Neutron Spin Echo: Probing Dynamics in Complex Fluids

By

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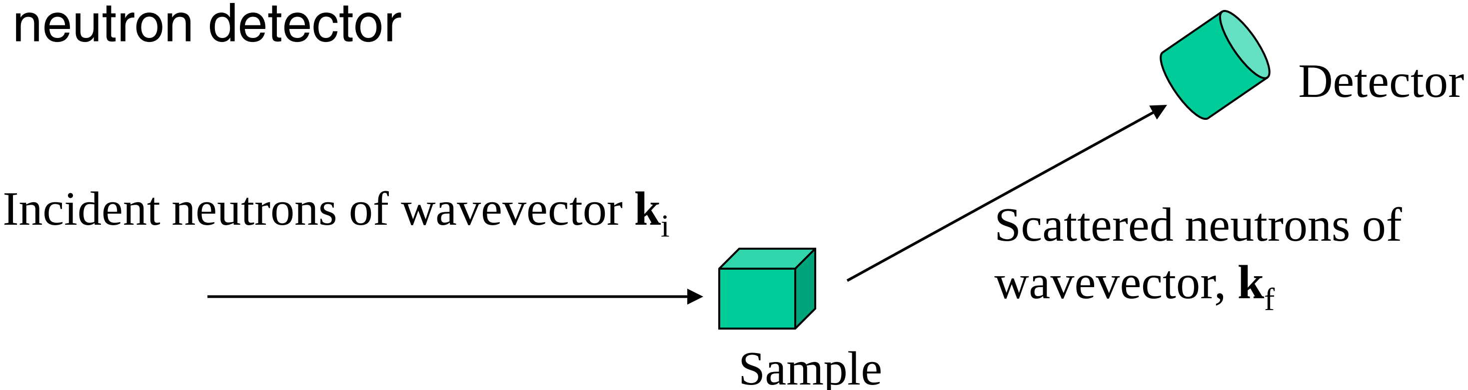
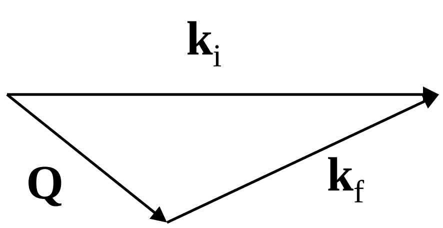
What Do We Need for a Basic Neutron Scattering Experiment?

- A source of neutrons
- A method to prescribe the wavevector of the neutrons incident on the sample
- (An interesting sample)
- A method to determine the wavevector of the scattered neutrons
- A neutron detector



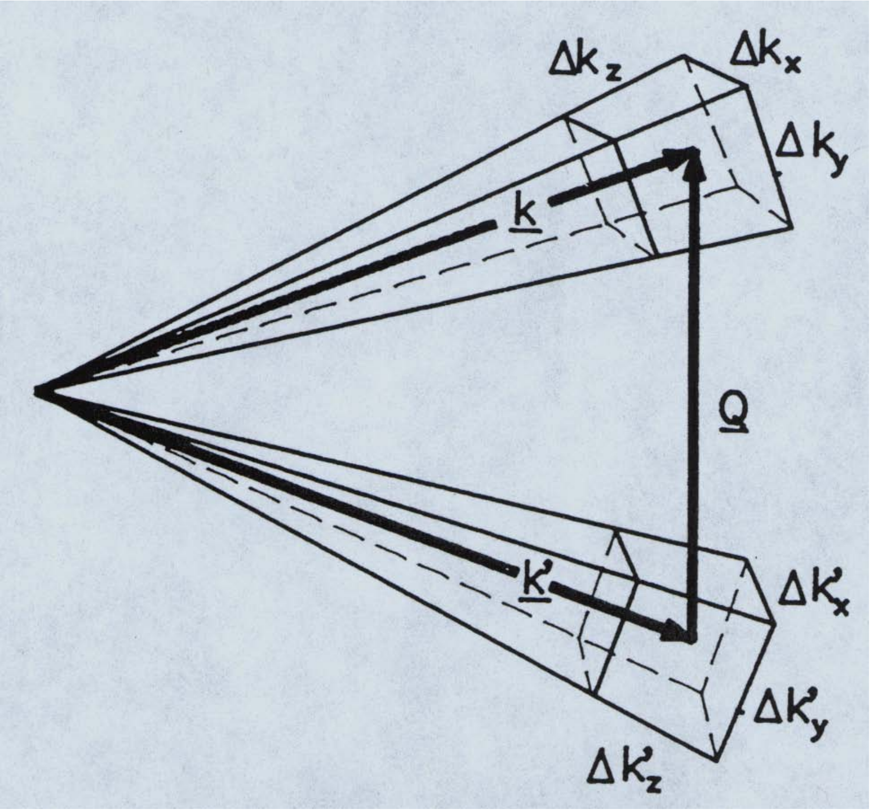


$$E = \hbar\omega = \frac{\hbar^2}{2m}(k_i^2 - k_f^2)$$

We usually measure scattering as a function of energy (E) and wavevector (**Q**) transfer

Instrumental Resolution

- Uncertainties in the neutron wavelength & direction of travel imply that Q and E can only be defined with a certain precision



- When the box-like resolution volumes in the figure are convolved, the overall resolution is Gaussian (central limit theorem) and has an elliptical shape in (Q,E) space

- The total signal in a scattering experiment is proportional to the phase space volume within the elliptical resolution volume – the better the resolution, the smaller the resolution volume and the lower the count rate

The Goal of Neutron Spin Echo is to Break the Inverse Relationship between Intensity & Resolution

- Traditional – define *both* incident & scattered wavevectors in order to define E and **Q** accurately

- Traditional – use collimators, monochromators, choppers etc to define both $\mathbf{k}_i$ and $\mathbf{k}_f$

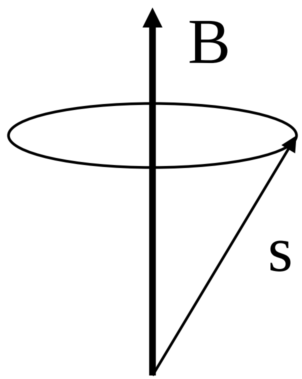
- NSE – measure as a function of the *difference* between appropriate components of $\mathbf{k}_i$ and $\mathbf{k}_f$ (original use: measure $k_i - k_f$ i.e. energy change)

- NSE – use the neutron's spin polarization to encode the difference between components of $\mathbf{k}_i$ and $\mathbf{k}_f$

- NSE – can use large beam divergence &/or poor monochromatization to increase signal intensity, while maintaining very good resolution

The Underlying Physics of Neutron Spin Echo (NSE) Technology is Larmor Precession of the Neutron's Spin

- The time evolution of the expectation value of the spin of a spin-1/2 particle in a magnetic field can be determined classically as:



$$\frac{d\vec{s}}{dt} = \gamma \vec{s} \wedge \vec{B} \qquad \Rightarrow \quad \omega_L = |\gamma| B$$

$$\gamma = -2913 * 2\pi \ \ Gauss^{-1}.s^{-1}$$

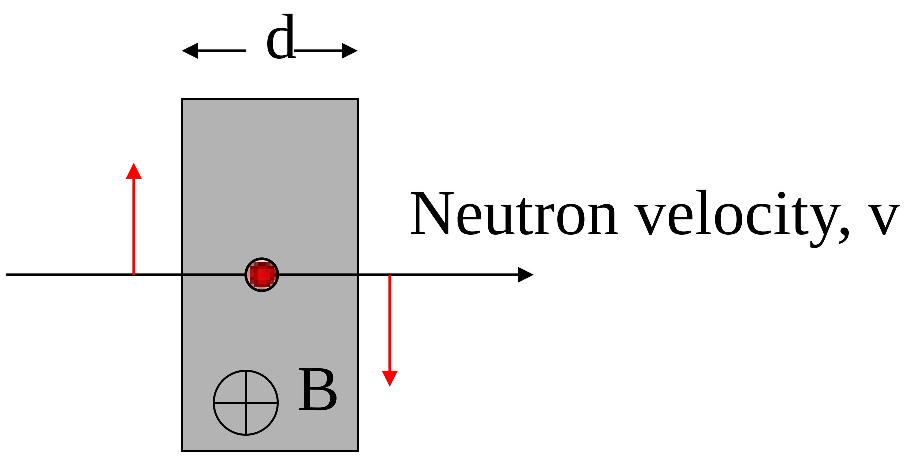
- The total precession angle of the spin, ϕ, depends on the time the neutron spends in the field: $\phi = \omega_L t$

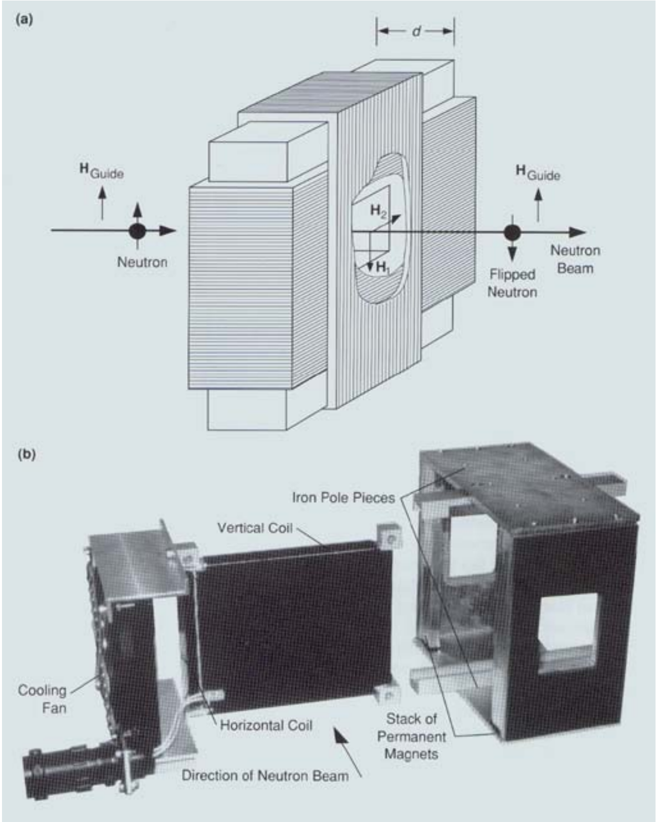
B(Gauss)	ω_L (10^3 rad.s^{-1})	N ($msec^{-1}$)	Turns/m for 4 Å neutrons
10	183	29	~29

Larmor Precession allows the Neutron Spin to be Manipulated using π or π/2 Spin-Turn Coils: Both are Needed for NSE

- The total precession angle of the spin, ϕ, depends on the time the neutron spends in the B field

$$\phi = \omega_L t = \gamma B d / \mathrm{v}$$



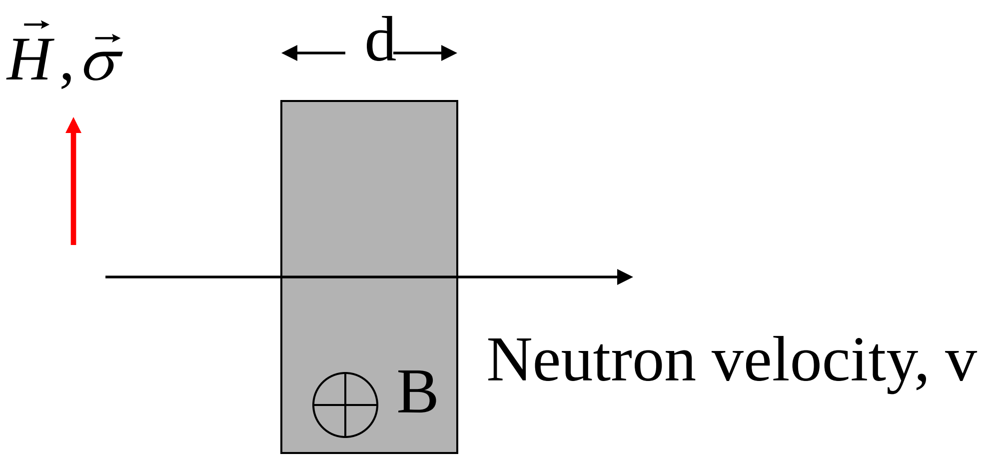


$$\text{Number of turns} = \frac{1}{135.65}.B[Gauss].d[cm].\lambda[Angstroms]$$

Neutron Spin Echo (NSE) uses Larmor Precession to "Code" Neutron Velocities

- A neutron spin precesses at the Larmor frequency in a magnetic field, B. $\omega_L = \gamma B$
- The total precession angle of the spin, ϕ, depends on the time the neutron spends in the field

$$\phi = \omega_L t = \gamma B d / \mathrm{v}$$



$$\text{Number of turns} = \frac{1}{135.65} . B[Gauss] . d[cm] . \lambda[Angstroms]$$

