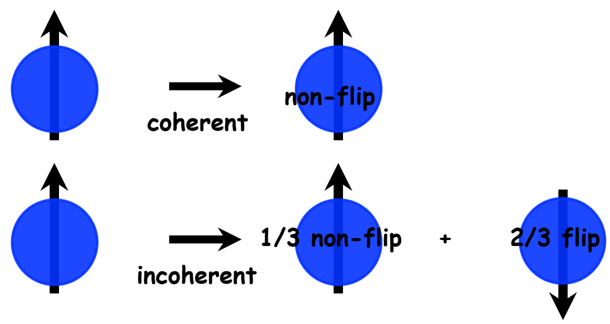
Polarization Analysis



Polarization Analysis



Nuclear Scattering Event and Neutron Moment



Nuclear Scattering Polarization Analysis

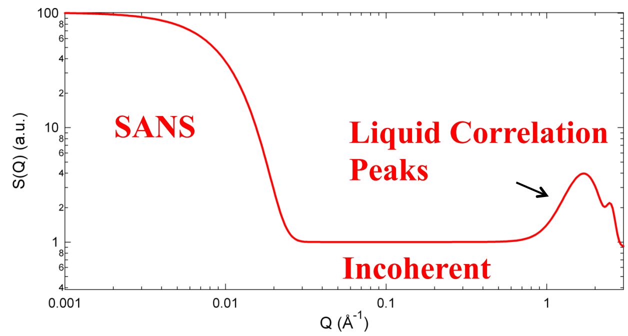
If no magnetic scattering is present and the sample is isotropic, the three polarization directions, xyz, are equivalent:

$$N^{incoh} = \frac{3}{2} SF$$

$$N^{coh} = NSF - \frac{1}{2} SF$$

Structure Factor

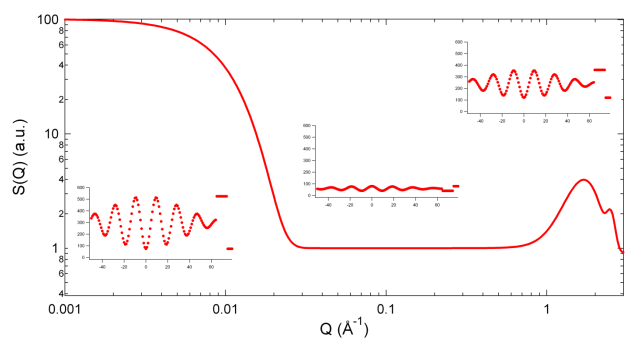
The Structure Factor, *S(Q)*, is the most relevant information for the preparation and, later, interpretation of a NSE experiment.



NSE: Coherent vs Incoherent

NSE is known for the investigation of the coherent dynamics.

Incoherent scattering intensity is reduced to -1/3 in NSE. The best achievable flipping ratio is 0.5.



However, the main limitation to the study of incoherent scattering by NSE is the *Q* coverage of the instrument.

Recent advancements in NSE instrumentation aim to overcome this limitation (IN-11C, the WASP project).

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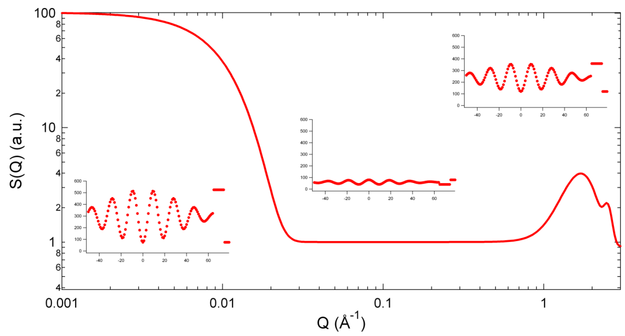


However, the main limitation to the study of incoherent scattering by NSE is the Q coverage of the instrument.

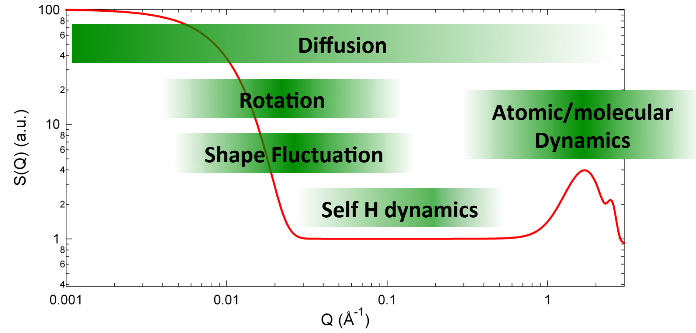
Most important is to avoid Q areas where coherent and incoherent intensity cancel each other.

