Neutron Spin Echo Spectroscopy (NSE)

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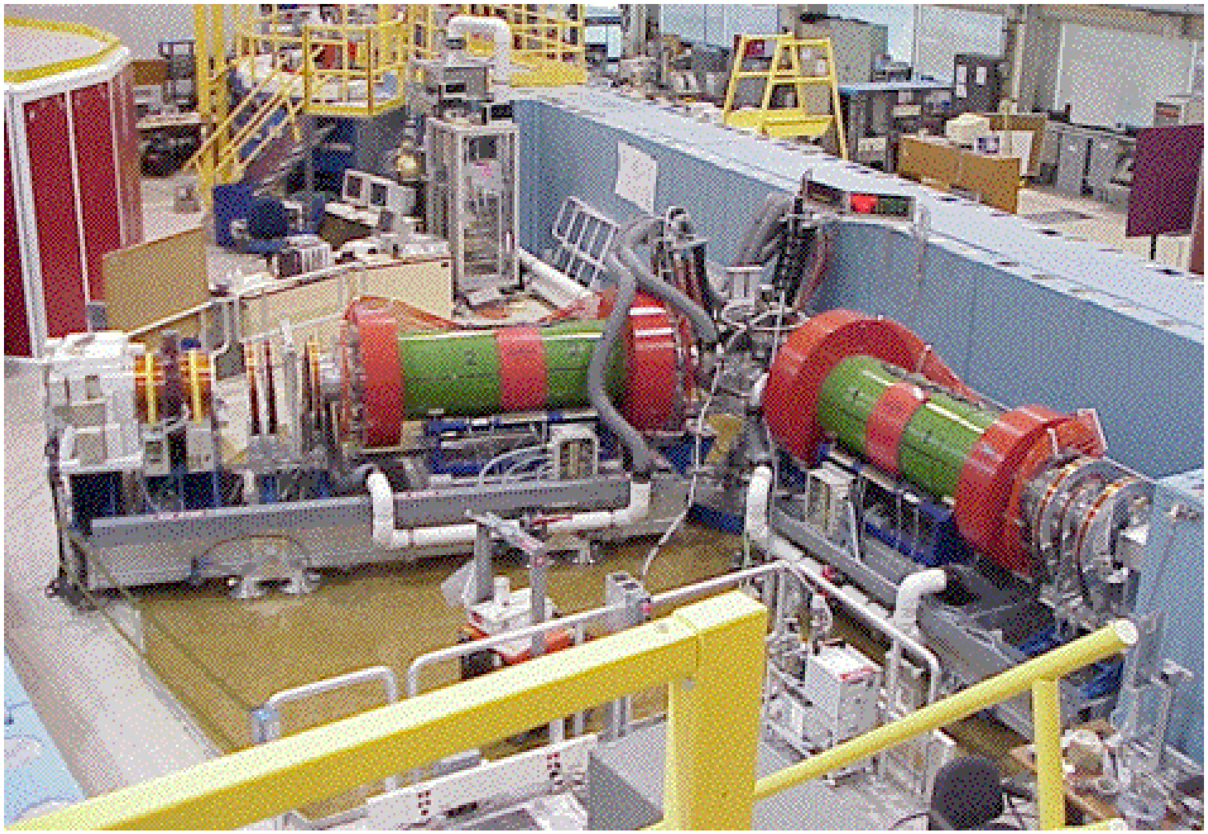

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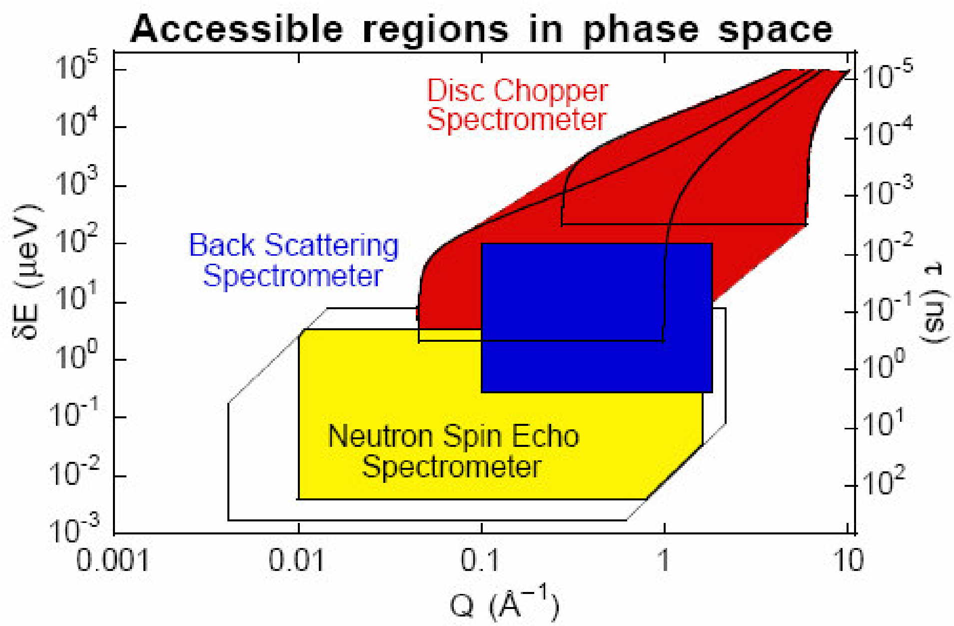
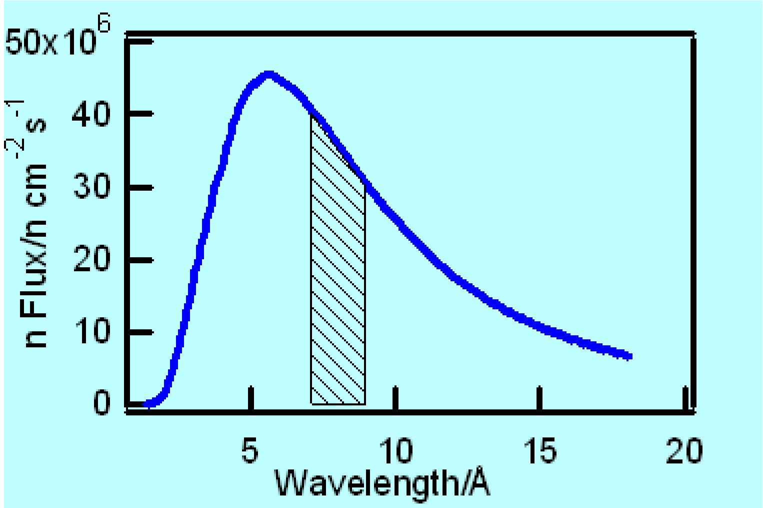
Why we need a magnetic field?