The precession angle ϕ is a measure of the neutron's speed v

The Principles of NSE are Very Simple

- 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, v, 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

Simulations

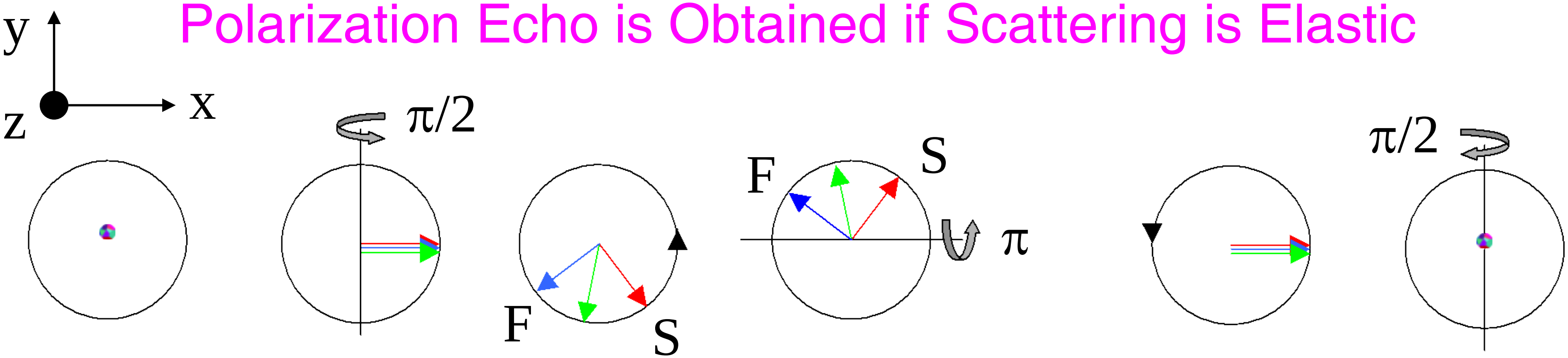
Classical picture

- Single neutron
- Neutrons of different velocities

Quantum Mechanical Picture

- Quasi-elastic scattering
- Inelastic scattering

In NSE*, Neutron Spins Precess Before and After Scattering & a Polarization Echo is Obtained if Scattering is Elastic



Initially, neutrons are polarized along z

Rotate spins into x-y precession plane

Allow spins to precess around z: slower neutrons precess further over a fixed path-length

Rotate spins through π about x axis

Elastic Scattering Event

Allow spins to precess around z: all spins are in the same direction at the echo point if ΔE = 0

Rotate spins to z and measure polarization

$$\textit{Final Polarization}, P = \langle \cos(\phi_1 - \phi_2) \rangle$$

* F. Mezei, Z. Physik, 255 (1972) 145

For Quasi-elastic Scattering, the Echo Polarization depends on Energy Transfer

- If the neutron changes energy when it scatters, the precession phases before & after scattering, ϕ_1 & ϕ_2, will be different:

$$\text{using} \qquad \hbar\omega = \frac{1}{2} m(v_1^2 - v_2^2) \approx mv\delta v$$

$$\phi_1 - \phi_2 = \gamma B d\left(\frac{1}{v_1} - \frac{1}{v_2}\right) \approx \frac{\gamma B d}{v^2}\delta v \approx \frac{\gamma B d \hbar\omega}{mv^3} = \frac{\gamma B d m^2 \lambda^3 \omega}{2\pi h^2}$$

- To lowest order, the difference between ϕ_1 & ϕ_2 depends only on ω (I.e. $v_1 - v_2$) & *not* on v_1 & v_2 separately
- The measured polarization, <P>, is the average of $\cos(\phi_1 - \phi_2)$ over all transmitted neutrons I.e.

$$\langle P \rangle = \frac{\iint I(\lambda) S(\vec{Q},\omega)\cos(\phi_1 - \phi_2)\, d\lambda\, d\omega}{\iint I(\lambda) S(\vec{Q},\omega)\, d\lambda\, d\omega}$$

Neutron Polarization at the Echo Point is a Measure of the Intermediate Scattering Function

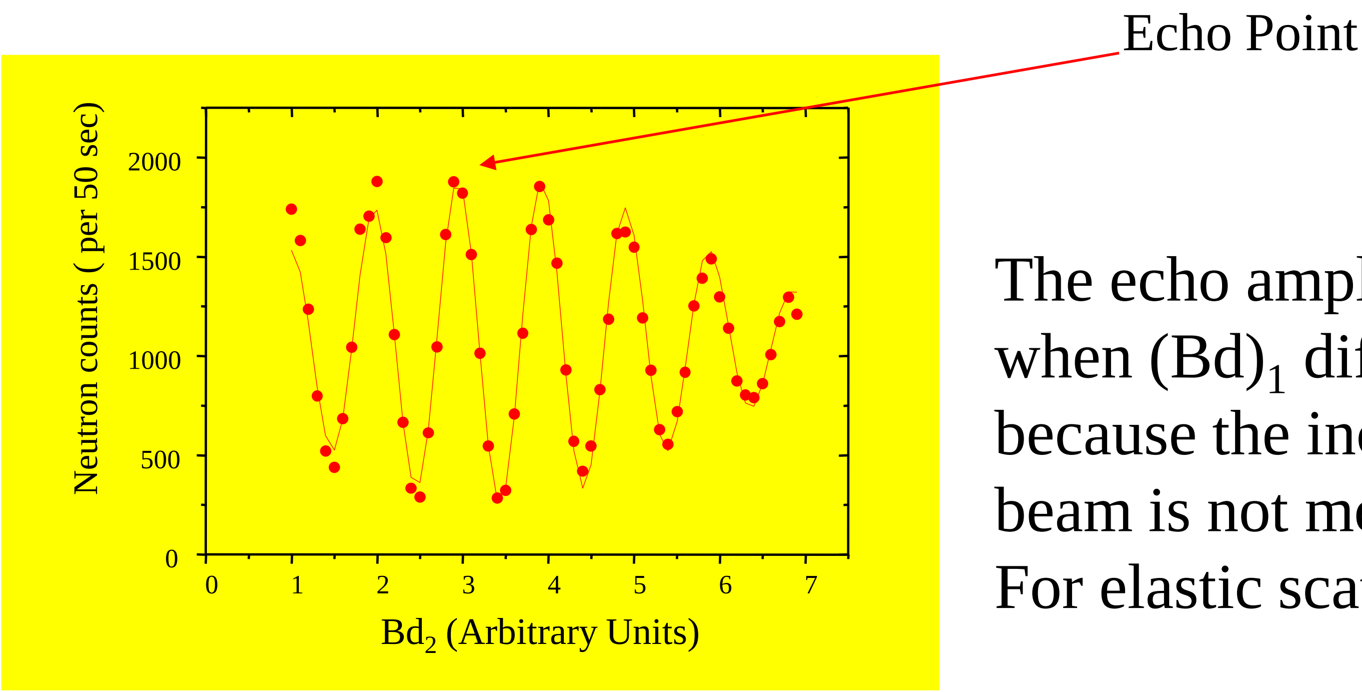
$$\langle P \rangle = \frac{\iint I(\lambda) S(\vec{Q},\omega)\cos(\phi_1 - \phi_2)\, d\lambda\, d\omega}{\iint I(\lambda) S(\vec{Q},\omega)\, d\lambda\, d\omega} \approx \left\langle \int S(\vec{Q},\omega)\cos(\omega\tau)\, d\omega \right\rangle = I(\vec{Q},\tau)$$

where the "spin echo time" $\tau = \gamma Bd \dfrac{m^2}{2\pi h^2}\lambda^3$

Bd (T.m)	λ (nm)	τ (ns)
1	0.4	12
1	0.6	40
1	1.0	186

- $I(\vec{Q},t)$ is called the intermediate scattering function
  - Time Fourier transform of $S(\vec{Q},\omega)$ or the $\vec{Q}$ Fourier transform of $G(\vec{r},t)$, the two particle correlation function
- NSE probes the sample dynamics as a function of time rather than as a function of ω
- The spin echo time, τ, is the "correlation time"

Neutron Polarization is Measured using an Asymmetric Scan around the Echo Point



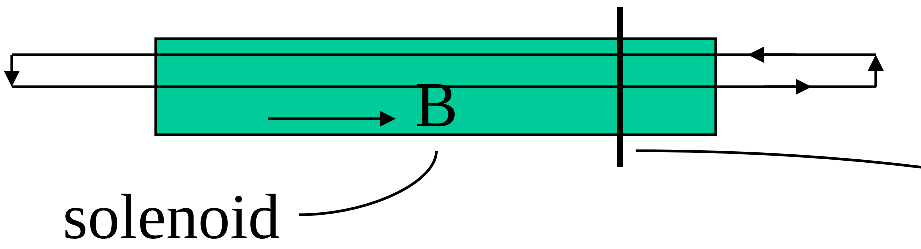
The echo amplitude decreases when $(Bd)_1$ differs from $(Bd)_2$ because the incident neutron beam is not monochromatic. For elastic scattering:

$$\langle P \rangle \sim \int I(\lambda) \cos\left[\frac{\gamma m}{h}\{(Bd)_1 - (Bd)_2\}\lambda\right].d\lambda$$

Because the echo point is the same for all neutron wavelengths, we can use a broad wavelength band and enhance the signal intensity

Field-Integral Inhomogeneities cause τ to vary over the Neutron Beam: They can be Corrected

- Solenoids (used as main precession fields) have fields that vary as r^2 away from the axis of symmetry because of end effects (div B = 0)

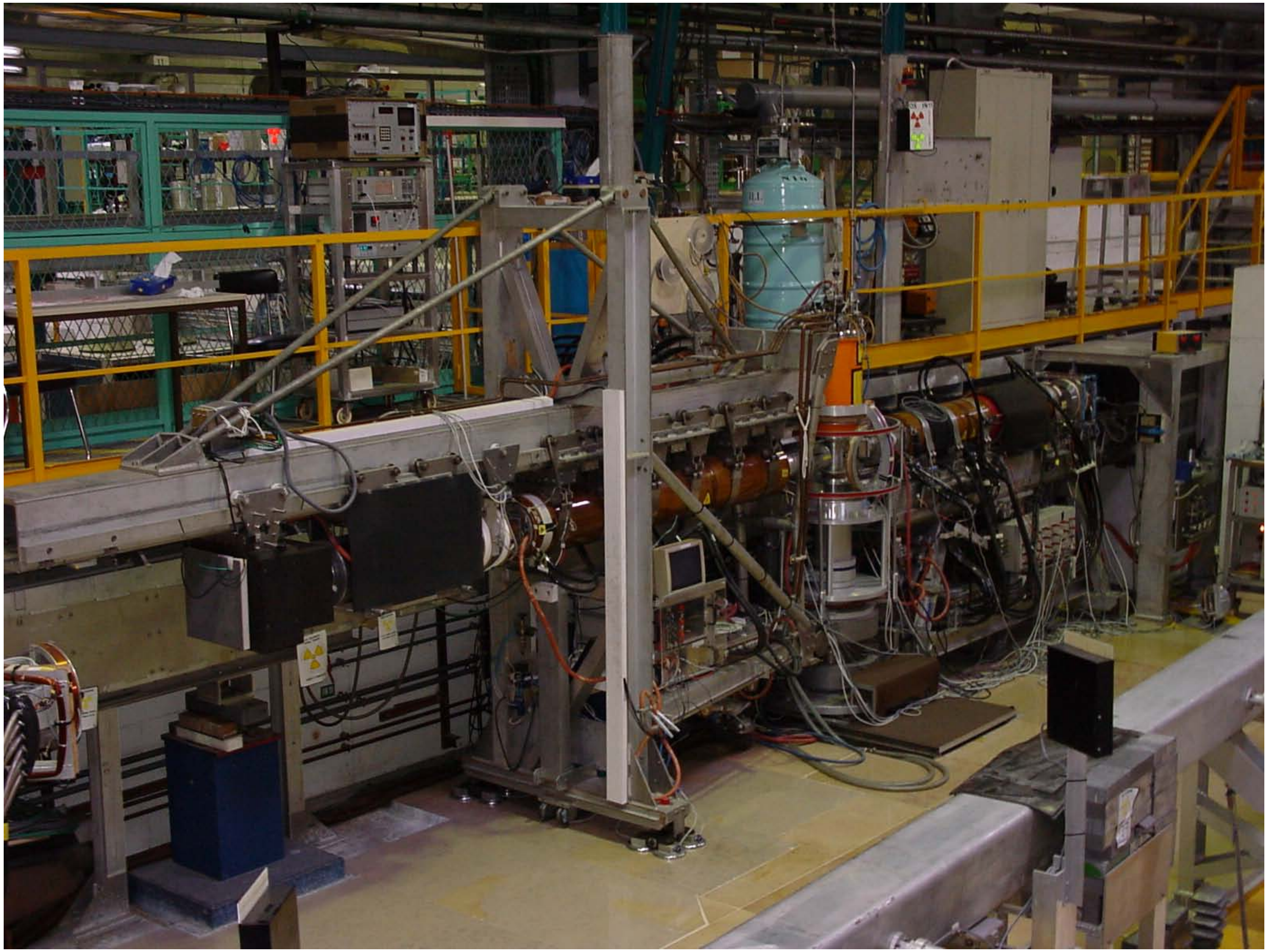


- According to Ampere's law, a current distribution that varies as r^2 can correct the field-integral inhomogeneities for parallel paths