The Intermediate Scattering Function

NSE measures the time decay of the structures defining $S(Q)$:

$$\frac{I(Q,t)}{I(Q,0)} = \frac{\langle \exp\{-iQ[r_i(t)-r_j(0)]\}\rangle}{\langle \exp\{-iQ[r_i(0)-r_j(0)]\}\rangle}$$

Types of Dynamics



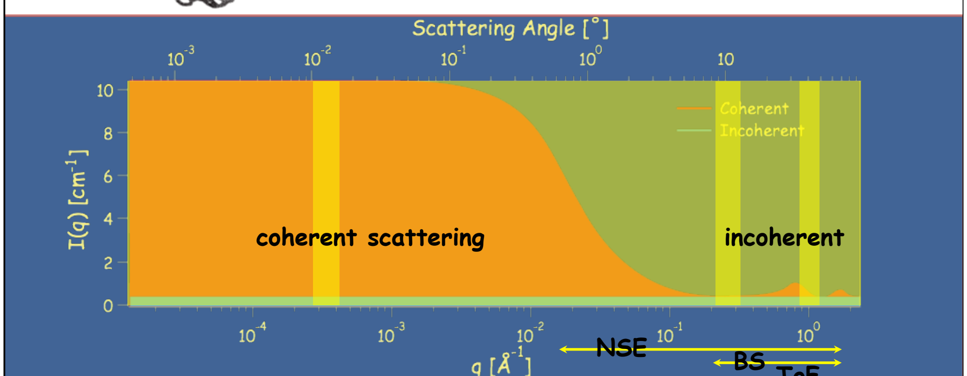
At length scales larger than the macromolecular size: Diffusion.

At length scales of the order the macromolecular size: Rotations and shape fluctuations.

Where the incoherent signal is dominant: Self H dynamics (translations, rotations, vibrations,…)

At the structural peaks: Atomic/Molecular Dynamics.

Lengthscales of Polymer Dynamics



larger scale objects: slower dynamics

coherent dynamics at low *Q* and at *Q* corresponding to relevant length scales
incoherent dynamics at high *Q*

D. Richter, et al., *Adv. in polym. Sci.*, **174**, 1 (2005).

Conclusions

Take home messages

- NSE studies dynamics in the ps to ns time range (4 orders of magnitude in time!), over lengthscales from tens of Ås to fractions of an Å. It covers the largest lengthscales and longest timescales of all neutron spectrometers.
- NSE works in the time domain. The instrumental resolution can be simply divided out.
- NSE works by encoding the neutron speed in its spin state. Do not depolarize the neutron beam.
- NSE is counting intensive. Large samples and/or significant scattering intensities are required.
- Good knowledge of the sample, in particular of its structure, is required for the preparation of the experiment and the understanding of the results.

Further Readings

- F. Mezei (ed.): Neutron Spin-Echo, *Lecture Notes in Physics*, 128, Springer, Heidelberg, 1980
- F. Mezei, C. Pappas, T. Gutberlet (Eds.): Neutron Spin-Echo Spectroscopy (2^{nd} workshop), *Lecture Notes in Physics,* 601, Springer, Heidelberg, 2003.
- D. Richter, M. Monkenbusch, A. Arbe, and J. Colmenero, "Neutron spin echo in polymer systems" *Adv. in polym. Sci*, **174**, 1 (2005).
- B. Farago, "Recent developments and applications of NSE in soft matter" *Curr. Opin. Colloid Interface Sci.*, **14**, 391 (2009).

Final Remarks

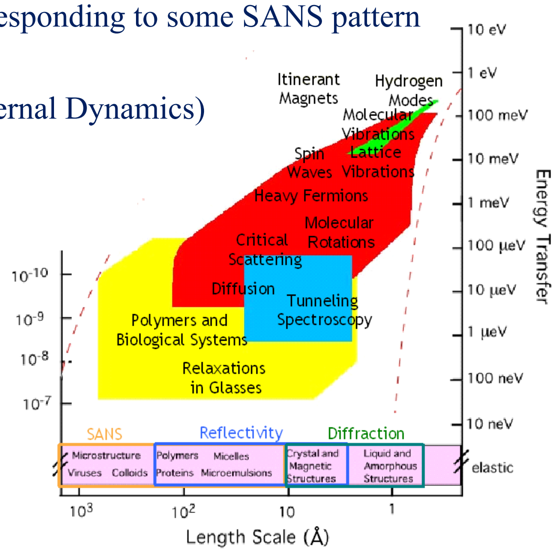
What Does NSE Study?

- Coherent Dynamics

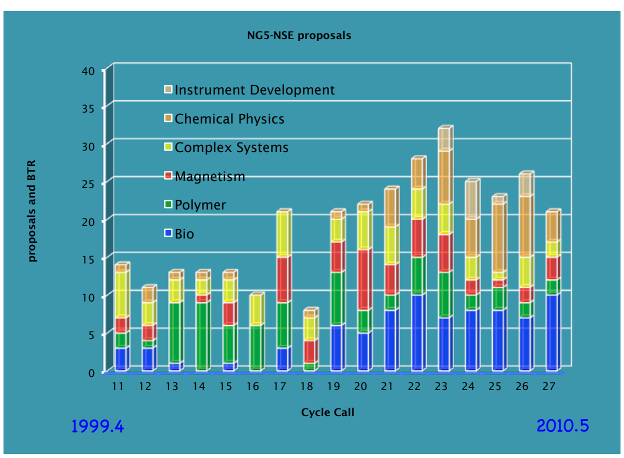
  Density Fluctuations corresponding to some SANS pattern
  - Diffusion
  - Shape Fluctuations (Internal Dynamics)
  - Polymer Dynamics
  - Glassy Systems
- Incoherent Dynamics

  Self-Dynamics (H atoms)
- Magnetic Dynamics

  Spin Glasses



NG5 NSE proposal statistics



Bio
- model bio-membrane
- protein motion

Polymer
- segment dynamics
- hydrogels
- polymer complex

Magnetism
- frustrated magnets
- spin glass

Chemical Physics
- confined water
- ionic liquids

Complex Fluids
- microemulsions
- surfactant membranes
- cluster dynamics