- **Goal:** $\delta E = 10^{-5} – 10^{-2}$ **meV** (very small!!!)
- We need low energy neutrons. Cold neutrons: $\lambda = 5 – 12$ Å, $E = 0.5 – 3.3$ meV.
- A "classical" inelastic technique works in two steps: preparation of the incoming monochromatic beam and analysis of the scattered beam.

- In Neutron spin echo the precessing neutron spin is employed as a kind of "individual" clock for each neutron. Thus, the velocity (energy) change of the neutrons can be measured directly in a single step.

- NSE technique allows the use of neutron beam wavelength spread $\Delta\lambda/\lambda = 5 – 20\%$, and therefore reasonably intense.



Neutron Flux along NG5 guide to NSE

Neutrons in magnetic fields: Precession

Mass, $m_n = 1.675\times10^{-27}$ kg

Spin, $S = 1/2$ [in units of $h/(2\pi)$]

Gyromagnetic ratio $\gamma = \mu_n/[S\times h/(2\pi)] =$

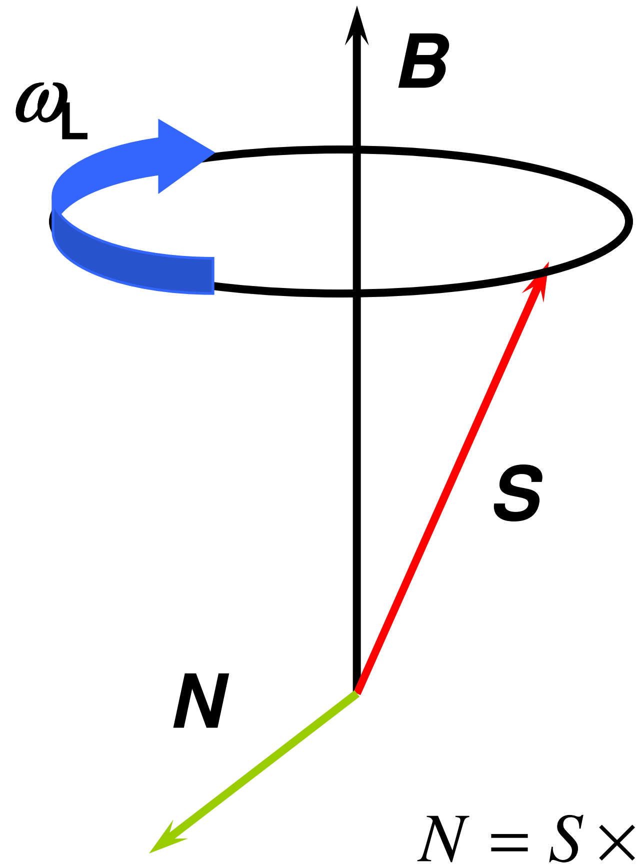
$1.832\times10^{8}\ s^{-1}T^{-1}$ (29.164 MHz T^{-1})

- The neutron will experience a torque from a magnetic field *B* perpendicular to its spin direction.

- Precession with the Larmor frequency:

$$\omega_L = \gamma B$$

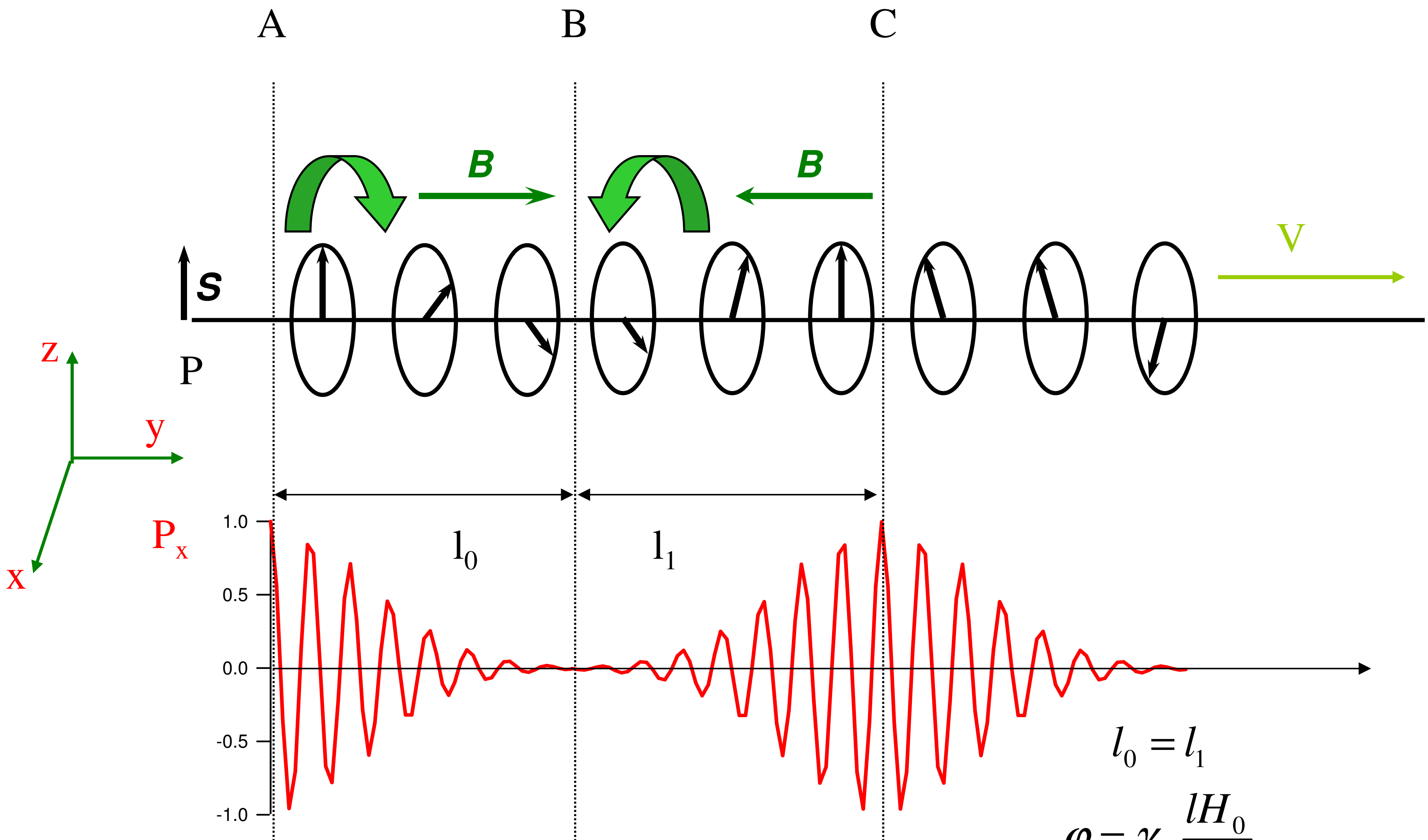
- The precession rate is predetermined by the strength of the field only.



$$N = S \times B$$

$$\frac{dS}{dt} = \gamma S \times B = S \times \omega_L$$

Spin echo effect



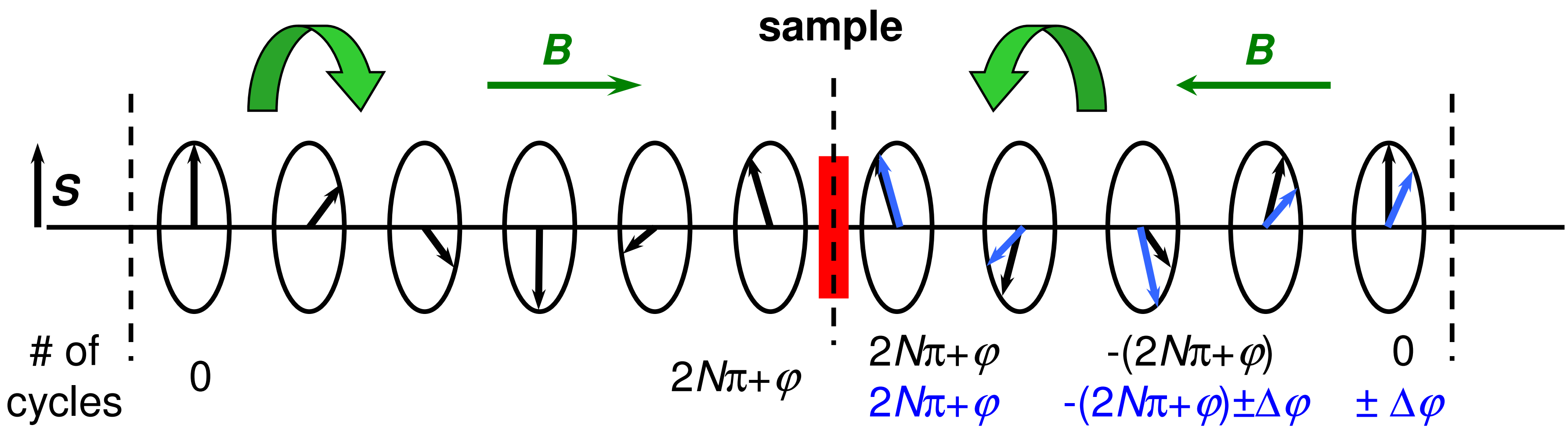
$$l_0 = l_1$$

$$\varphi = \gamma_L \frac{lH_0}{v}$$

$$P_x = \langle \cos\varphi \rangle = \int f(v) \cos\left[\frac{\gamma_L l H_0}{v}\right] dv$$

Monochromatic beam

- elastic scattering
- inelastic scattering



$$\varphi = \gamma B \frac{L}{v} \qquad \Delta\varphi = \gamma B L\left(\frac{1}{v} - \frac{1}{v'}\right) = \frac{\gamma B L \Delta v}{v^2}$$