- Similar devices can be used to correct the integral along divergent paths

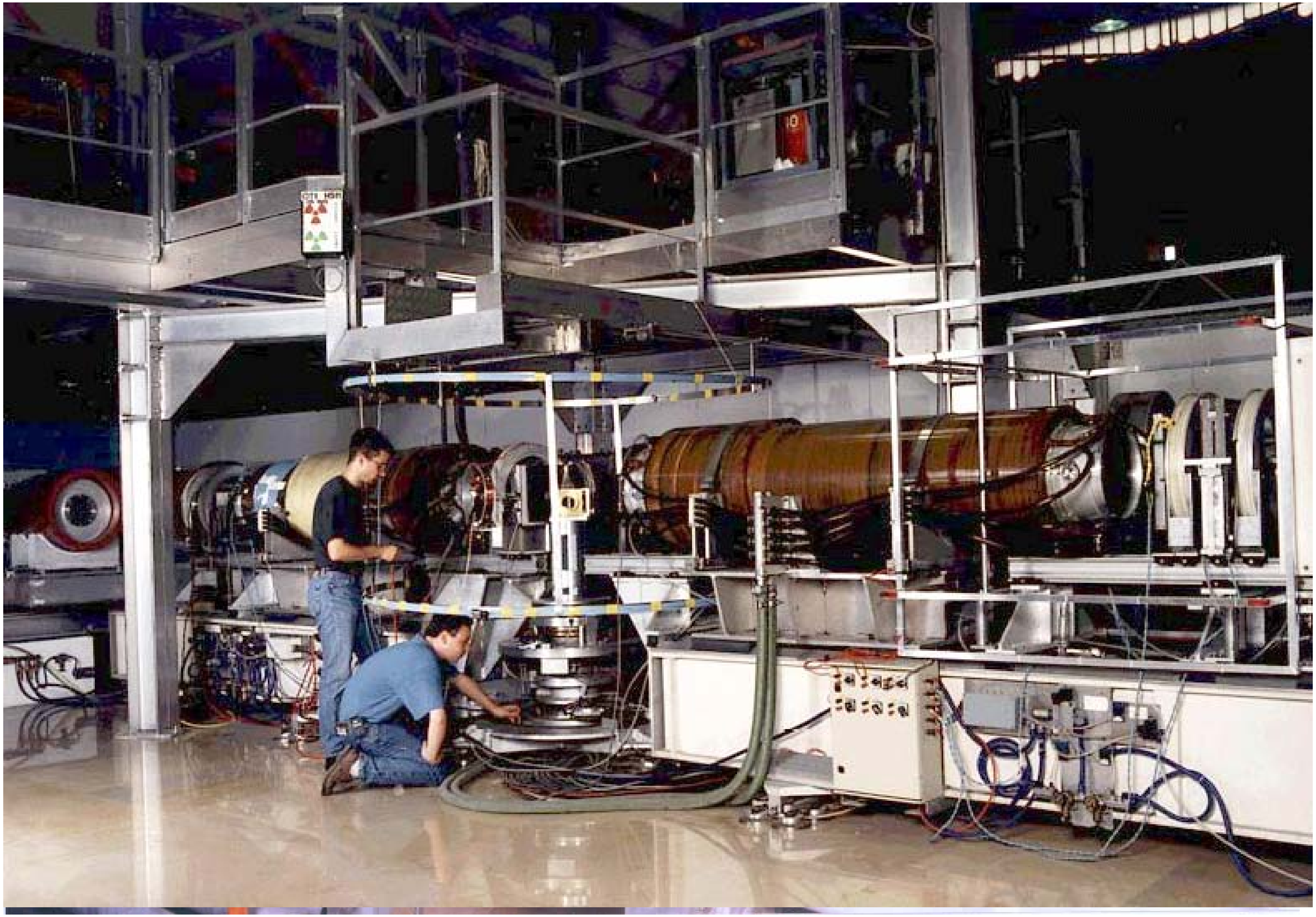
Fresnel correction coil for IN15

What does a NSE Spectrometer Look Like? IN11 at ILL was the First



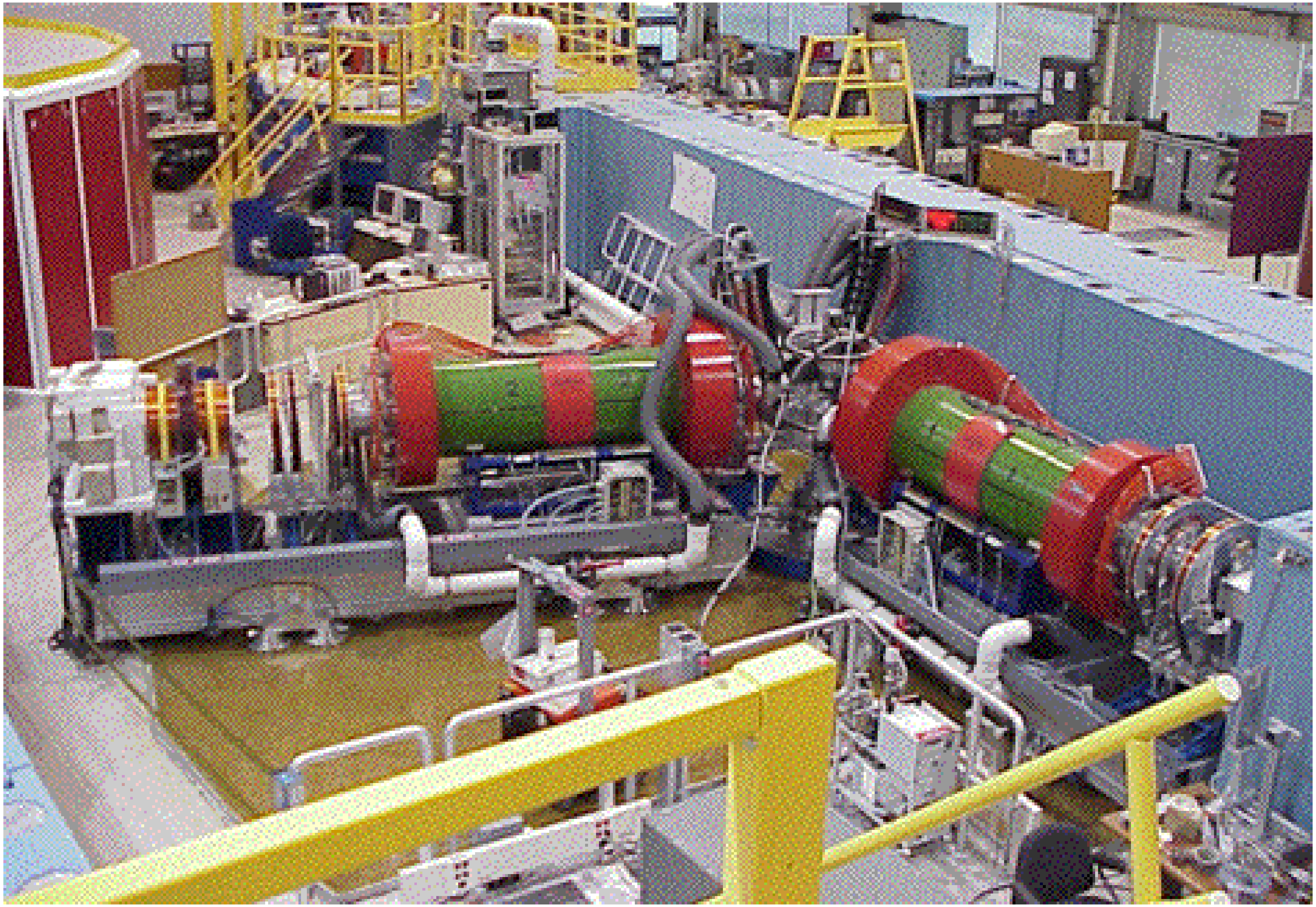
$\tau_{max} \sim 50$ ns at $\lambda = 10$ Å

IN-11C and IN15

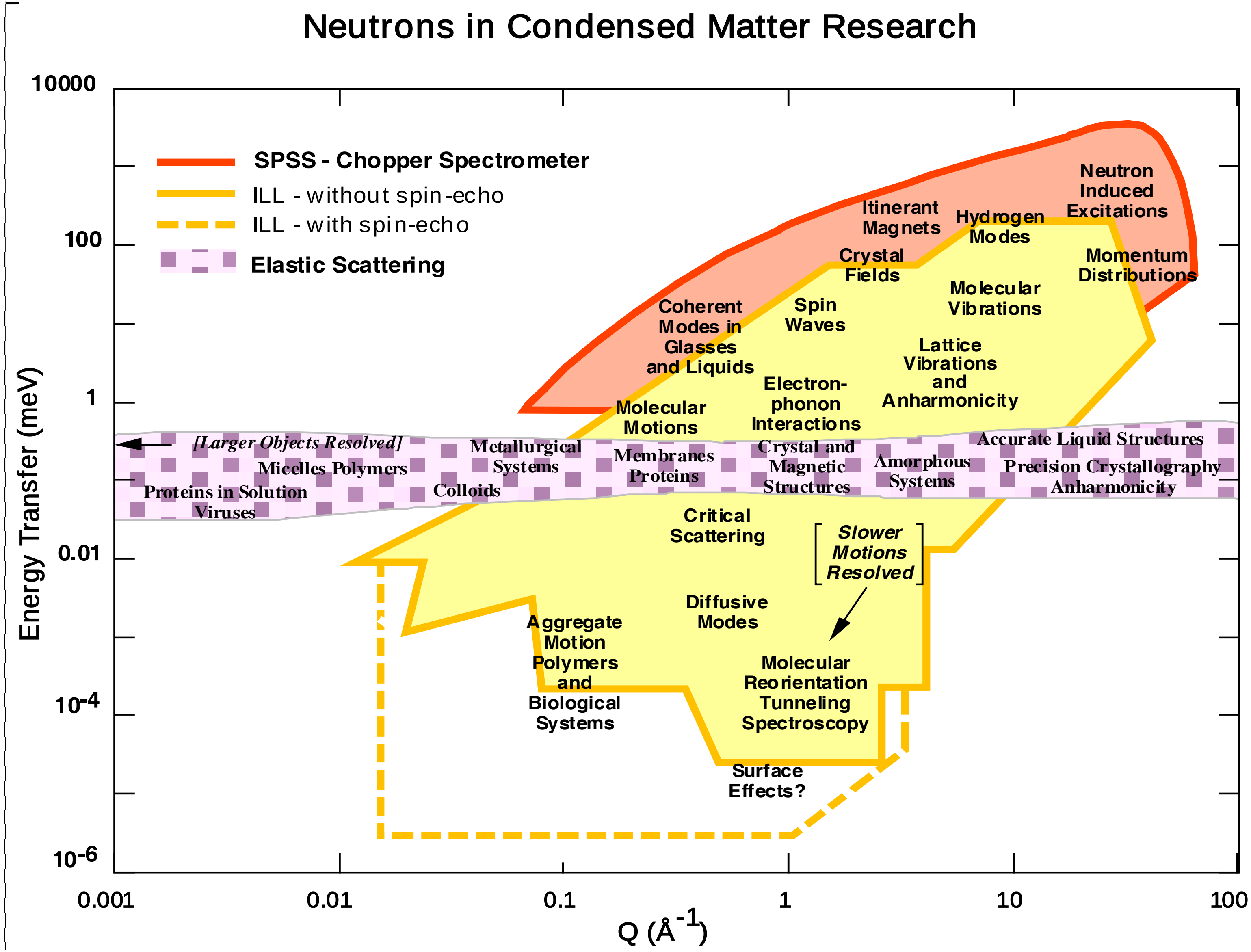


τ_{max} ~ 12 ns at λ = 10Å for IN11-C and τ_{max} ~ 400 ns at λ = 15Å

NSE is also available at the NCNR



$\tau_{max} \sim 50$ ns

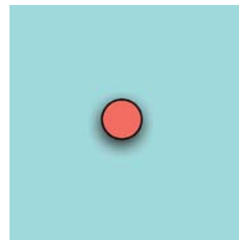


Neutron Spin Echo has significantly extended the (Q,E) range to which neutron scattering can be applied

