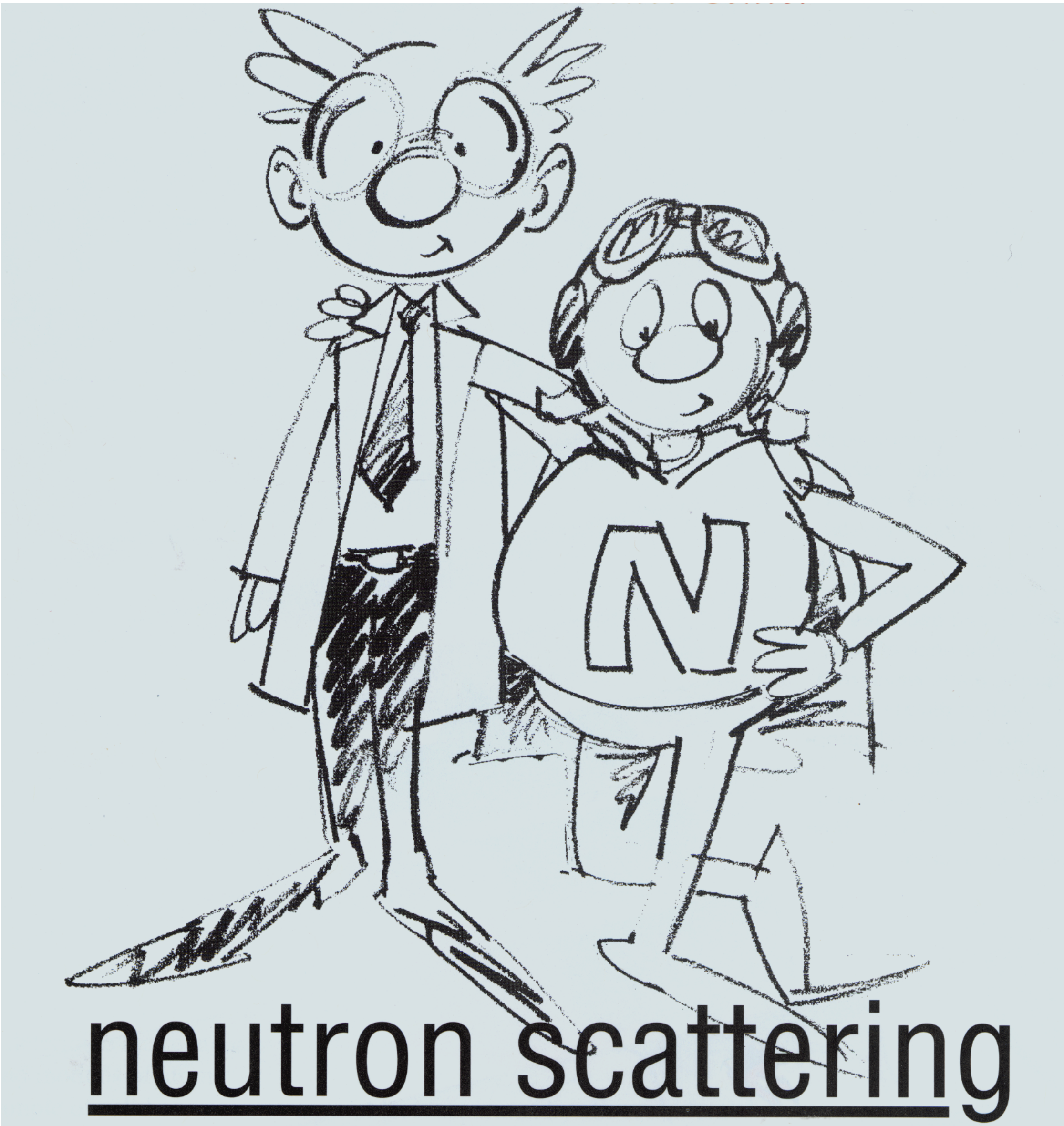


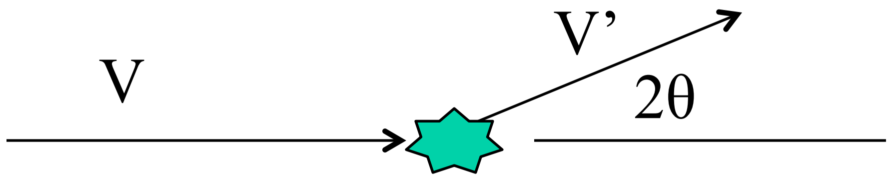
by

Roger Pynn

Larmor Methods for Neutron Scattering

Neutron Scattering

- What fraction of neutrons were scattered through an angle 2θ and with a change in velocity of ΔV?



$$\Delta V = V - V'$$

- Neutrons are **particles**
- If Suzie measures neutrons scattered through 36° from a sample when using 4 Angstrom neutrons, what is the inter-plane spacing of her crystal?

Iφ Σζιε μεασυρεσ νευτρονσ σχαττερεδ τηρουγη 36° φρομ α σαμπλε ωηεν υσινγ 4 Ανγστρομ νευτρονσ, ωηατ ισ τηε ιντερ− πλανε σπαχινγ οφ ηερ χρψσταλ?

Plug and chug

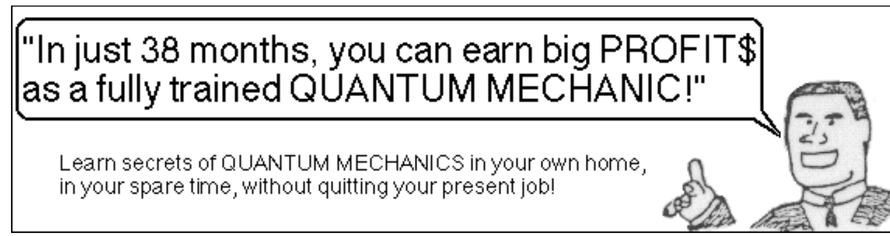
- Relate neutron scattering intensity distribution to sample properties -- Neutrons are **waves**

$$\lambda=2d\sin\theta$$

- But I slipped something past you……..

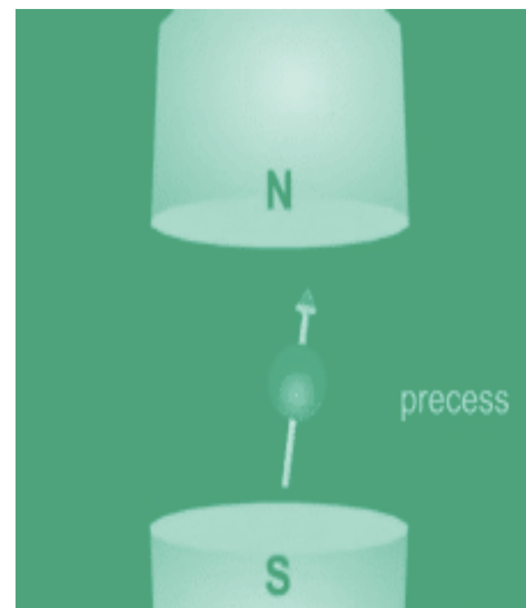
Larmor labeling

- Neutrons are **quantum particles** with a spin that has two possible states – “up” and “down”. It’s state can be written as a linear combination of “up” and “down” which evolve separately in a magnetic field



- Neutrons carry a pointed stick called the “**classical spin**” that rotates when the neutron is in a magnetic field
- The rotation is called Larmor precession and occurs at a rate of $1.83 \times 10^8 \times B(T)$ per second
- This precession can be used as a clock to measure the neutron speed

Plug & chug



Play Movies

Larmor Methods use the Neutron Spin to improve neutron instrument performance

- An example is Neutron Spin Echo (NSE)

- Traditional methods
  - define *both* incident & scattered wavevectors in order to define E and **Q** accurately
  - 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)
  - use the neutron's spin polarization to encode the difference between components of $\mathbf{k}_i$ and $\mathbf{k}_f$
  - can use large beam divergence &/or poor monochromatization to increase signal intensity, while maintaining very good resolution

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 to reverse rotation
  - The effect is 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 flip
- 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

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. δv) & *not* on v_1 & v_2 separately
- The measured polarization, <P>, is the average of cos(ϕ_1 - ϕ_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