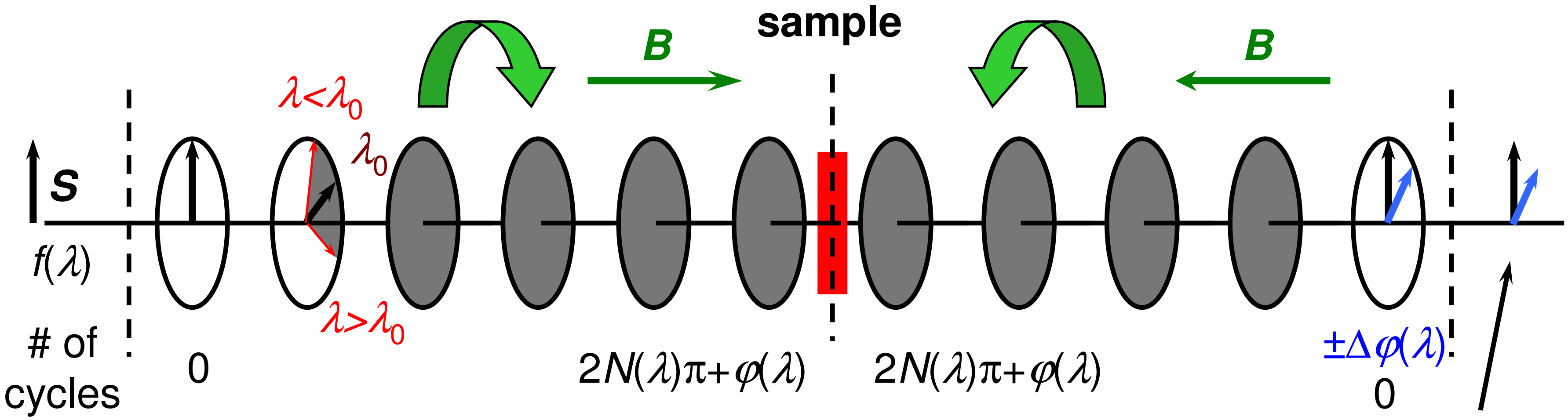
$$N(\lambda) = \frac{1}{2\pi}\int \frac{4\pi\gamma\mu_N B m\lambda}{h^2} dl = \frac{2\gamma\mu_N m\lambda}{h^2}\int B dl = 7370 \times J[T \cdot m] \times \lambda[\text{Å}]$$

$$J = \int B dl$$

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

$$\frac{\Delta v}{v} \approx \frac{1}{N} \approx 10^{-5}\ !$$

Polychromatic beam



The measured quantity is the spin component along z: $\cos(\Delta\varphi(\lambda))$:

$$N_0 \equiv N(\lambda_0);\text{then } N(\lambda) = N_0 \frac{\lambda}{\lambda_0}$$

$$\Delta\varphi(\lambda) = N_0 \frac{\delta\lambda}{\lambda_0} + \Delta N_0 \frac{\lambda}{\lambda_0} + \Delta N_0 \frac{\delta\lambda}{\lambda_0}.$$

Energy change

Asymmetry between coil field integrals

Neglect 2nd order terms for small asymmetries or quasielastic scattering

$$\cos\left[2\pi(\mathrm{N}_0\delta\lambda + \Delta N_0\lambda)/\lambda_0\right] =$$

$$= \cos\left(2\pi\mathrm{N}_0\frac{\delta\lambda}{\lambda_0}\right)\cos\left(2\pi\Delta N_0\frac{\lambda}{\lambda_0}\right) + \text{2nd order terms}$$

Neglected

The Principles of NSE

- If a spin rotates anticlockwise & then clockwise by the same amount it comes back to the same orientation

  - Need to reverse the direction of the applied field

  - Independent of neutron speed provided the speed is constant

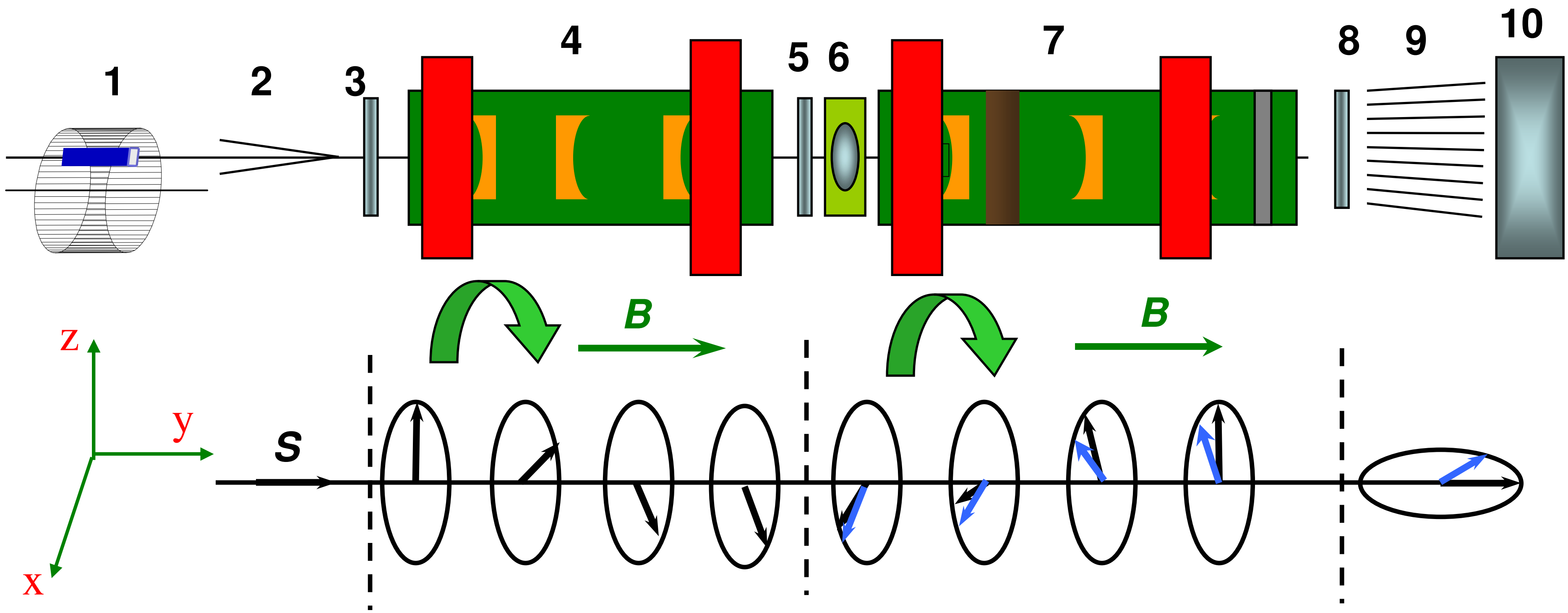
- The same effect can be obtained by reversing the precession angle at the mid-point and continuing the precession in the same sense

  - Use a π rotation

- If the neutron's velocity is changed by the sample, its spin will not come back to the same orientation

  - The difference will be a measure of the change in the neutron's speed or energy.