Something Simple: A Single Diffusing Particle*

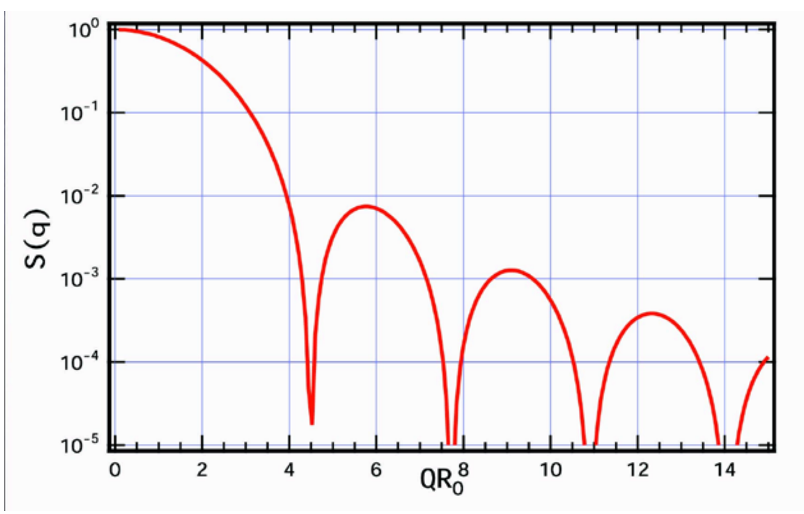
$$S(\vec{Q}) = \left\langle \sum_{i,j} b_i b_j e^{-\vec{Q}.(\vec{r}_i - \vec{r}_j)} \right\rangle$$

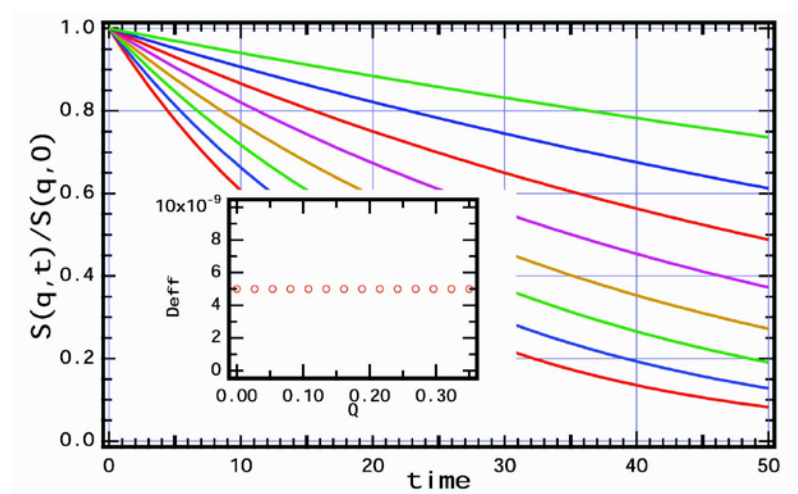
$$S(\vec{Q}) = \left(3\rho R^3 \frac{j_1(QR)}{QR} \right)^2$$



$$S(\vec{Q},t) = \left\langle \sum_{i,j} b_i b_j e^{i\vec{Q}.[\vec{r}_i(0) - \vec{r}_j(t)]} \right\rangle$$

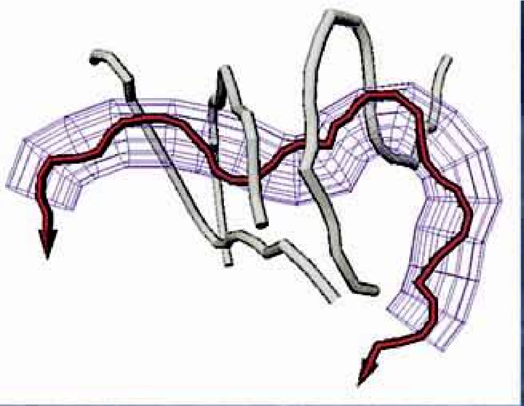
$$S(\vec{Q},t) = S(\vec{Q}) e^{-DQ^2 t}$$





*Viewgraph courtesy of B. Farago

Polymer Reptation*



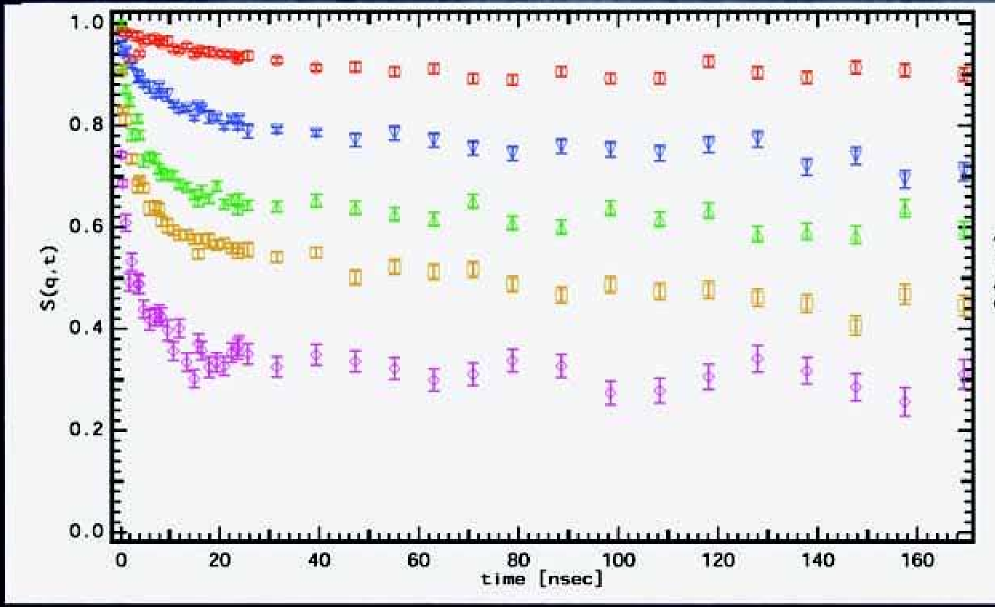
10% marked polymer chain(H) in deuterated matrix of the same polymer melt

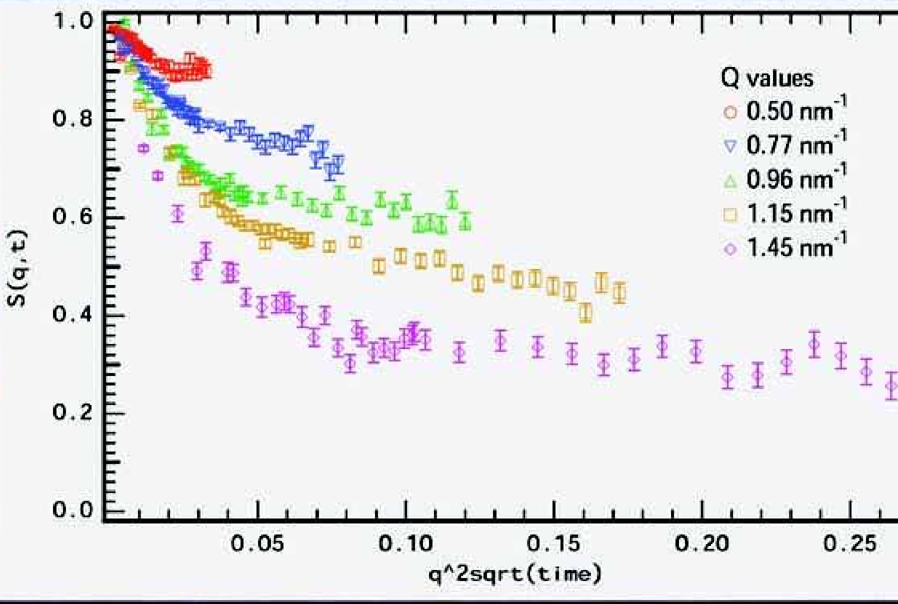
at short time => Rouse dynamics 1/tau ~ q^4

at longer times starts to feel the "tube" formed by the other chains (deGennes)