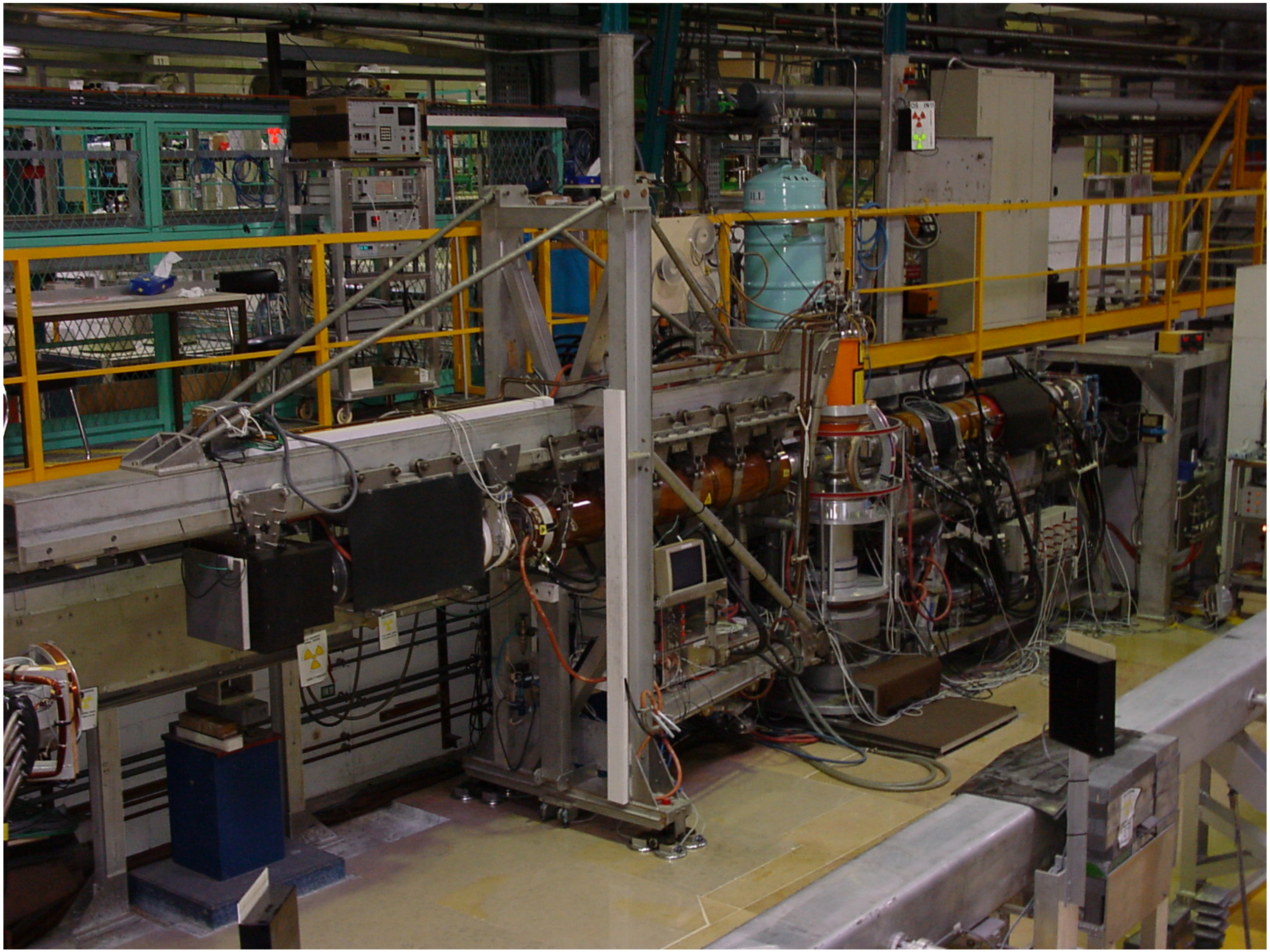
$$\langle P \rangle = \frac{\iint I(\lambda) S(\vec{Q},\omega) \cos(\phi_1 - \phi_2) d\lambda d\omega}{\iint I(\lambda) S(\vec{Q},\omega) d\lambda d\omega} \approx \left\langle \int S(\vec{Q},\omega) \cos(\omega\tau) d\omega \right\rangle = I(\vec{Q},\tau)$$

$$\text{where the "spin echo time" } \tau = \gamma B d \frac{m^2}{2\pi h^2} \lambda^3$$

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

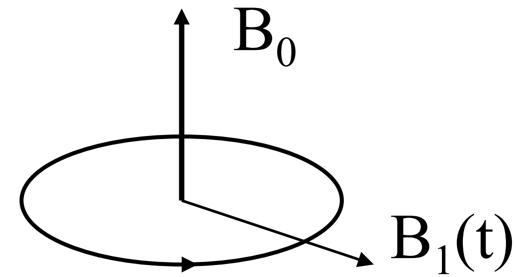
- $I(\vec{Q},t)$ is called the intermediate scattering function
  - Time Fourier transform of $S(\vec{Q},\omega)$ or the $\vec{Q}$ Fourier transform of $G(\vec{r},t)$, the two particle correlation function
- NSE probes the sample dynamics as a function of time rather than as a function of ω
- The spin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

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

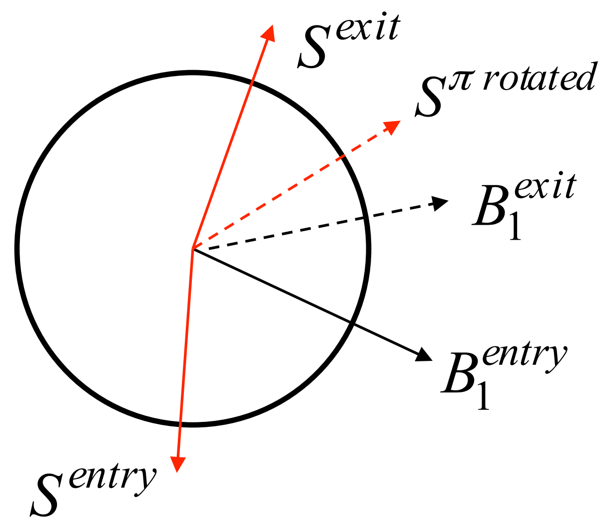


The Principle of Neutron Resonant Spin Echo

- 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:

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

Neutron Spin Phases in an NRSE Spectrometer*

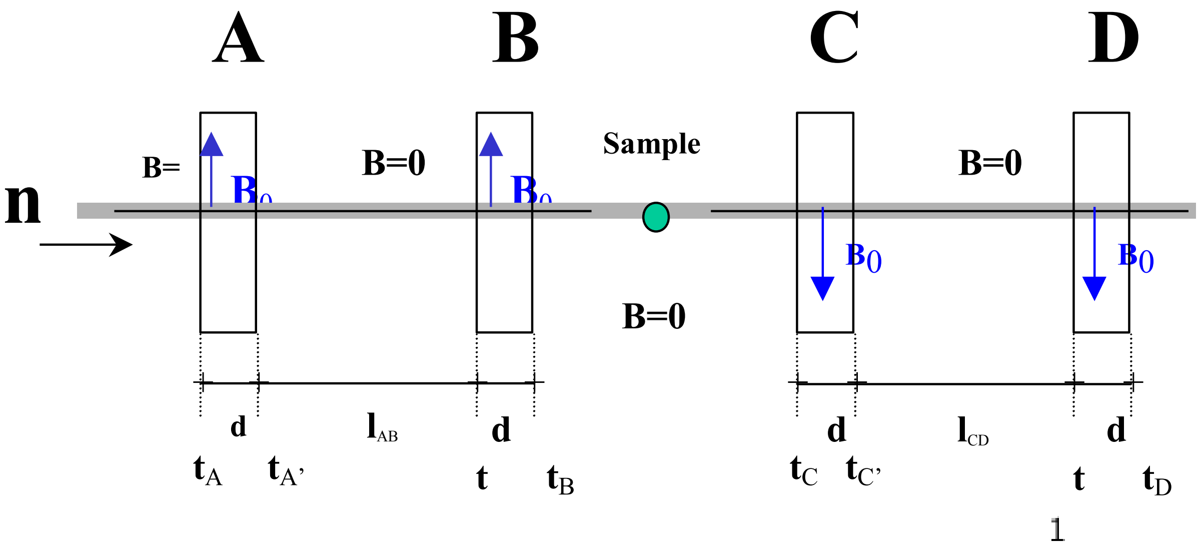


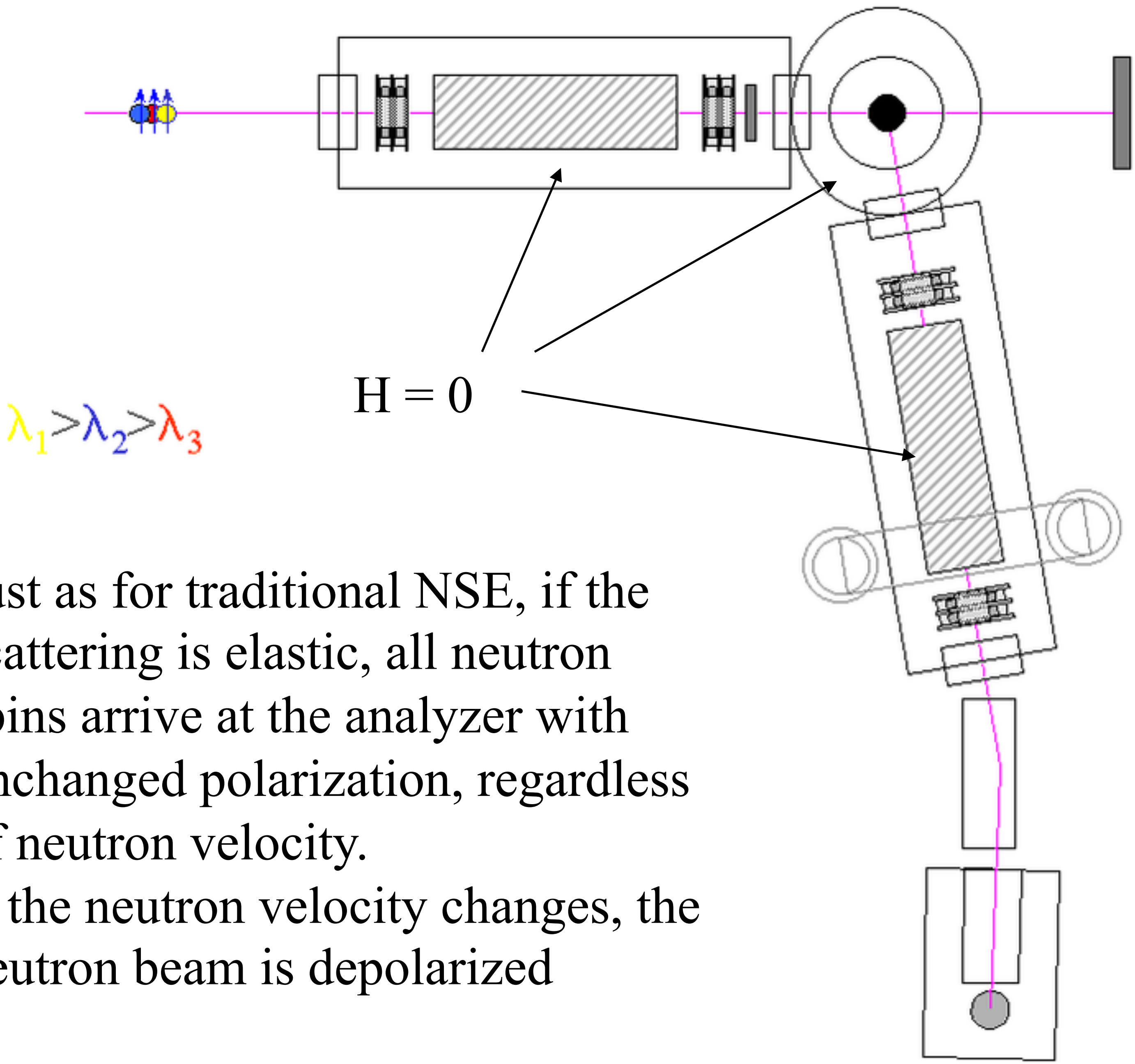
Table 1. Spin orientation

	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:

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

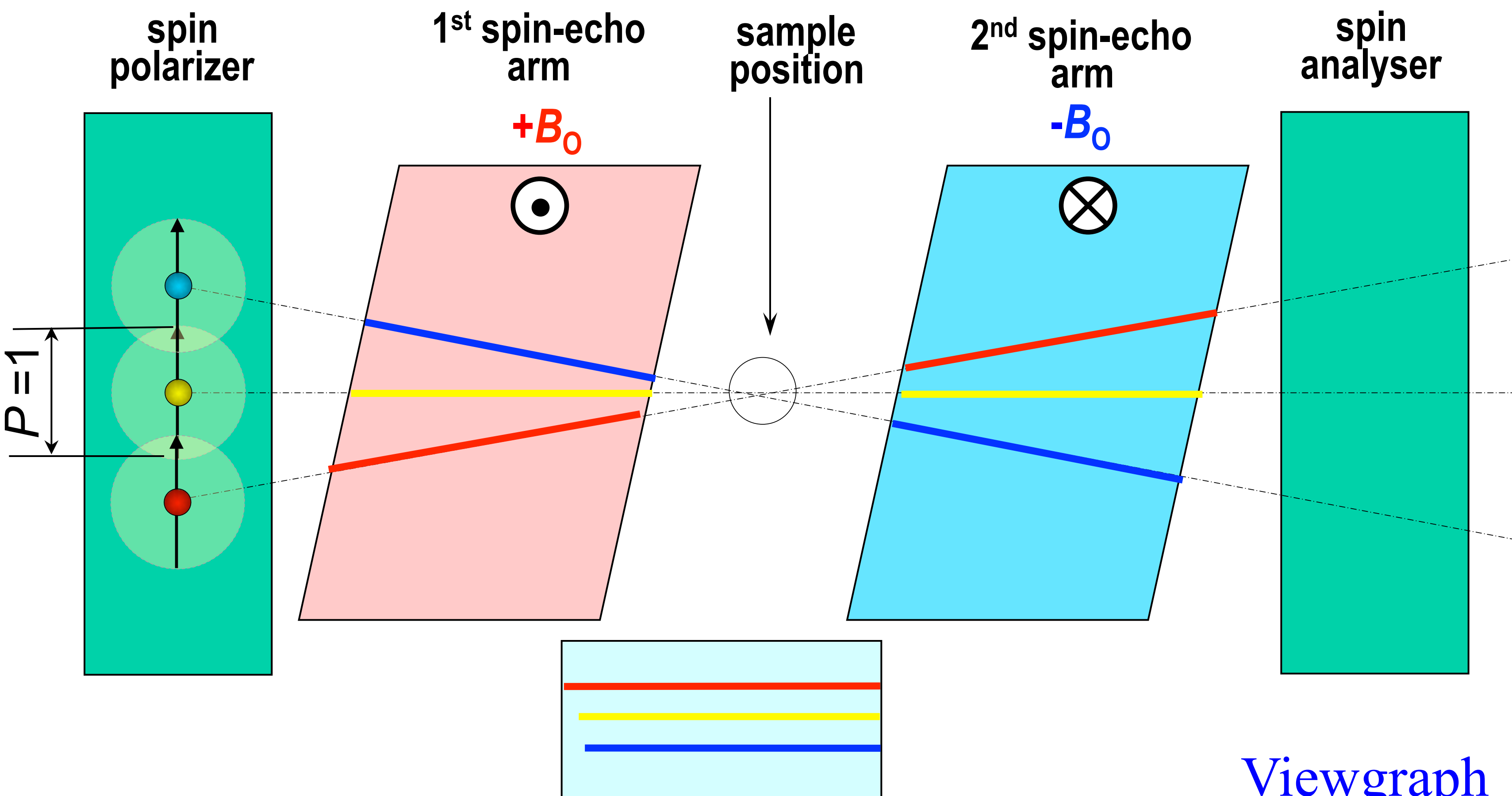
where the "spin echo time" $\tau_{NRSE} = 2\gamma B_0 (l + d) \frac{m^2}{2\pi h^2} \lambda^3$

- 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

The Classical Picture of Spin Precession

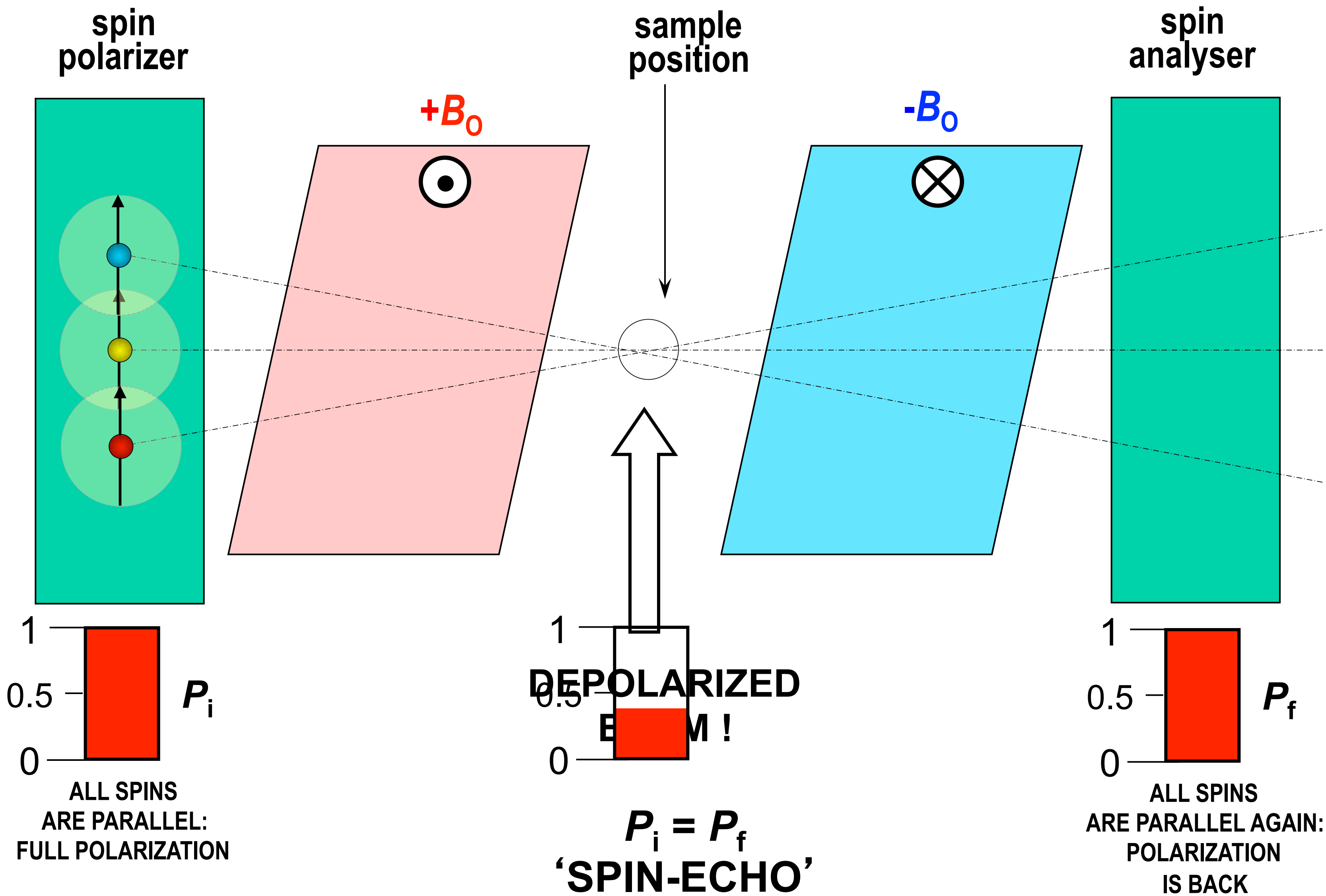


DIFFERENT PATH LENGTHS FOR THE DIFFERENT TRAJECTORIES !