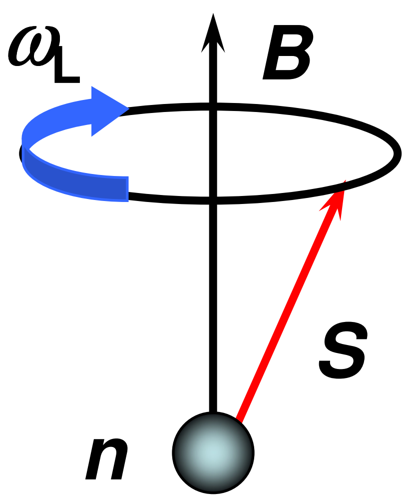
NSE Spectrometer schematic



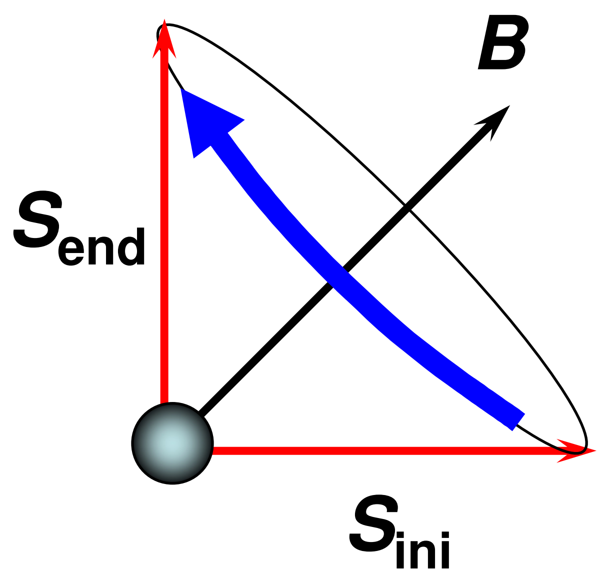
1. Velocity selector (selects neutron with certain λ_0)	5. π flipper (Provides phase inversion)	9. Polarization analyzer (radial array of polarizing supermirrors)
2. Polarizer (Polarizing supermirrors)	6. Sample	10. Area detector (20×20 cm^2)
3. $\pi/2$ flipper (starts Larmor precession)	7. Second main solenoid (phase and correction coils)	
4. First main solenoid (phase and correction coils)	8. $\pi/2$ flipper (stops Larmor precession)	

Spin flippers

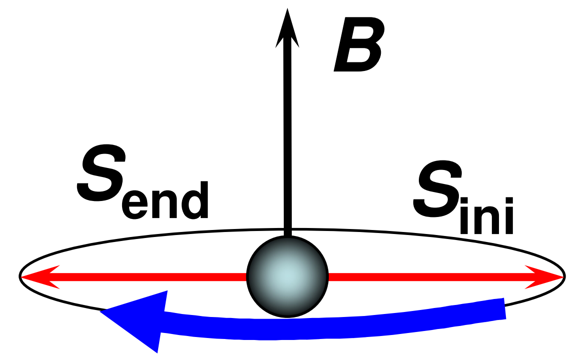
Precession



π/2 flipper



π flipper



Intensity at the detector

For a single wavelength:

$$\cos\left(2\pi\Delta N_0 \frac{\lambda}{\lambda_0}\right)\cos\left(2\pi \mathrm{N}_0 \frac{\delta\lambda}{\lambda_0}\right)$$

For wavelength distribution, $f(\lambda)$, with mean wavelength, λ_0:

$$\langle P\rangle = \int_0^\infty f(\lambda)\cos\left(2\pi\Delta N_0 \frac{\lambda}{\lambda_0}\right)\left[\int_{-\infty}^{\infty} S(\mathbf{Q},\omega)\cos(\omega t(\lambda))d\omega\right]d\lambda$$

$$\text{where } t \equiv \frac{N_0 m\lambda^3}{h\lambda_0} \quad \text{since } \delta\lambda = \frac{m\lambda^3}{2\pi h}\omega$$

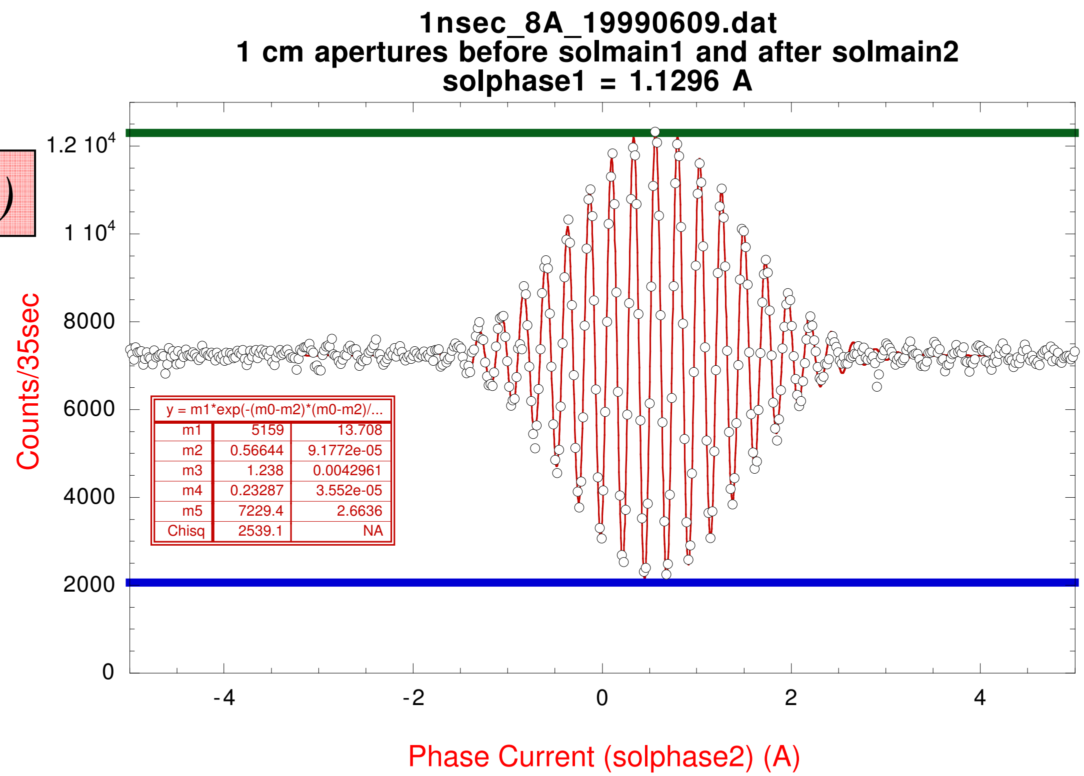
$$\left[\int_{-\infty}^{\infty} S(\mathbf{Q},\omega)\cos(\omega t(\lambda))d\omega\right]$$