D. Richter, B. Ewen, B. Farago, et al., Physical Review Letters **62**, 2140 (1989).

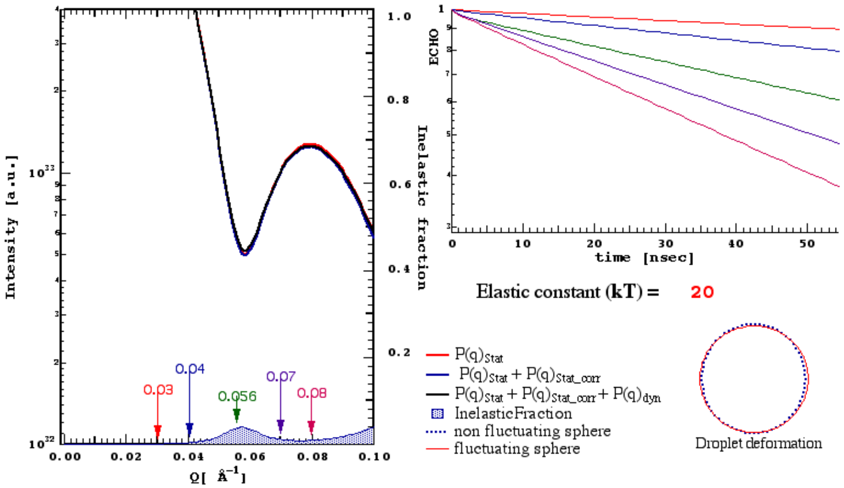
*viewgraph courtesy of B. Farago

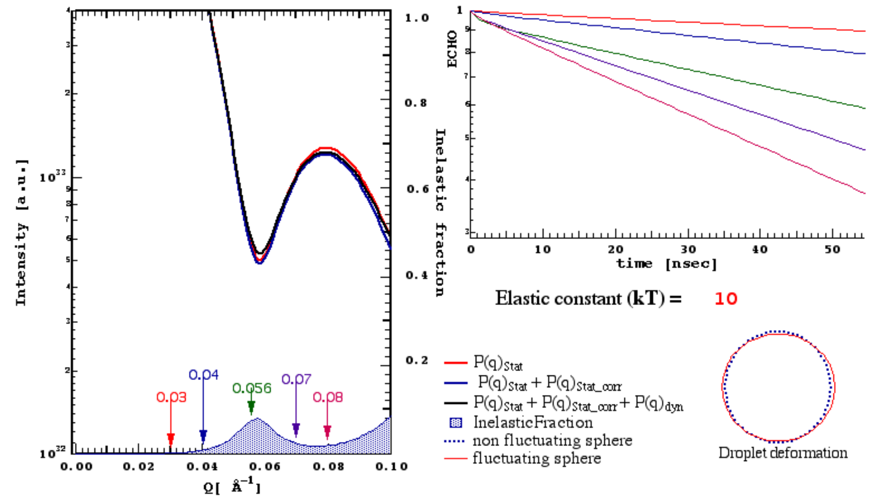


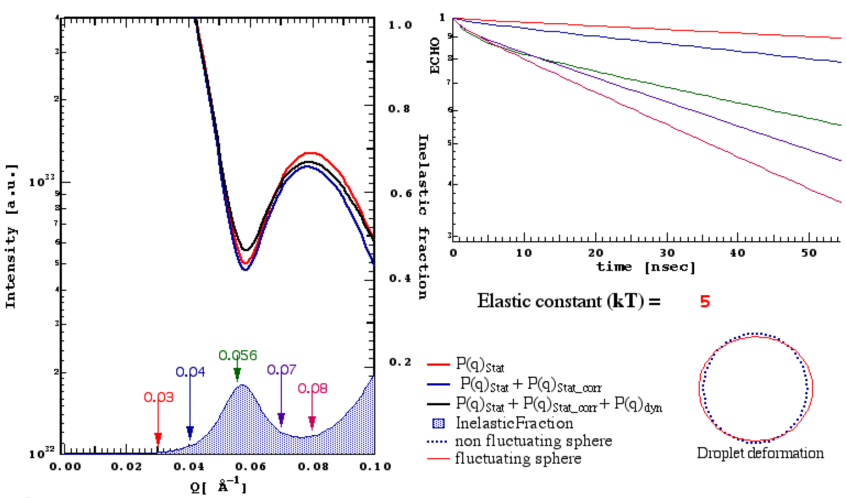


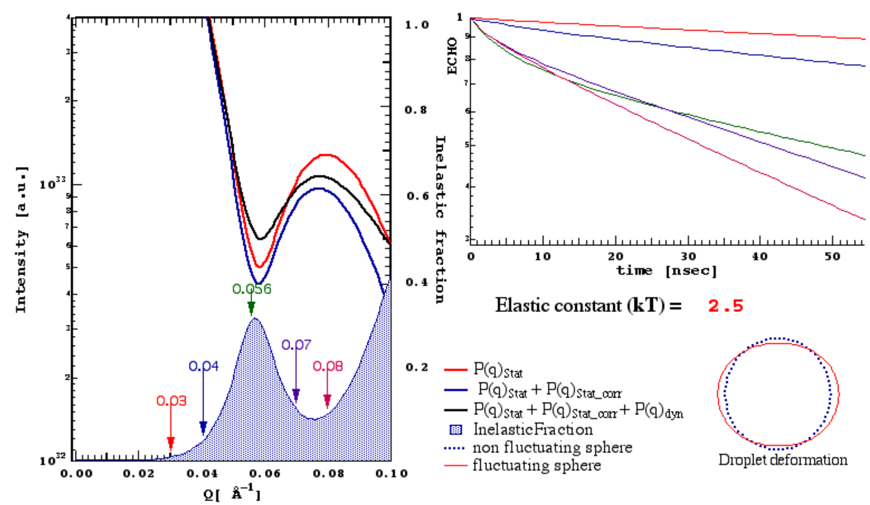
P. Schleger, B. Farago, C. Lartigue, et al., Physical Review Letters **81**, 124 (1998).

Neutron Spin Echo study of Deformations of Spherical Droplets*





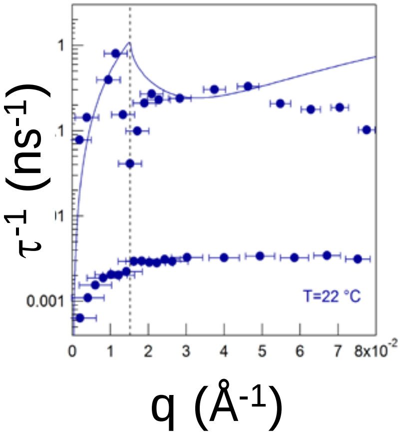




* Courtesy of B. Farago

Mesoscopic Membrane Fluctuations

Dispersion relation

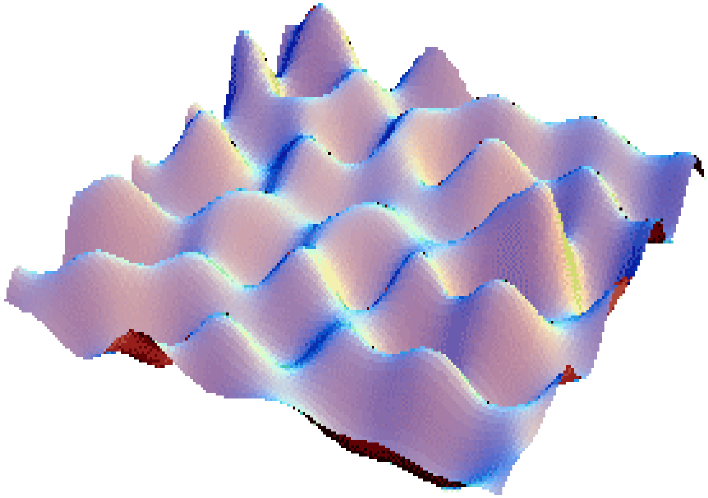


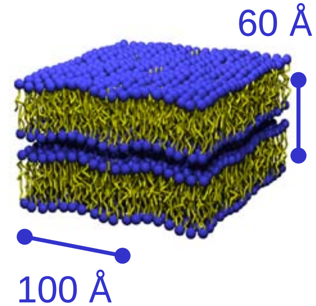
Contains ‘dynamic’ information

q-dependence of excitation frequencies

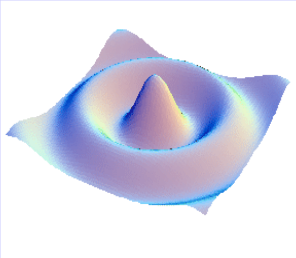
and relaxation rates

Thermal membrane fluctuations



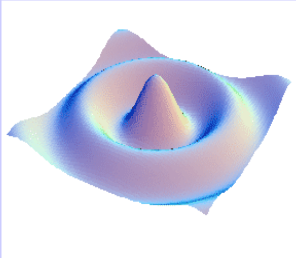


Elementary excitations



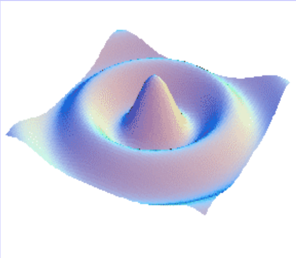
Propagating

+



Oscillating

+

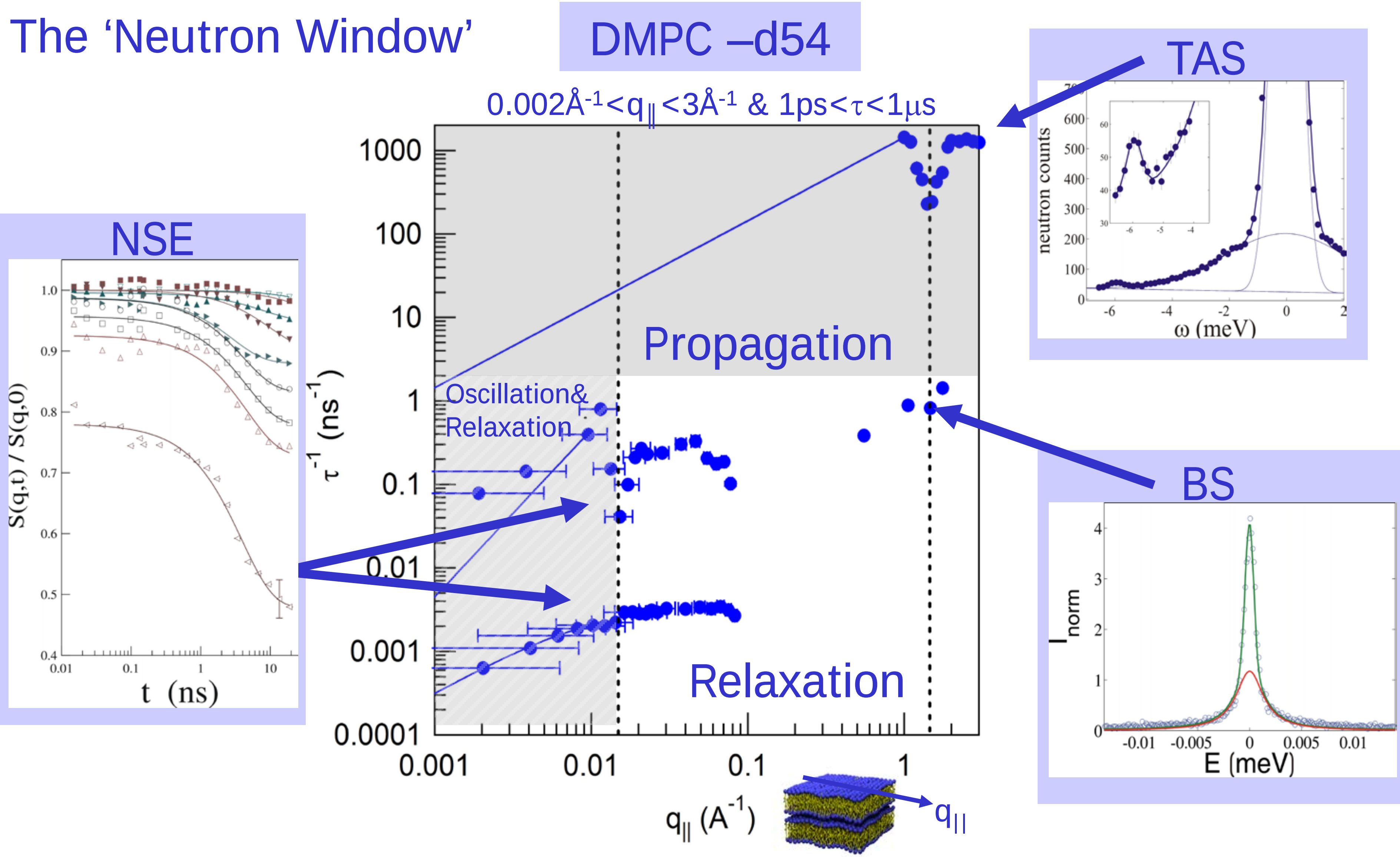


Relaxing

Mode

Collective Excitations in Model Membranes*

The ‘Neutron Window’