Viewgraph sequence by A. Vorobiev

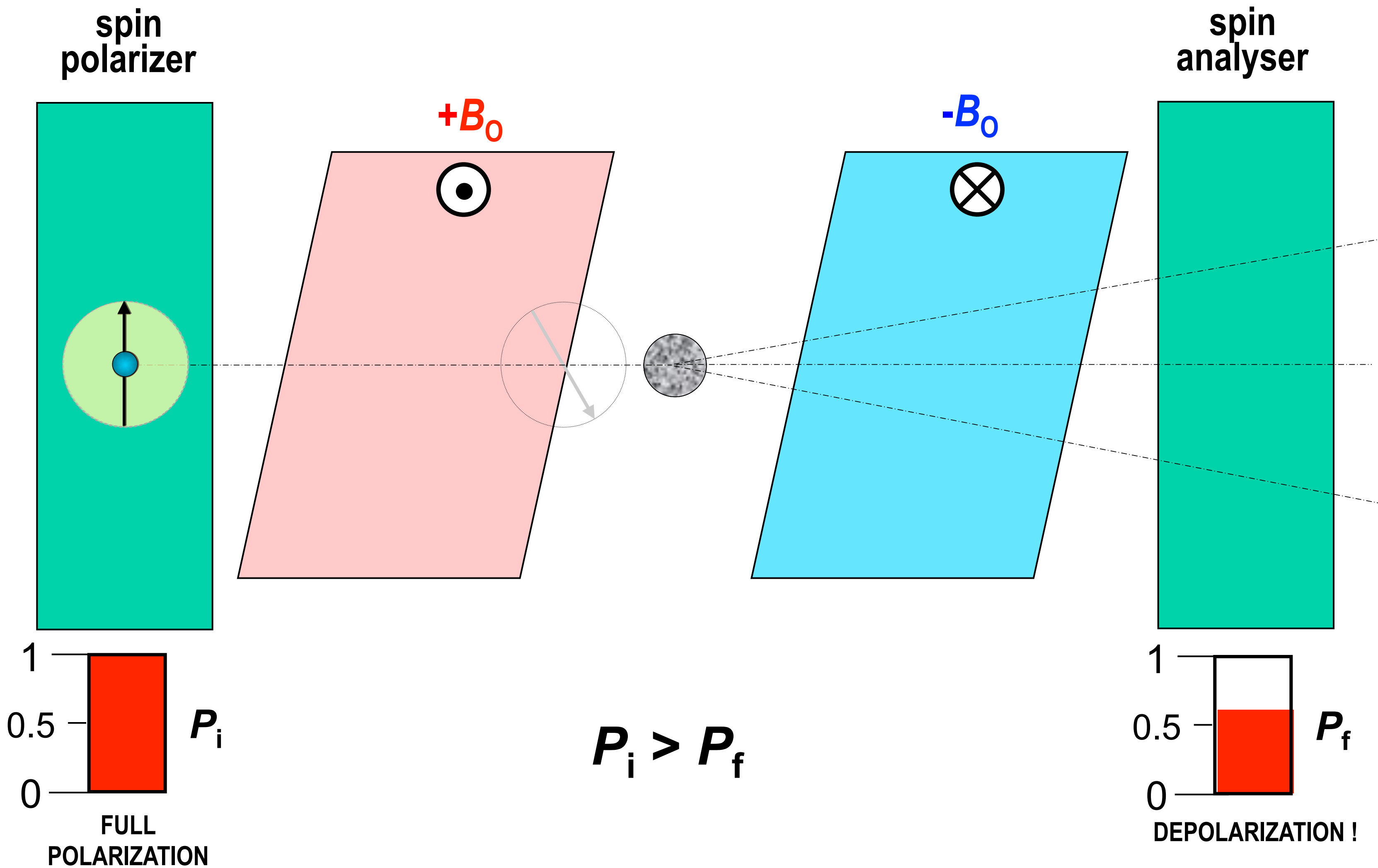
Spin Echo Scattering Angle Measurement (SESAME)

No Sample in Beam

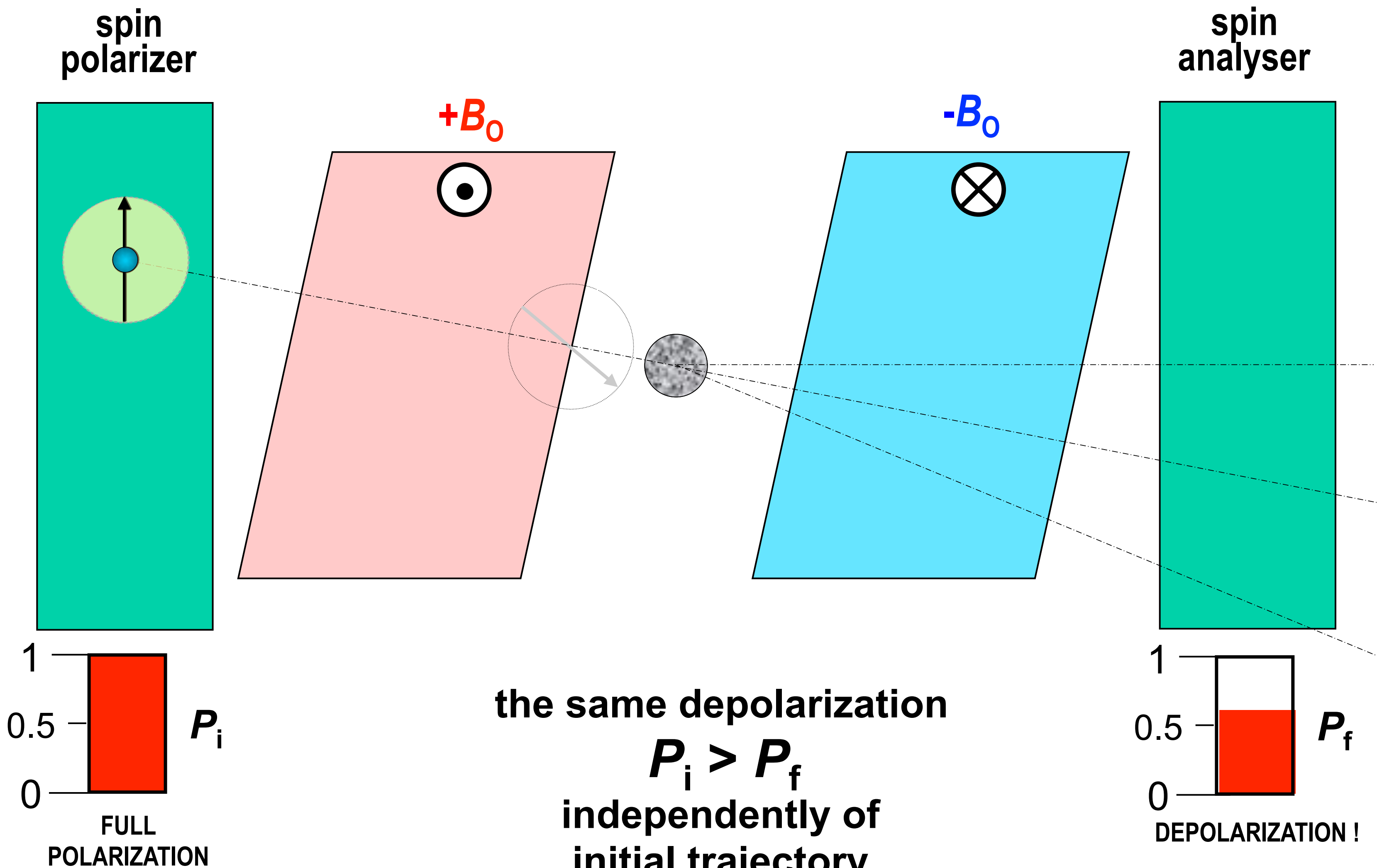


$P_i = P_f$
'SPIN-ECHO'