Intermediate Scattering Function $I(Q,t)$

at $t = 0$

$$\langle P\rangle = \int_0^\infty f(\lambda)\cos\left(2\pi\Delta N_0 \frac{\lambda}{\lambda_0}\right)d\lambda$$

At small N_0 vary ΔN_0:
- Period gives λ_0
- Envelope gives $f(\lambda)$



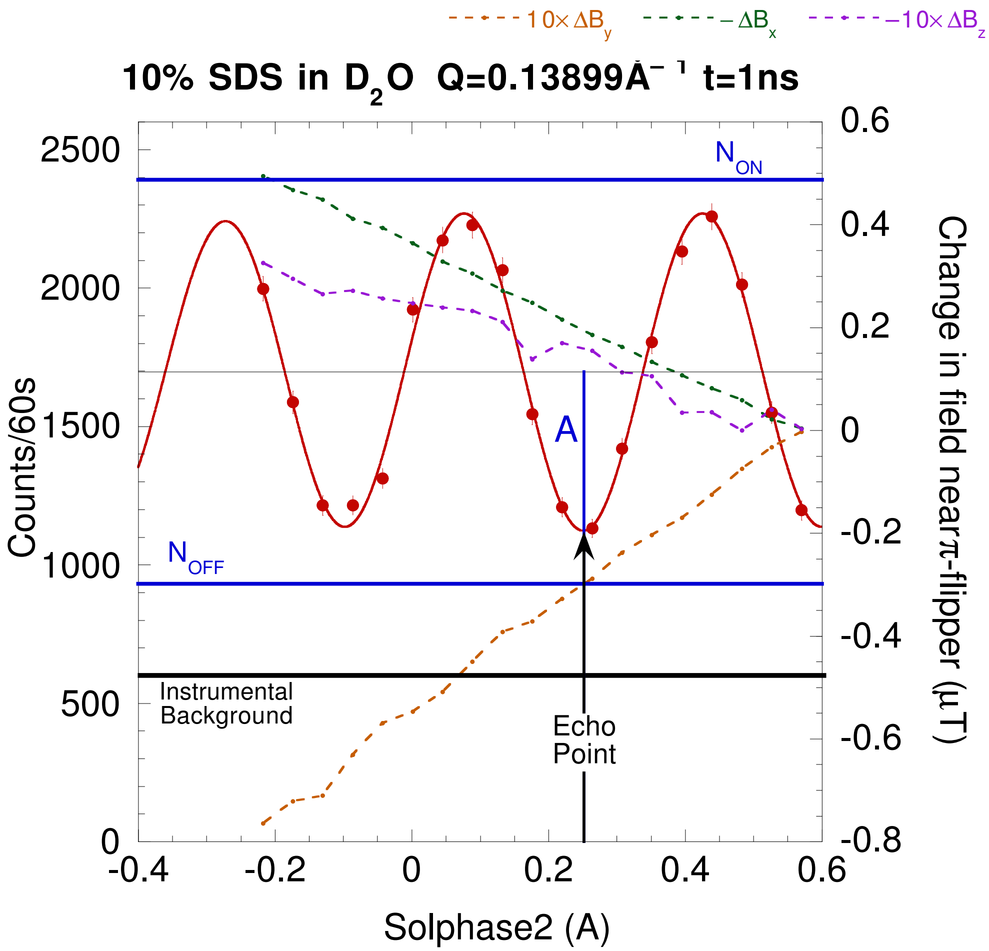
Measuring *I*(*Q*,*t*)

- The difference between the flipper ON and flipper OFF data gives $I(Q,0)$

- The echo is fit to a gaussian-damped cosine.

Signal before resolution correction is

$$\frac{2A}{N_{ON} - N_{OFF}}$$



How to deal with the resolution?

$$\langle P\rangle = \int_0^\infty f(\lambda)\cos\left(2\pi\Delta N_0 \frac{\lambda}{\lambda_0}\right)\left[\int_{-\infty}^{\infty} S(\mathbf{Q},\omega)\cos(\omega t(\lambda))d\omega\right]d\lambda$$

Inhomogeneities in the magnetic field may further reduce the polarization. Since they are not correlated with $S(Q,\omega)$ or $f(\lambda)$, their effect may be divided out by measuring the polarization from a purely elastic scatterer.

$$\left[\int_{-\infty}^{\infty} S(\mathbf{Q},\omega)\cos(\omega t(\lambda))d\omega\right] = I(\mathbf{Q},t(\lambda))$$