*Measurements made by M. Rheinstadter

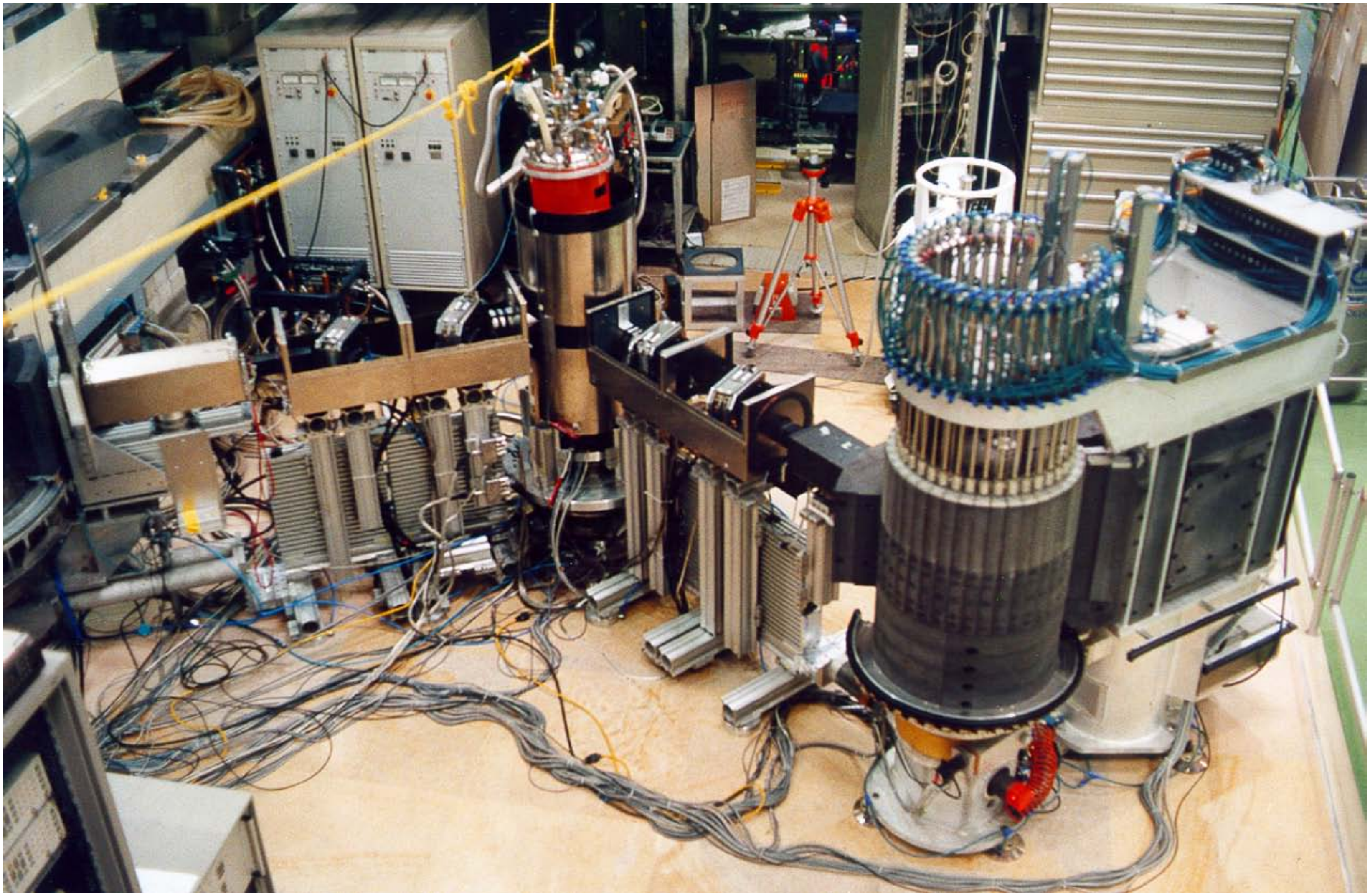
Other Larmor Precession Methods

- Neutron resonance spin echo (NRSE)
  - Very similar to traditional NSE
  - Can also be added to a triple axis spectrometer for “phonon focusing”
  - Available at several European centers (LLB, Munich, HMI)

- Spin Echo Scattering Angle Measurement (SESAME)
  - Measure spatial correlations over large distances
  - Currently only available for SESANS at Delft
  - Several prototypes being developed in the U.S. for SESANS and SERGIS

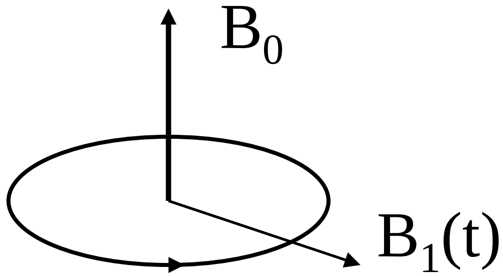
- MIEZE
  - Energy resolved SANS
  - Not yet implemented anywhere (as far as I know) although prototype was built at IPNS

An NRSE Triple Axis Spectrometer at HMI: Note the Tilted Coils



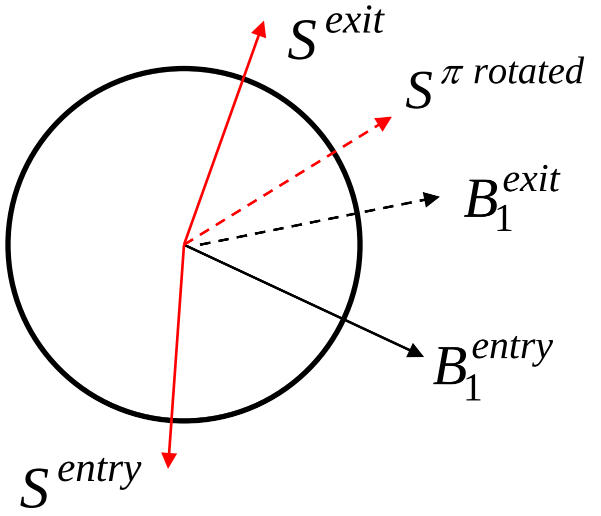
The Principle of Neutron Resonant Spin Echo

- Within a coil, the neutron is subjected to a steady, strong field, B_0, and a weak rf field $B_1\cos(\omega t)$ with a frequency $\omega = \omega_0 = \gamma B_0$
  - Typically, $B_0 \sim 100$ G and $B_1 \sim 1$ G



- In a frame rotating with frequency ω_0, the neutron spin sees a constant field of magnitude B_1

- The length of the coil region is chosen so that the neutron spin precesses around B_1 thru an angle π.



- The neutron precession phase is:

$$\phi_{neutron}^{exit} = \phi_{RF}^{exit} + (\phi_{RF}^{entry} - \phi_{neutron}^{entry})$$
$$= 2\phi_{RF}^{entry} - \phi_{neutron}^{entry} + \omega_0 d / v$$

Neutron Spin Phases in an NRSE Spectrometer*

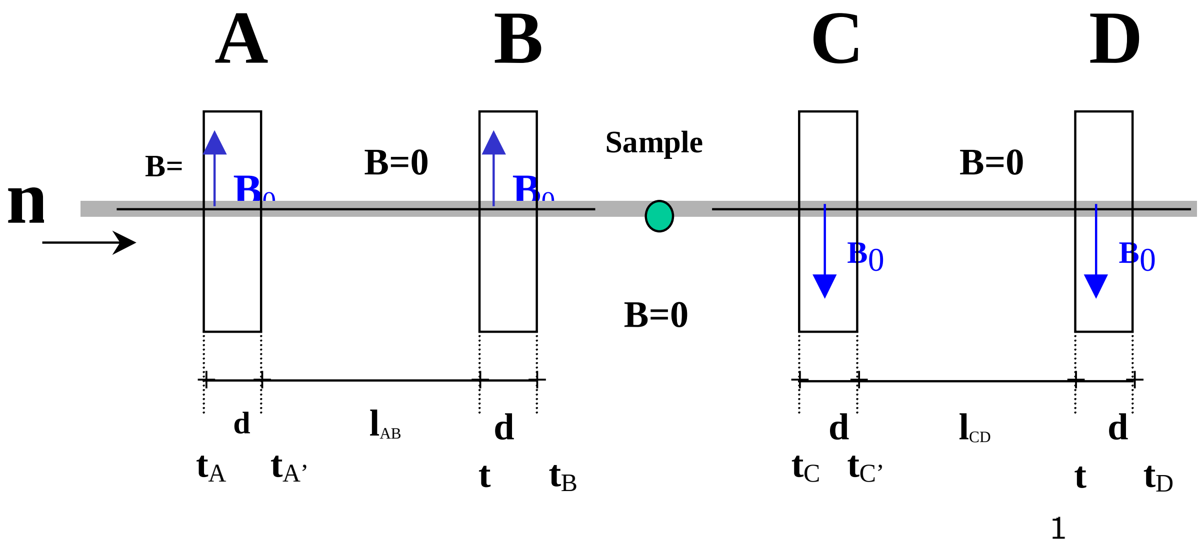


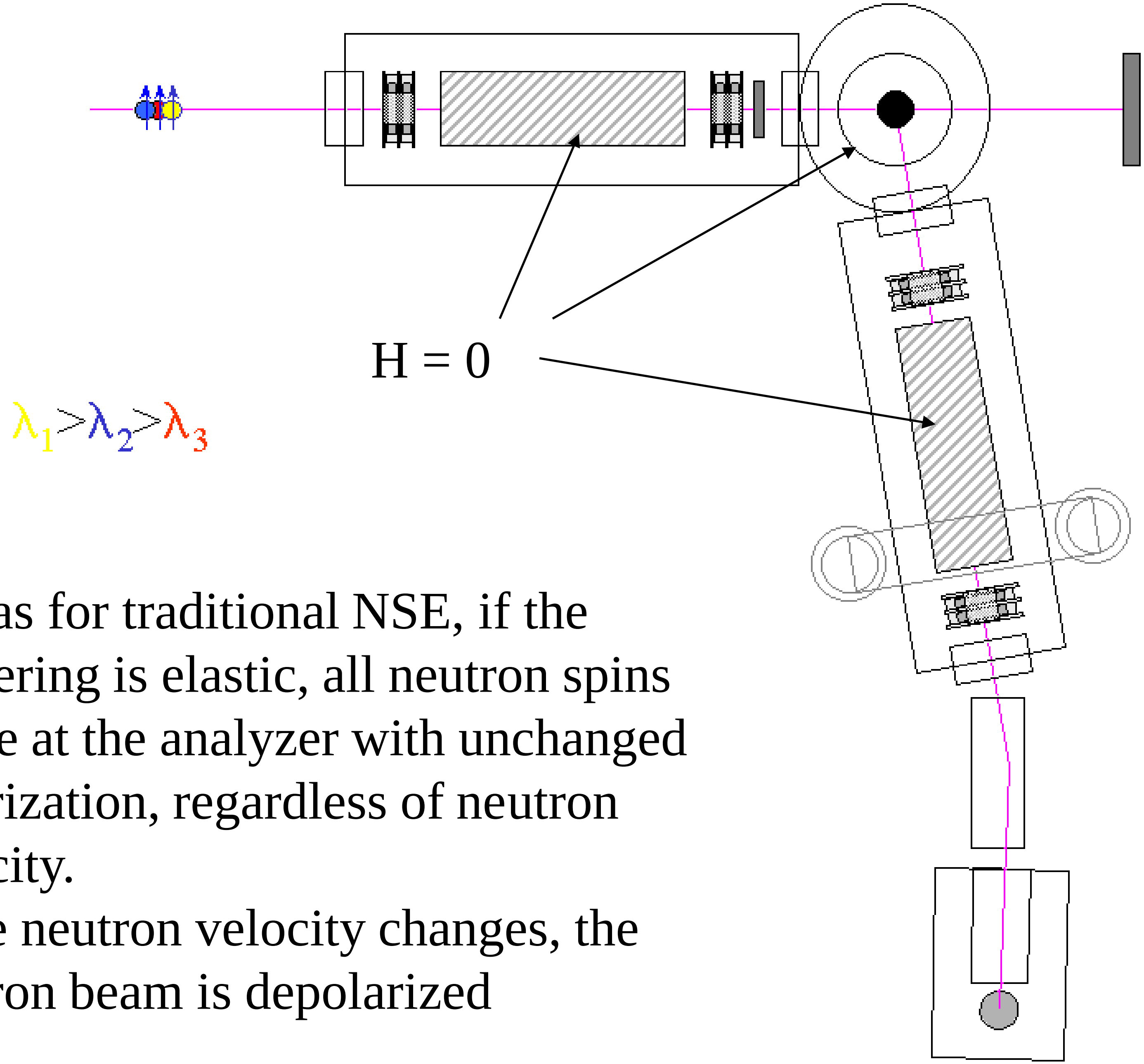
Table 1. Spin orientation

	Time t	Phase field B_r	neutron Spin phase S
A	t_A	ωt_A	0
A'	$t_{A'} = t_A + \frac{d}{v}$	$\omega t_{A'}$	$2\omega t_A + \omega \frac{d}{v}$
B	$t_B = t_A + \frac{l_{AB}+d}{v}$	ωt_B	$2\omega t_A + \omega \frac{d}{v}$
B'	$t_{B'} = t_A + \frac{l_{AB}+2d}{v}$	$\omega t_{B'}$	$2\omega \frac{l_{AB}+d}{v}$
C	t_C	$-\omega t_C$	$2\omega \frac{l_{AB}+d}{v}$
C'	$t_{C'} = t_C + \frac{d}{v}$	$-\omega t_{C'}$	$-\omega \frac{d}{v'} - 2\omega t_C - 2\omega \frac{l_{AB}+d}{v}$
D	$t_D = t_C + \frac{l_{CD}+d}{v'}$	$-\omega t_D$	$-\omega \frac{d}{v'} - 2\omega t_C - 2\omega \frac{l_{AB}+d}{v}$
D'	$t_{D'} = t_C + \frac{l_{CD}+2d}{v'}$	$-\omega t_{D'}$	$2\omega(\frac{l_{AB}+d}{v} - \frac{l_{CD}+d}{v'})$

Echo occurs for elastic scattering when

$l_{AB} + d = l_{CD} + d$

* Courtesy of S. Longeville



Just as for traditional NSE, if the scattering is elastic, all neutron spins arrive at the analyzer with unchanged polarization, regardless of neutron velocity.