Spin Echo Scattering Angle Measurement (SESAME)
Scattering by the Sample



Spin Echo Scattering Angle Measurement (SESAME)
Scattering of a Divergent Beam

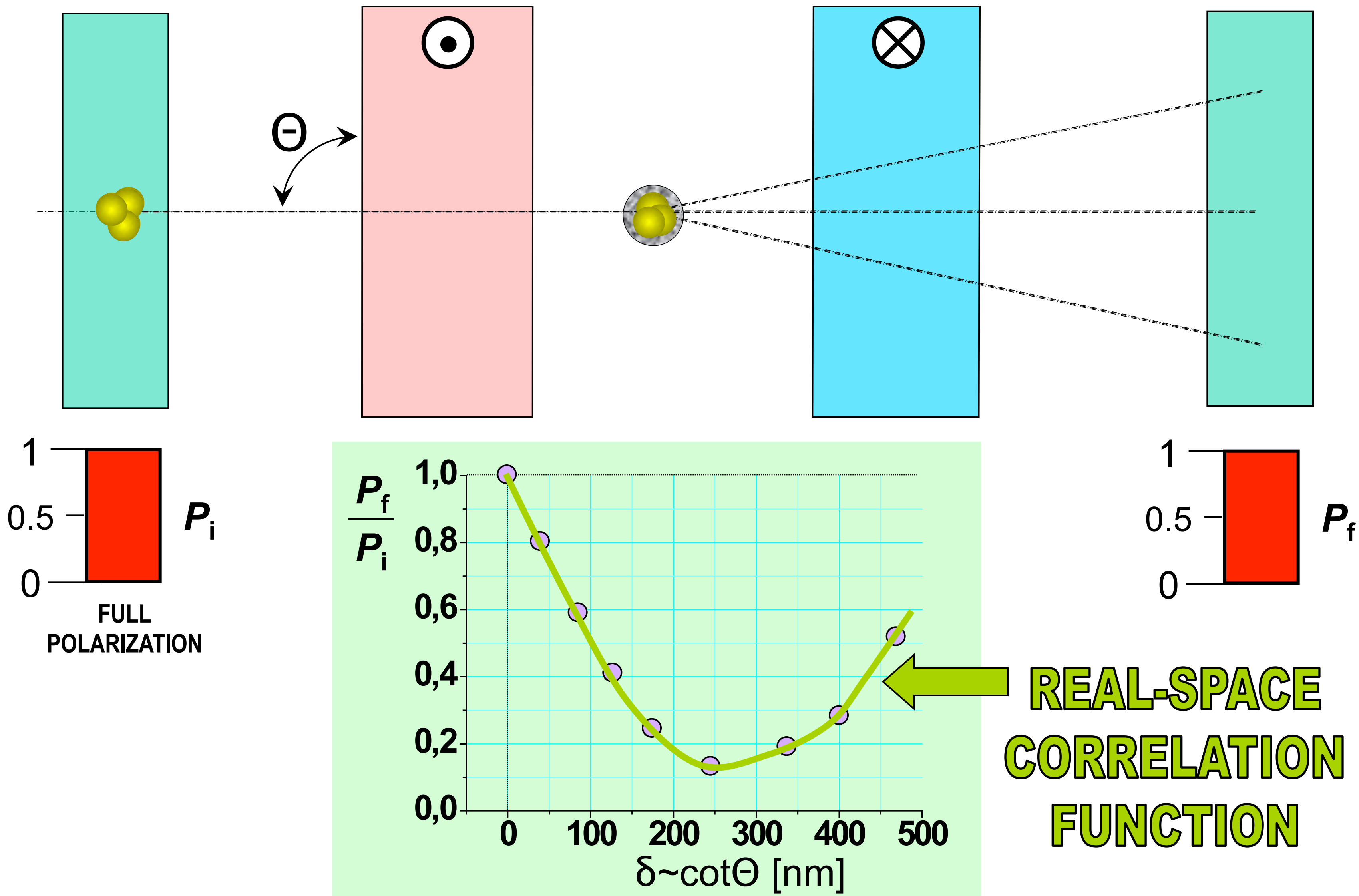


the same depolarization

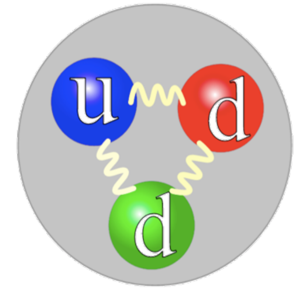
$$P_i > P_f$$

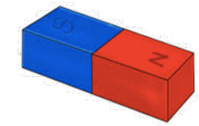
independently of initial trajectory

Spin-echo angular encoding: the experiment



The neutron is a spin-1/2 particle with a magnetic moment



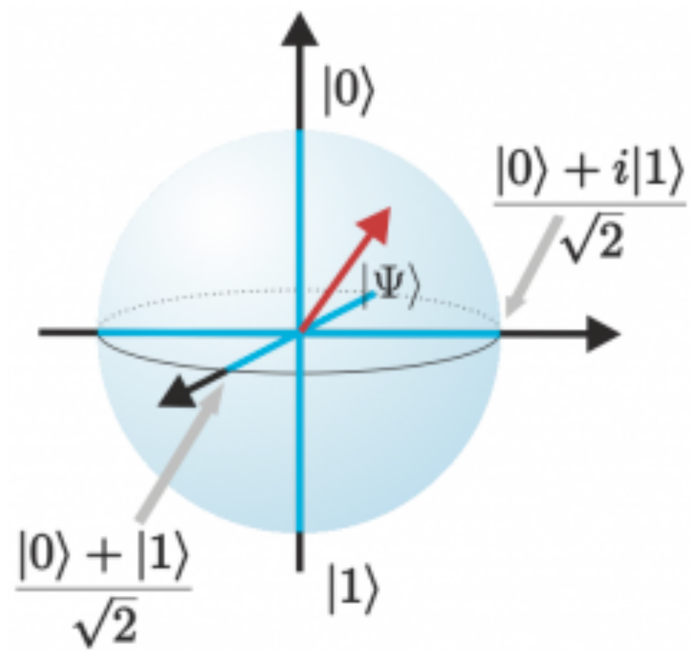


- In an applied magnetic field, B_z, the neutron has two spin eigenstates, “up” and “down” denoted l0> & l1>
  - Magnetic moment either along z or –z (magnetic moment is antiparallel to the spin)
- Quantum mechanics tells us that any neutron spin state is a linear combination of the two eigenstates

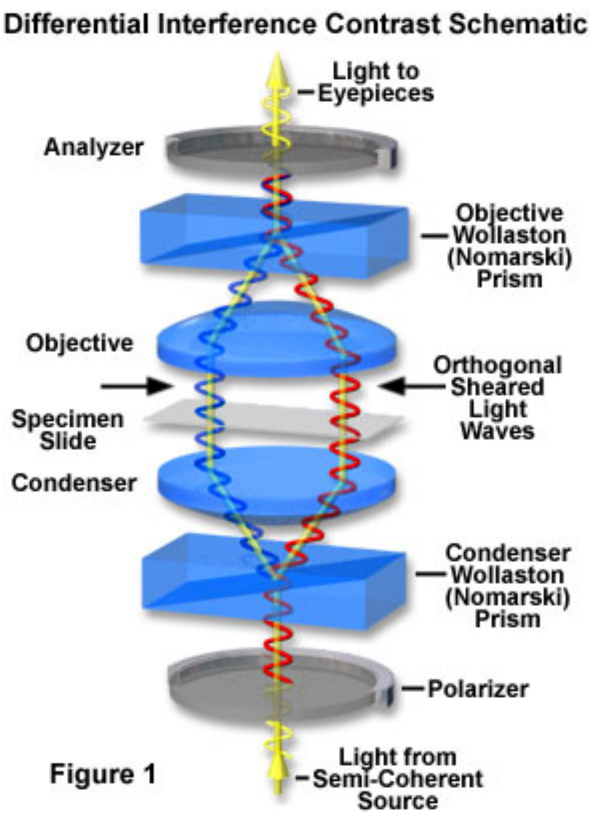
$$\chi = a|0\rangle + b|1\rangle \quad \text{where } |a|^2 + |b|^2 = 1\chi$$

$$\chi = \cos(\theta/2)|0\rangle + e^{i\phi}\sin(\theta/2)|1\rangle$$

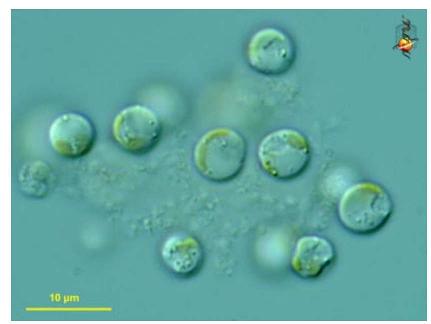
- The general state can be viewed as a point on the Bloch sphere



Differential Interference Contrast Microscopy



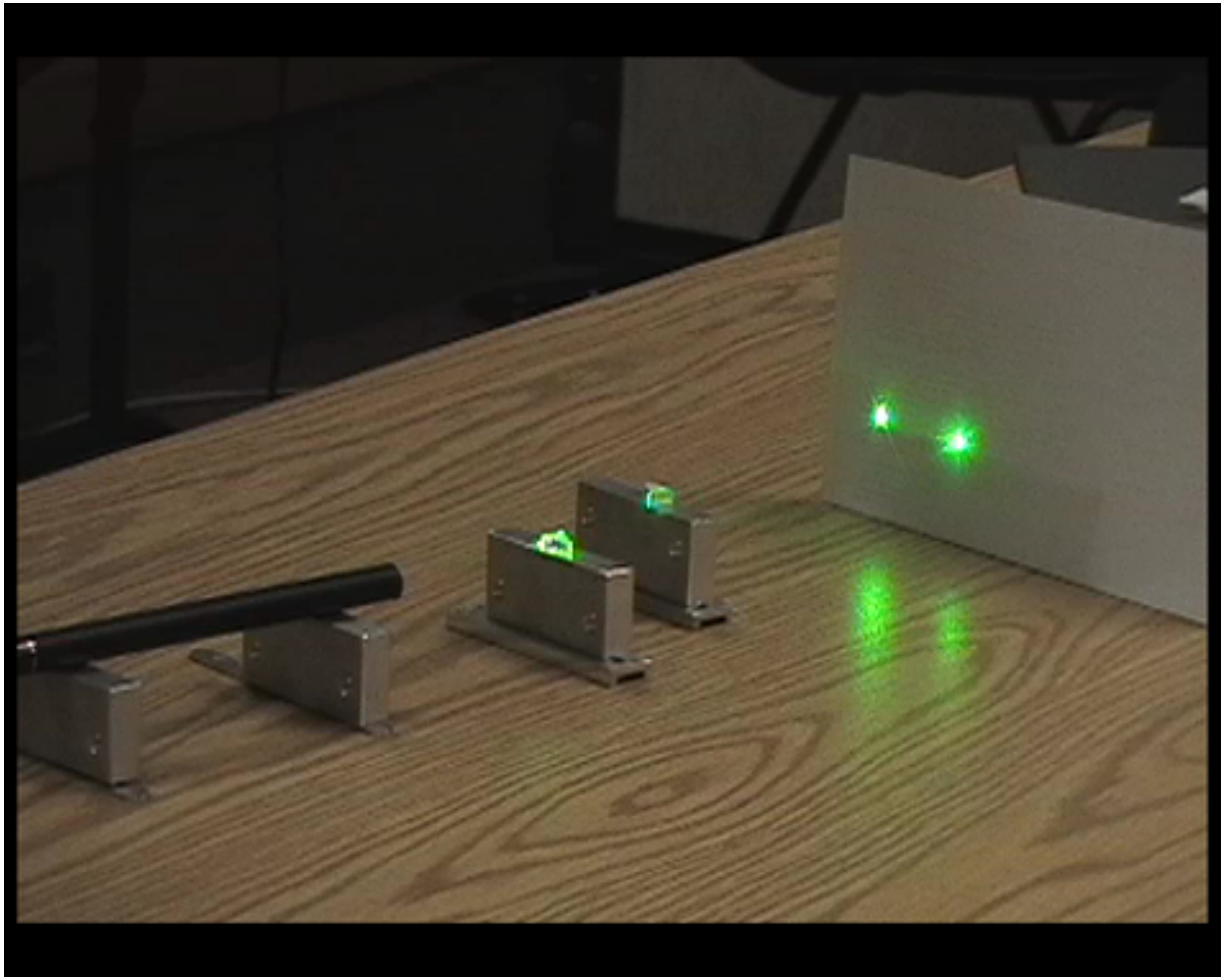
on Prism



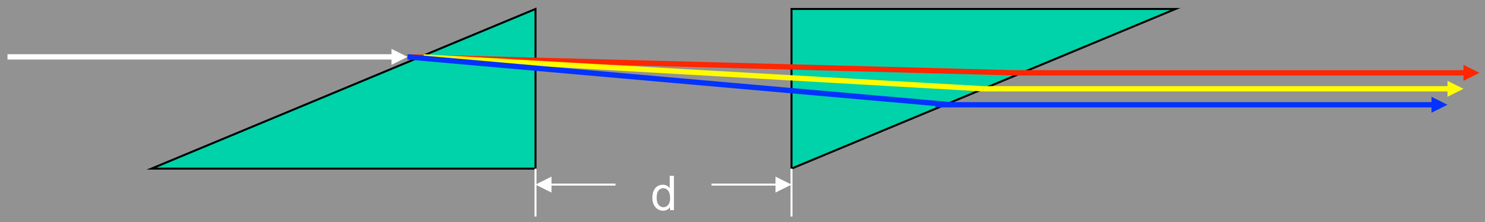
Eukaryotic Algae

Two polarization states of light "visit" neighboring parts of a sample and interfere to produce contrast that depends on the phase difference between the paths.