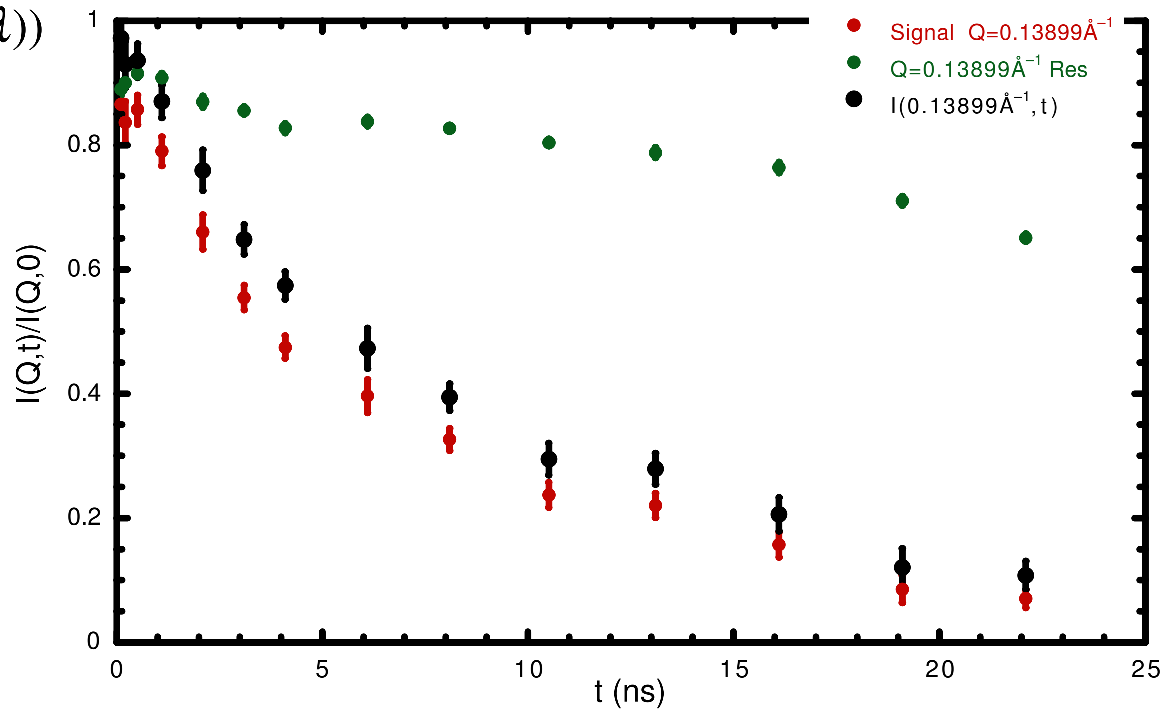
At the echo point, $\Delta N_0 = 0$,

$$\langle P\rangle = \int_0^\infty f(\lambda) I(\mathbf{Q},t(\lambda))d\lambda$$

$$J(\mathbf{Q},t) = I(\mathbf{Q},t)\cdot R(\mathbf{Q},t)$$

$$I(\mathbf{Q},t) = \frac{J(\mathbf{Q},t)}{R(\mathbf{Q},t)}$$

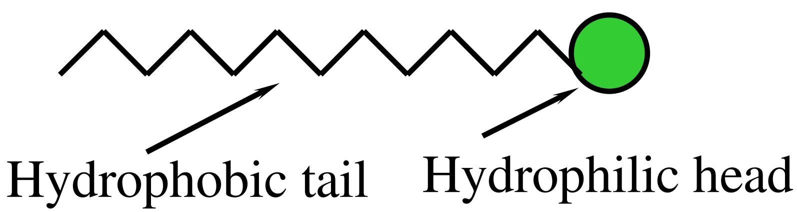
In the time domain the resolution is simply divided

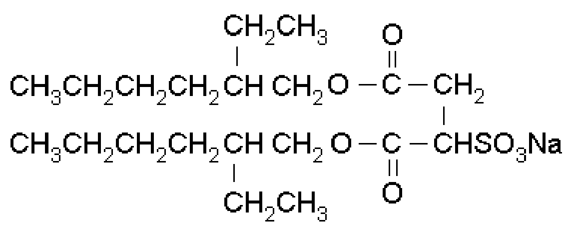


The main application of NSE is to measure the intermediate coherent scattering function $I_{coh}(Q,t)$, the coherent density fluctuations that correspond to some SANS intensity pattern.

- Diffusion
- Internal dynamics (shape fluctuations)
- …

Example: Diffusion of Surfactant Molecules

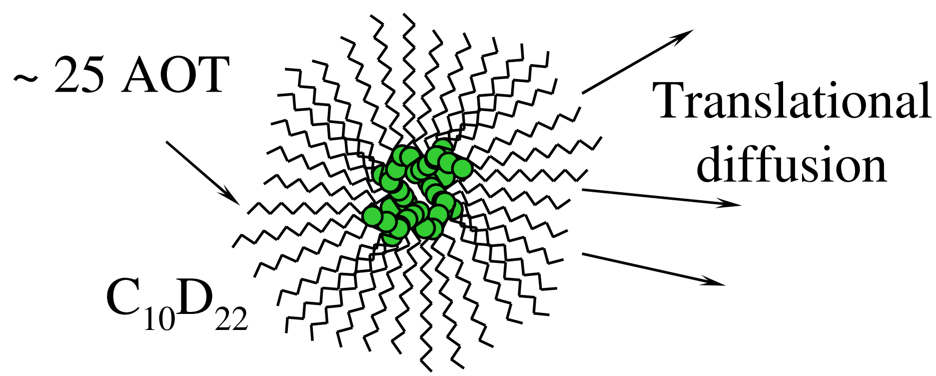




AOT

AOT micelles in n-decane ($C_{10}D_{22}$)