If the neutron velocity changes, the neutron beam is depolarized

The Measured Polarization for NRSE is given by an Expression Similar to that for Classical NSE

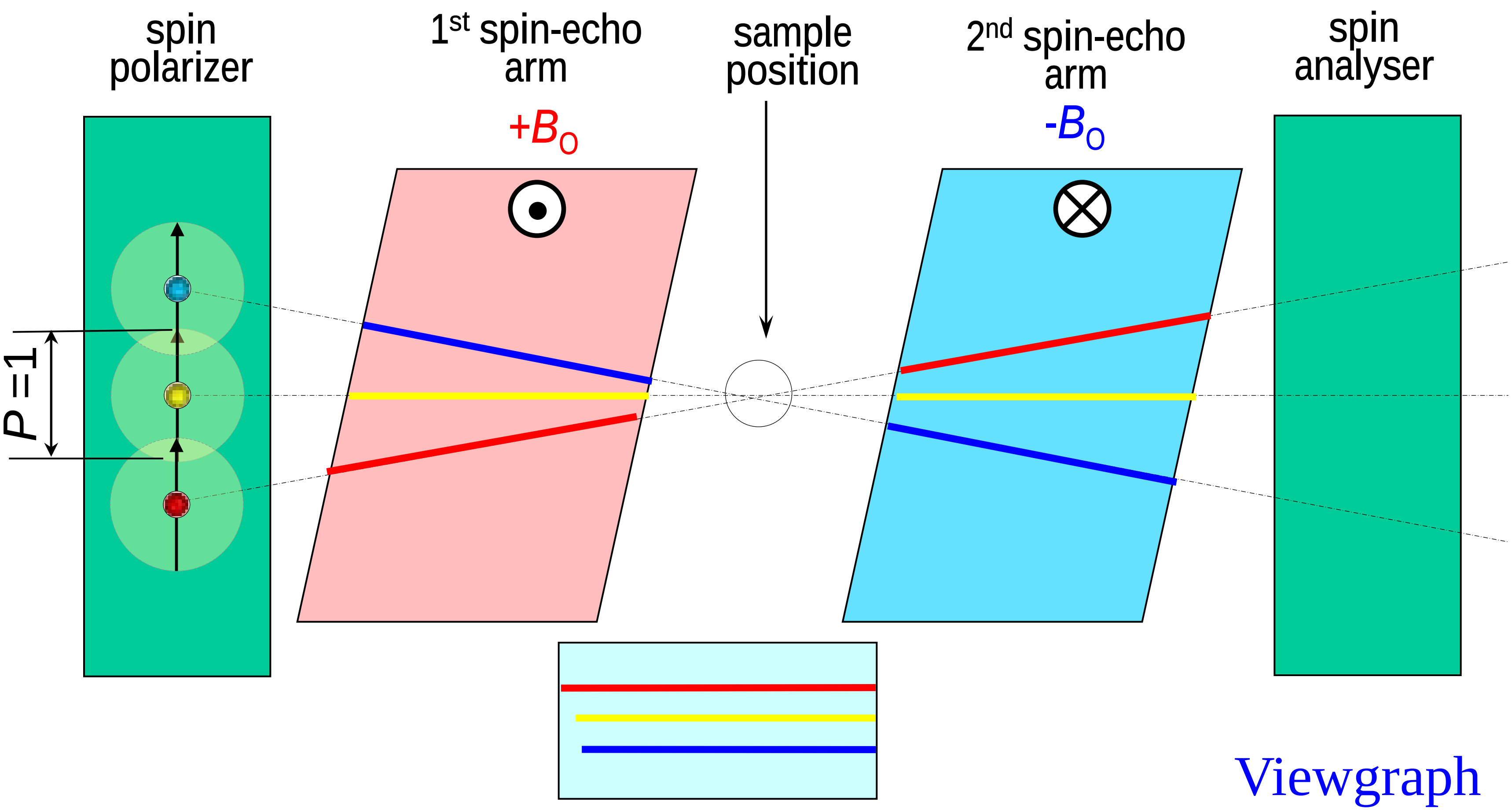
- Assume that $v' = v + \delta v$ with δv small and expand to lowest order, giving:

$$\langle P \rangle = \frac{\iint I(\lambda) S(\vec{Q},\omega) \cos(\omega \tau_{NRSE}) d\lambda d\omega}{\iint I(\lambda) S(\vec{Q},\omega) d\lambda d\omega}$$

where the "spin echo time" $\tau_{NRSE} = 2\gamma B_0 (l + d) \frac{m^2}{2\pi h^2} \lambda^3$

- Note the additional factor of 2 in the echo time compared with classical NSE (a factor of 4 is obtained with “bootstrap” rf coils)
- The echo is obtained by varying the distance, l, between rf coils
- In NRSE, we measure neutron velocity using fixed “clocks” (the rf coils) whereas in NSE each neutron “carries its own clock” whose (Larmor) rate is set by the local magnetic field

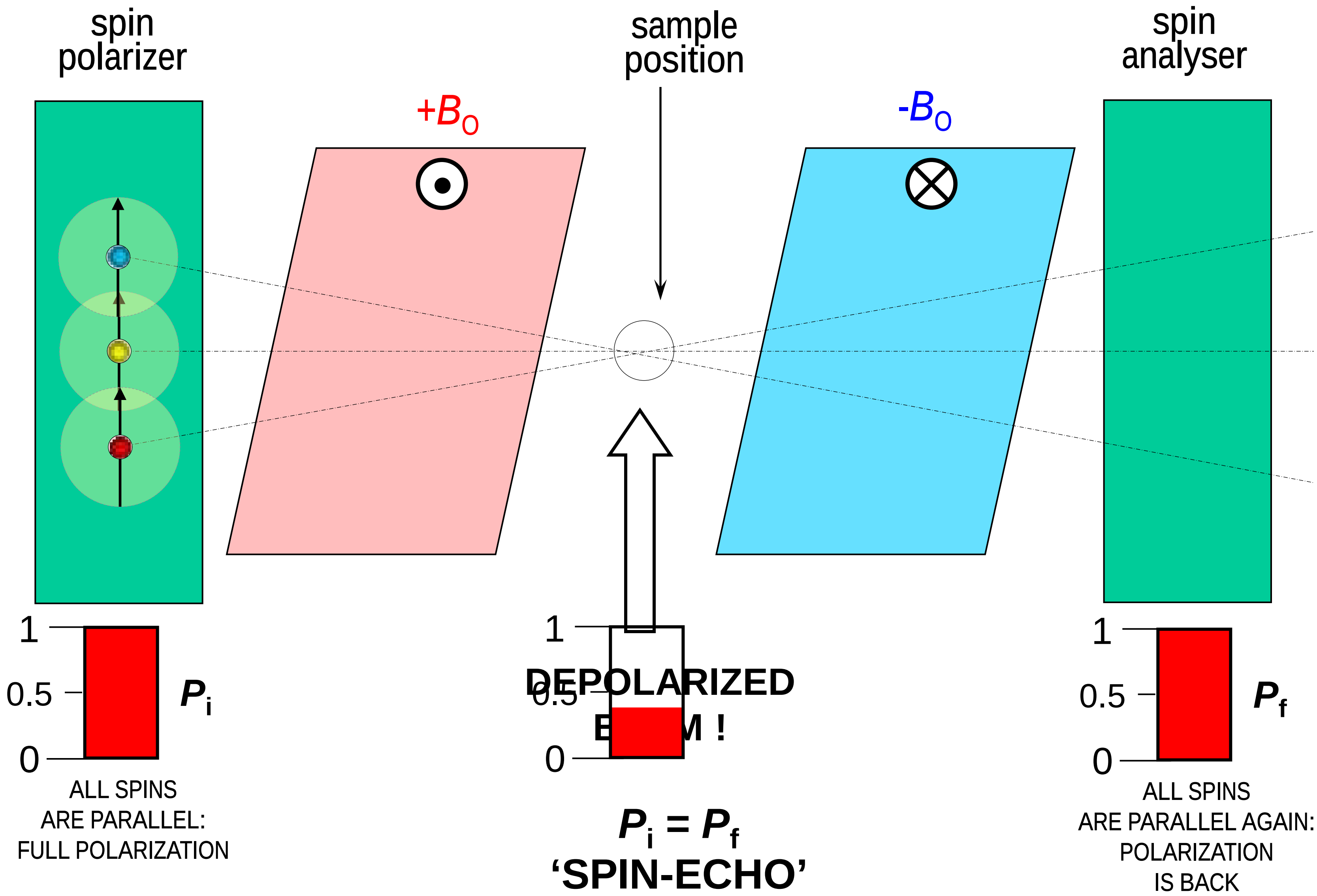
SESAME: Tilted Field Boundaries to Code Scattering Angles



DIFFERENT PATH LENGTHS FOR THE DIFFERENT TRAJECTORIES !

Viewgraph sequence by A. Vorobiev

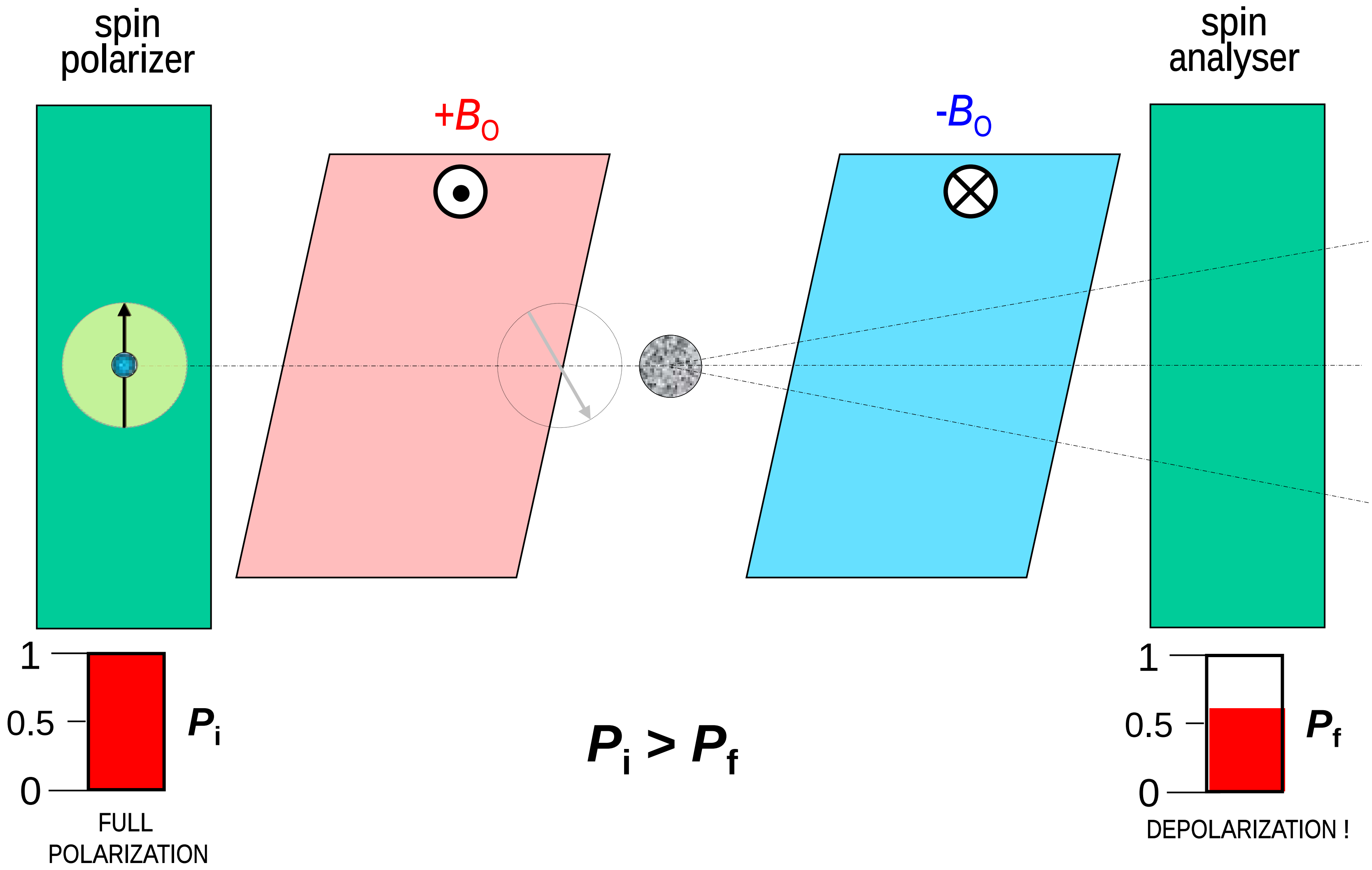
Spin Echo Scattering Angle Measurement (SESAME)
No Sample in Beam