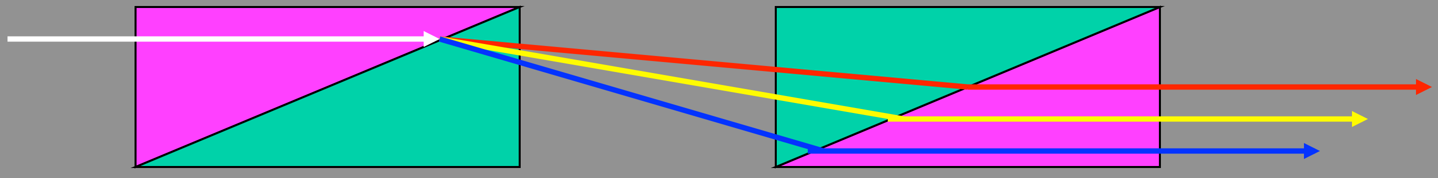
The Quantum View



Increasing the Spin Echo Length



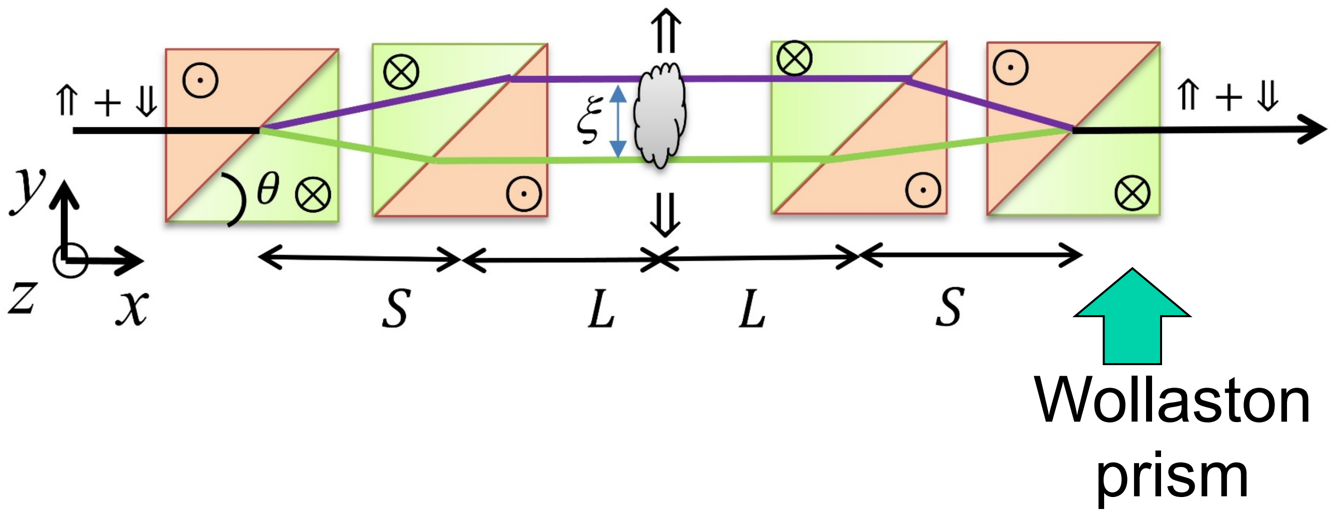
Cutting the slab into prisms allows the spatial separation of red and blue rays to be controlled by the separation of the prisms



Adding prisms with the opposite dispersion increases the separation of the red and blue rays

Spin echo small angle neutron scattering-
a polarized neutron interferometer

- Quantum view



$$\xi = c\lambda^2 BS\cot\theta$$

$$c = 1.47\times 10^{14}\, T^{-1} m^{-2}$$

ξ: spin echo length-max accessible length scale, which depends on,

- ❑ The separation of the two devices, S
- ❑ Inclination angle, θ
- ❑ Field intensity

The final neutron polarization is a measure of the degree of correlation between scattering from points separated by a distance ξ.

Conclusion: NSE Provides a Way to Separate Resolution from Monochromatization & Beam Divergence

- The method currently provides the best energy resolution for inelastic neutron scattering (~ neV)
  - Both classical NSE and NRSE achieve similar energy resolution
  - NRSE is more easily adapted to "phonon focusing" I.e. measuring the energy line-widths of phonon excitations

- The method can also be used to improve (Q) resolution for elastic scattering
  - Extend size range for SANS (SESANS)
  - May allow 100 – 1000 x gain in measurement speed for some SANS exps
  - Separate specular and diffuse scattering in reflectometry
  - Measure in-plane ordering in thin films (SERGIS)?

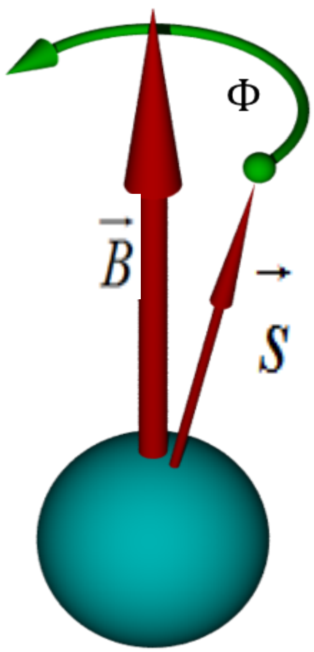
Summary of NSE

- Field boundaries perpendicular to the neutron beam code neutron's speed (not it's trajectory)
  - Correlation time (up to ~ 300 ns on IN15) scales as λ^3
- Field boundaries inclined to neutron beam code neutron's trajectory
  - Correlation length (also called the spin echo length) scales as λ^2
  - Up to 25 microns correlation length has been achieved (Delft)
- Neither method requires highly collimated or very monochromatic neutron beams
  - NSE is suitable for long correlation times or large length scales typical of soft materials such as complex fluid films

THE END

Questions?

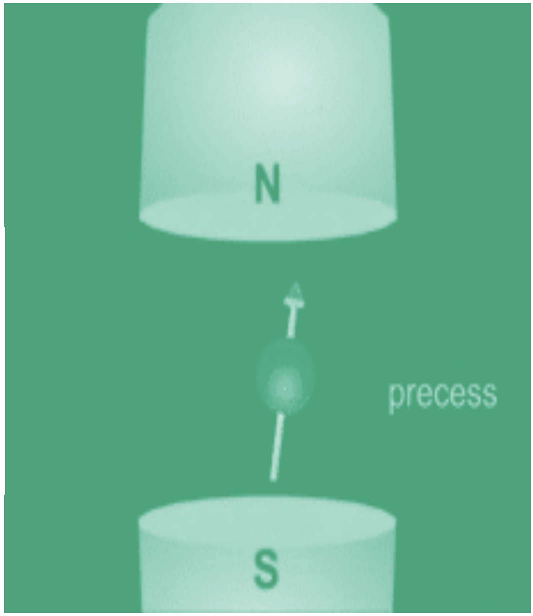
In a uniform magnetic field the neutron spin precesses around the the field



$$\frac{d\mathbf{S}}{dt} = \gamma_L \mathbf{S} \times \mathbf{B}$$

Bloch equation

$$\Phi = \frac{\gamma_L m \lambda}{h} \int_{path} Sign(B)|B|dl \propto \lambda \cdot FI$$

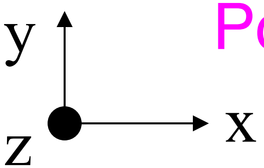


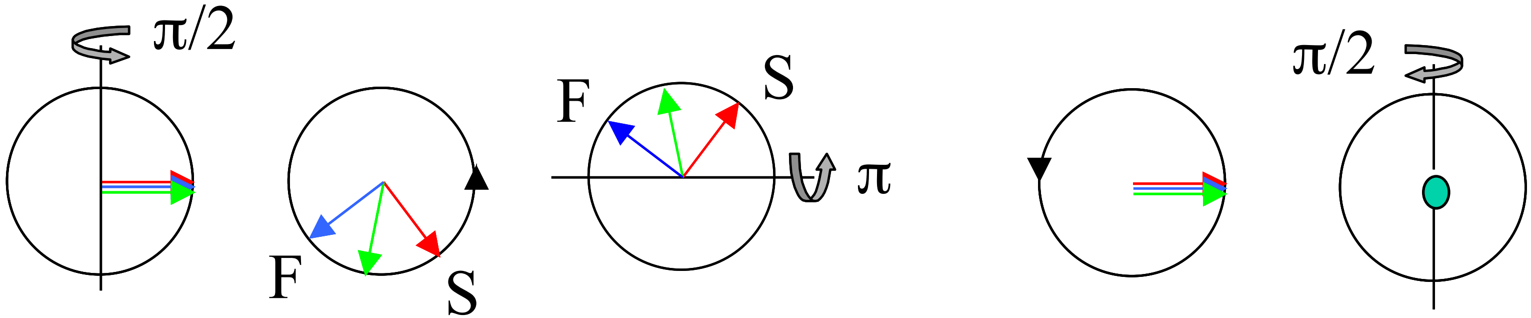
In a 10G field, 3cm of travel will rotate the spin of a 4Å neutron by a complete turn.