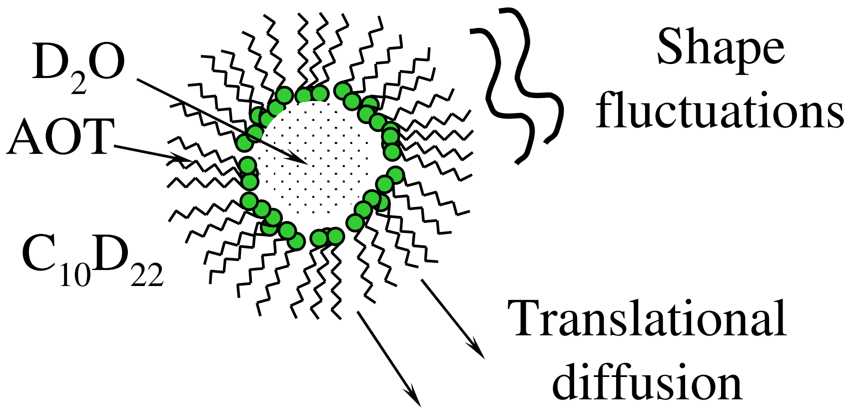
Inverse spherical micelle

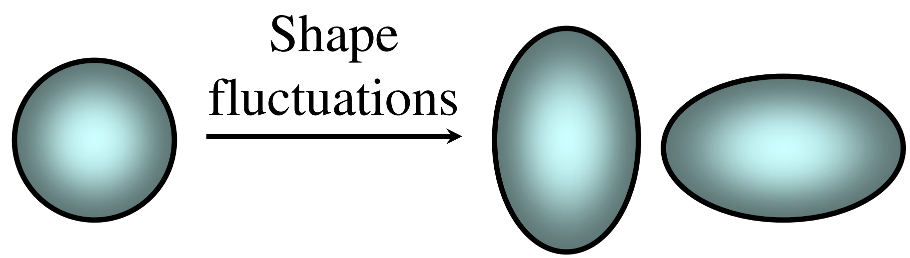


$$\frac{I(Q,t)}{I(Q,0)} = Exp\left[-D_{eff}Q^2t\right]$$

Experiment

Shape fluctuations in AOT/D_2O/C_6D_{14} inverse microemulsion droplet



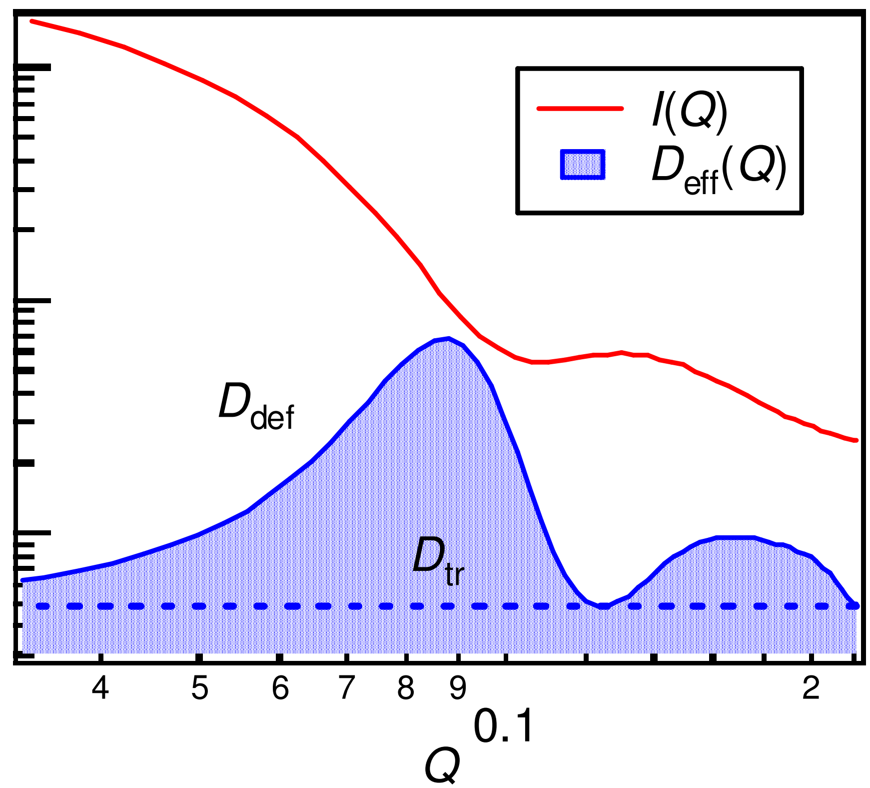


$$\frac{I(Q,t)}{I(Q,0)} = Exp\left[-D_{eff}(Q)Q^2 t\right]$$

$$D_{eff}(Q) = D_{tr} + D_{def}(Q) =$$

$$D_{tr} + \frac{5\lambda_2 f_2(QR_0)\left\langle |a_2|^2 \right\rangle}{Q^2\left[4\pi[j_0(QR_0)]^2 + 5 f_2(QR_0)\left\langle |a_2|^2 \right\rangle\right]}$$

$$f_2(QR_0) = 5[4 j_2(QR_0) + QR_0 j_3(QR_0)]$$



Experiment

$$D_{eff}(Q) = D_{tr} + \frac{5\lambda_2 f_2(QR_0)\left\langle |a_2|^2 \right\rangle}{Q^2\left[4\pi[j_0(QR_0)]^2 + 5f_2(QR_0)\left\langle |a_2|^2 \right\rangle\right]}$$

Goal: Bending modulus of elasticity

$$k = \frac{1}{48}\left[\frac{k_B T}{\pi p^2} + \lambda_2 \eta R_0^3 \frac{23\eta' + 32\eta}{3\eta}\right]$$

λ_2 – the damping frequency – **frequency of deformation**

$\langle |a|^2 \rangle$ – mean square displacement of the 2-nd harmonic – **amplitude of deformation**

p^2 – size polydispersity, measurable by SANS or DLS

η is the bulk viscosity of deuterated n-hexane

η' is the bulk viscosity of deuterated water