$P_i = P_f$
'SPIN-ECHO'

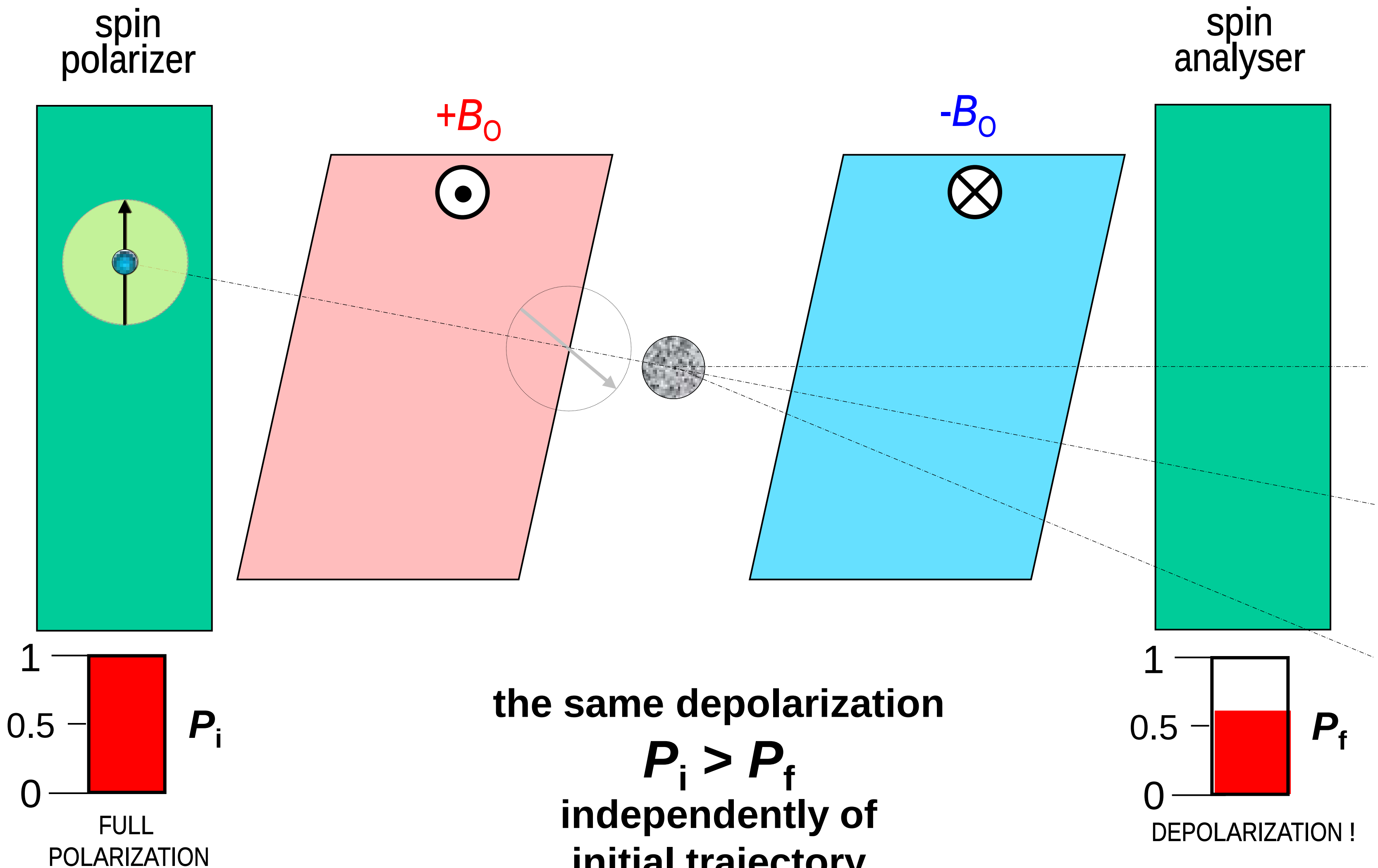
Spin Echo Scattering Angle Measurement (SESAME)

Scattering by the Sample



Spin Echo Scattering Angle Measurement (SESAME)

Scattering of a Divergent Beam



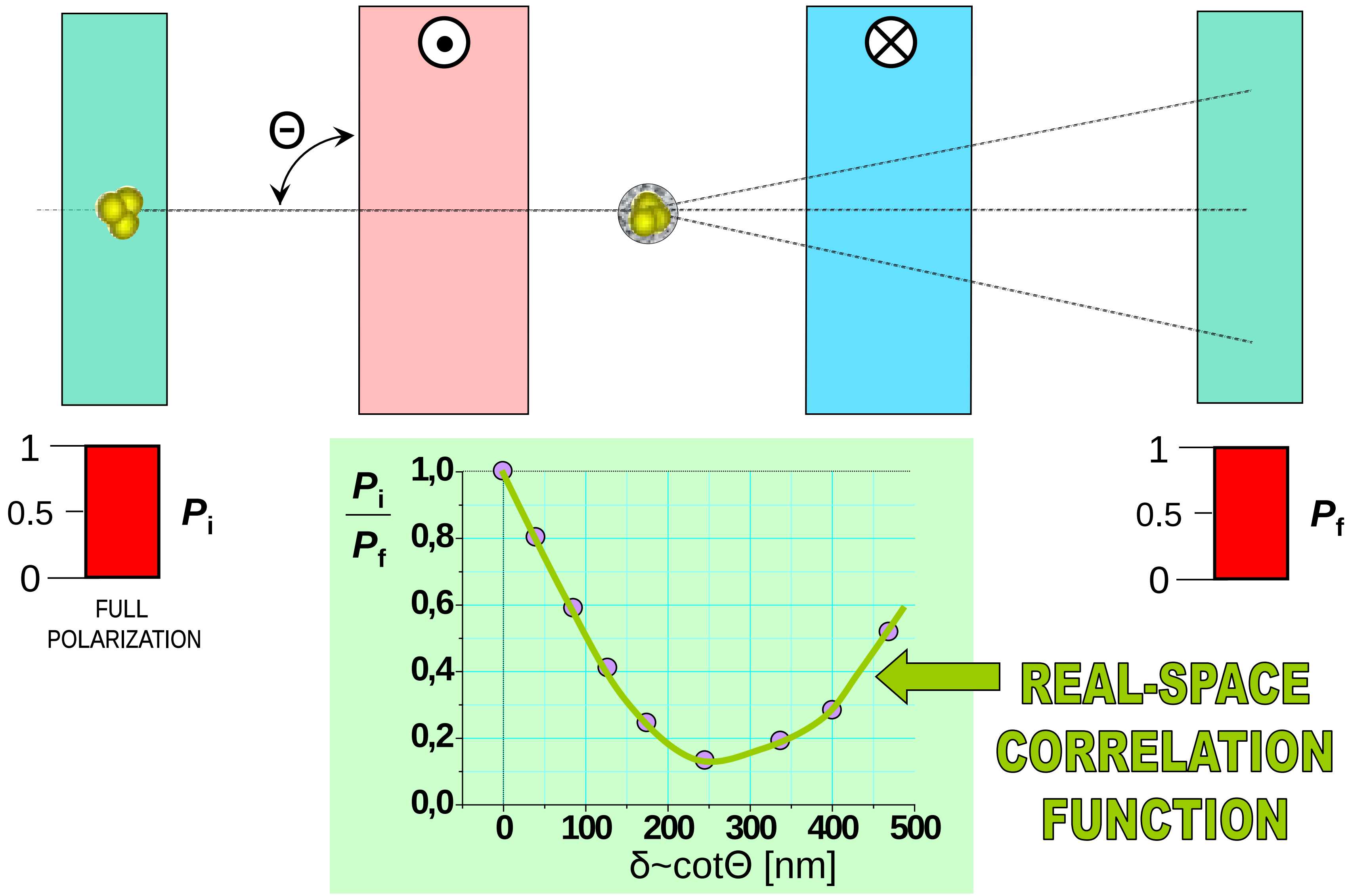
the same depolarization

$$P_i > P_f$$

independently of initial trajectory

spin-echo angular coding

5. THE EXPERIMENT

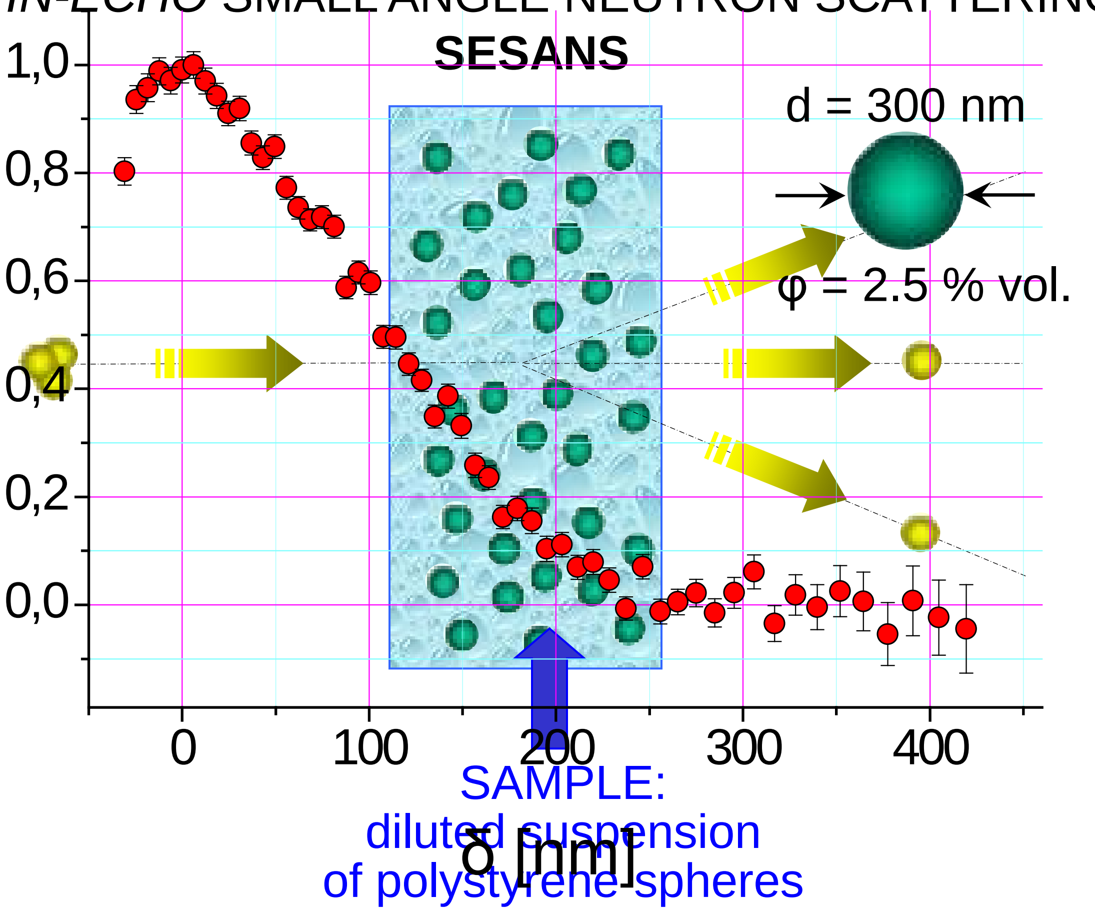


spin-echo angular coding

6. SESANS EXAMPLE-1

TRANSMISSION GEOMETRY:

SPIN-ECHO SMALL ANGLE NEUTRON SCATTERING



Autocorrelation function obtained for the polystyrene spheres (water suspension).