The “up” and “down” states have different Zeeman energies in a magnetic field

$$E_{Zeeman} = -\vec{\mu}_N . \vec{B}$$

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

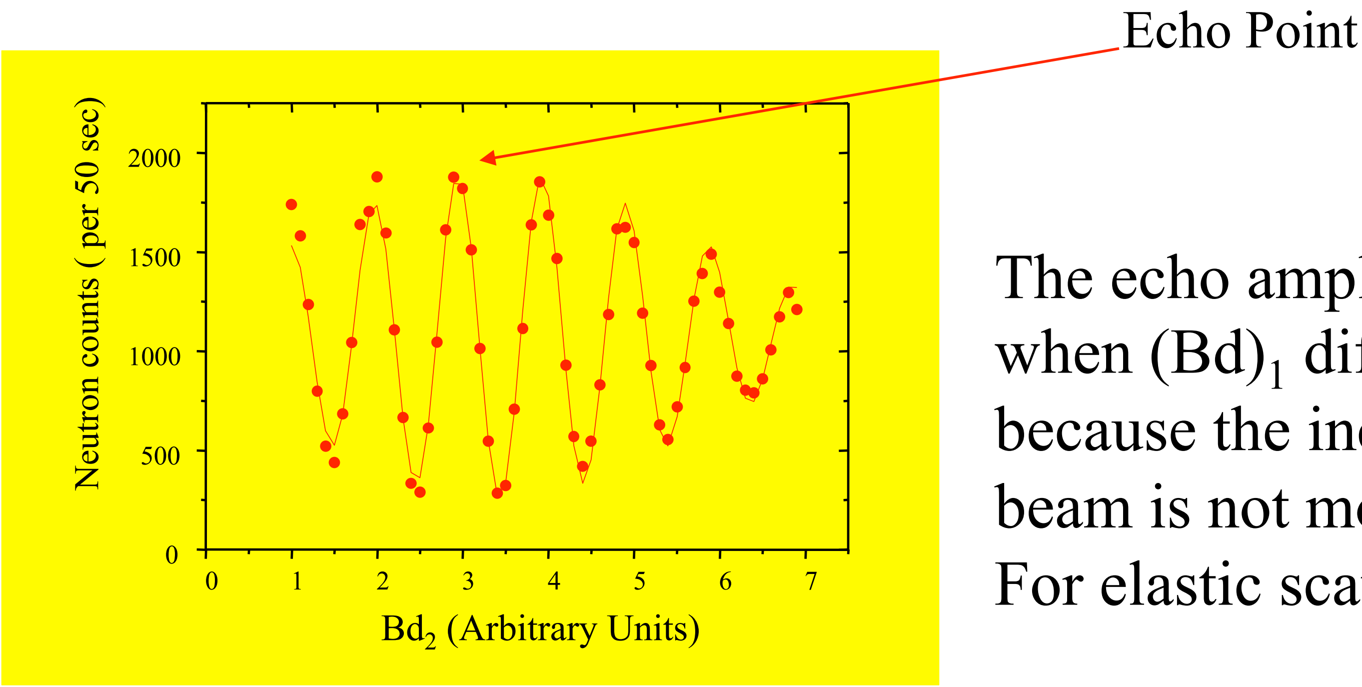
Allow spins to precess around z: all spins are in the same direction at the echo point if $\Delta E = 0$

Rotate spins to z and measure polarization

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

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

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



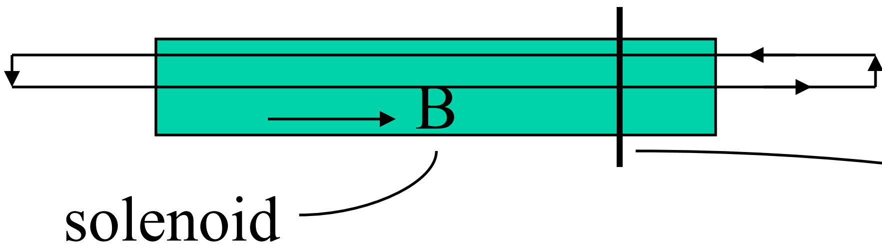
The echo amplitude decreases when $(Bd)_1$ differs from $(Bd)_2$ because the incident neutron beam is not monochromatic. For elastic scattering:

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

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

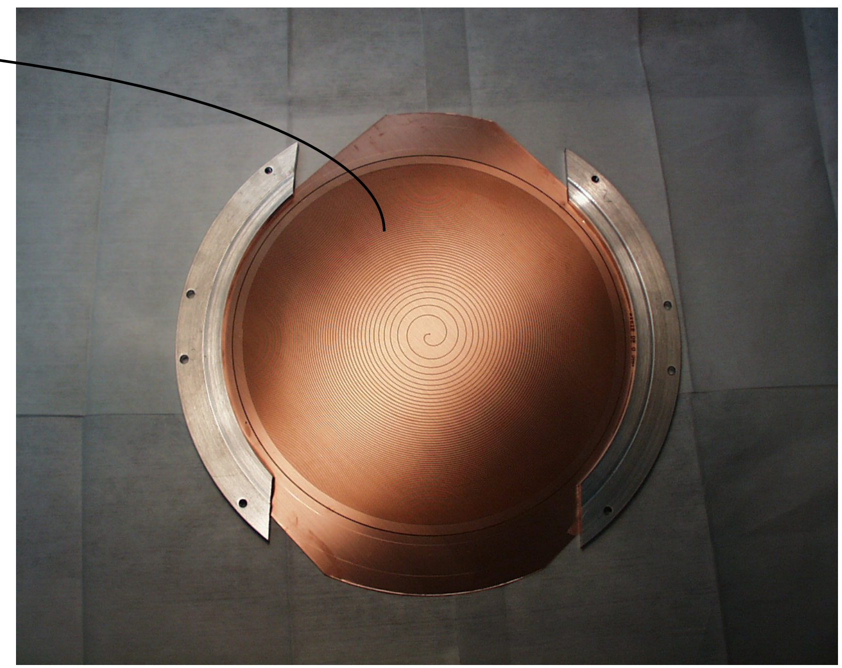
Field-Integral Inhomogeneities cause τ to vary over the Neutron Beam: They can be Corrected

- 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)



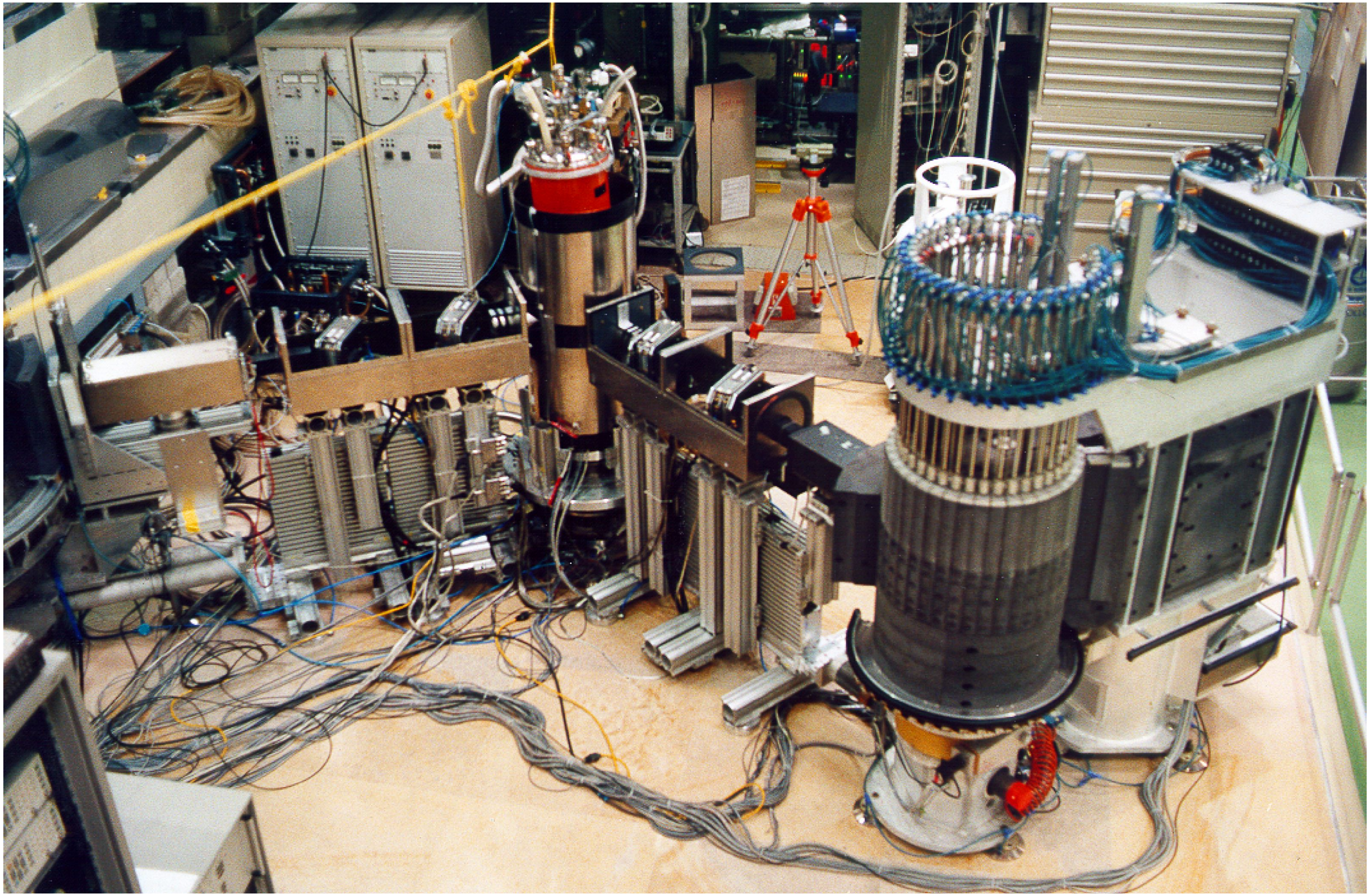
- According to Ampere's law, a current distribution that varies as r^2 can correct the field-integral inhomogeneities for parallel paths

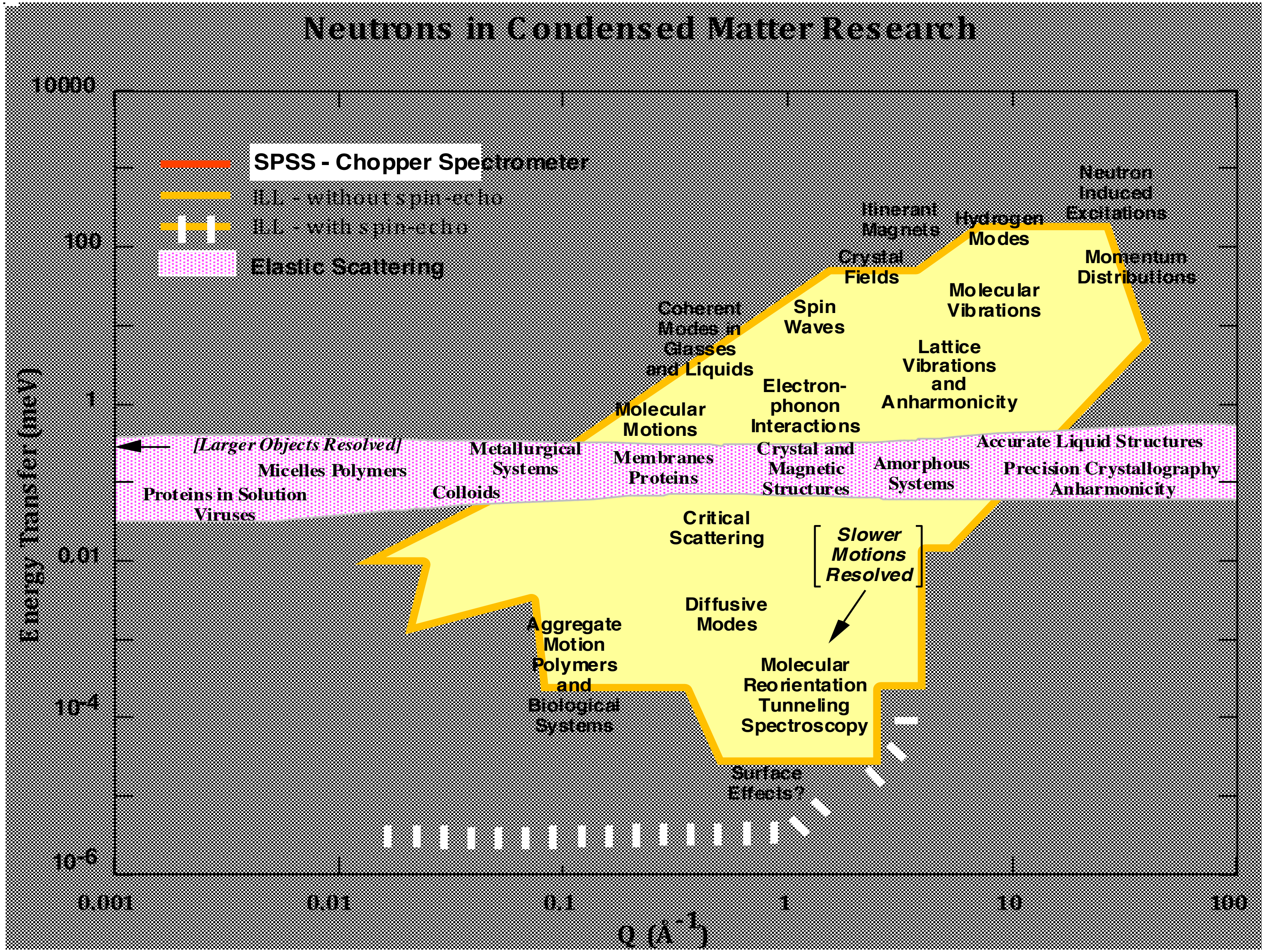
- Similar devices can be used to correct the integral along divergent paths



Fresnel correction coil for IN15

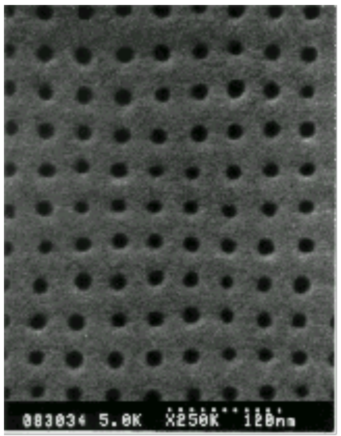
An NRSE Triple Axis Spectrometer at HMI: Note the Tilted Coils



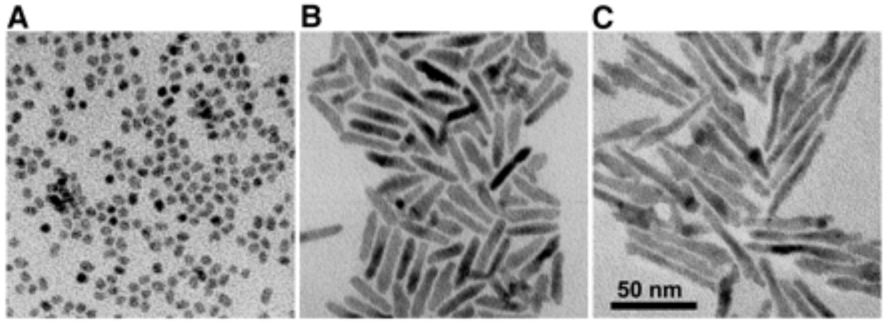


Neutron Spin Echo has significantly extended the (Q,E) range to which neutron scattering can be applied

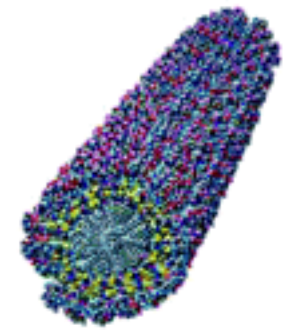
Nanoscience & Biology Need Structural Probes for 1-100 nm



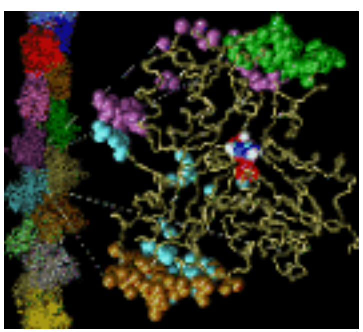
10 nm holes in PMMA



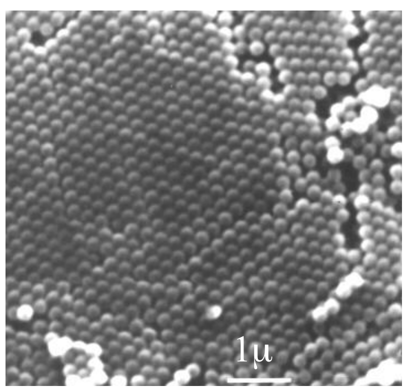
CdSe nanoparticles



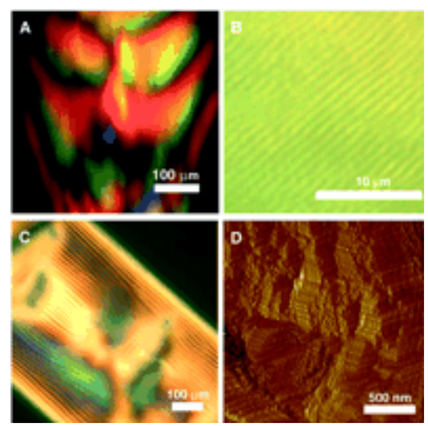
Peptide-amphiphile nanofiber



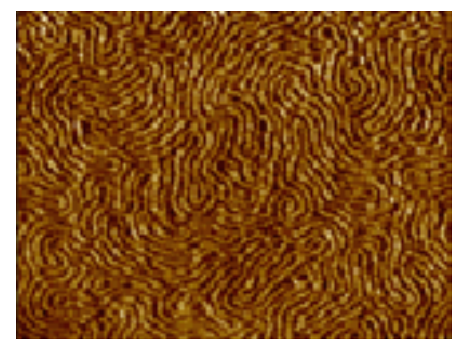
Actin



Si colloidal crystal

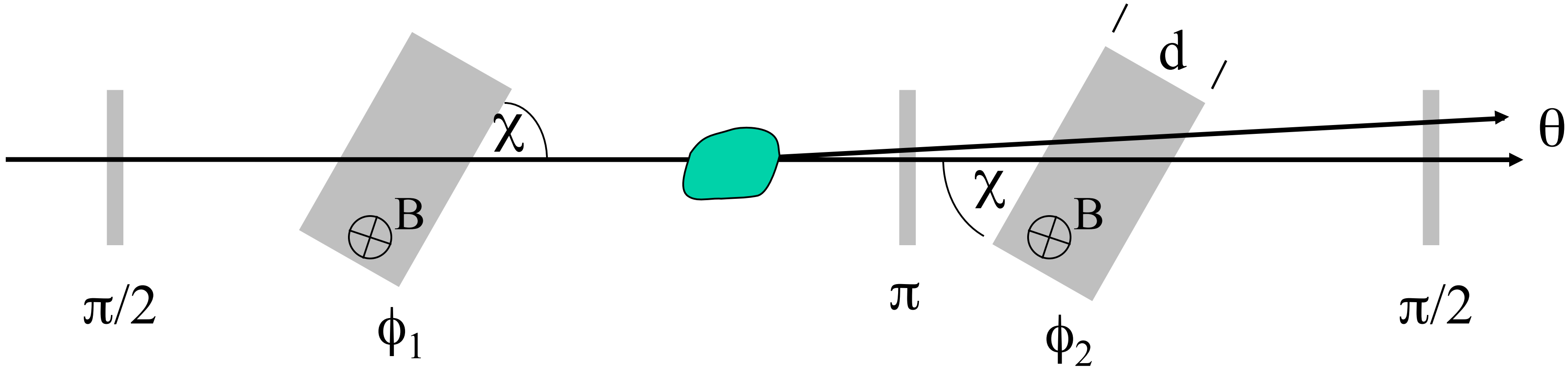


Structures over many length scales in self-assembly of ZnS and cloned viruses



Thin copolymer films

"Tilted" Fields for SESANS



- Any unscattered neutron (θ=0) experiences the same precession angles (ϕ_1 and ϕ_2) before and after scattering, whatever its angle of incidence
- Precession angles are different for scattered neutrons

$$\phi_1 = \frac{KBd}{k \sin \chi} \text{ and } \phi_2 = \frac{KBd}{k \sin(\chi + \theta)} \Rightarrow \cos(\phi_1 - \phi_2) \approx \cos\left[\frac{KBd \cos \chi}{k \sin^2 \chi} \theta\right]$$

$$P = \int dQ.S(Q).\cos\left[\frac{KBd \cos \chi}{k^2 \sin^2 \chi} Q\right]$$

Polarization proportional to Fourier Transform of *S(Q)*

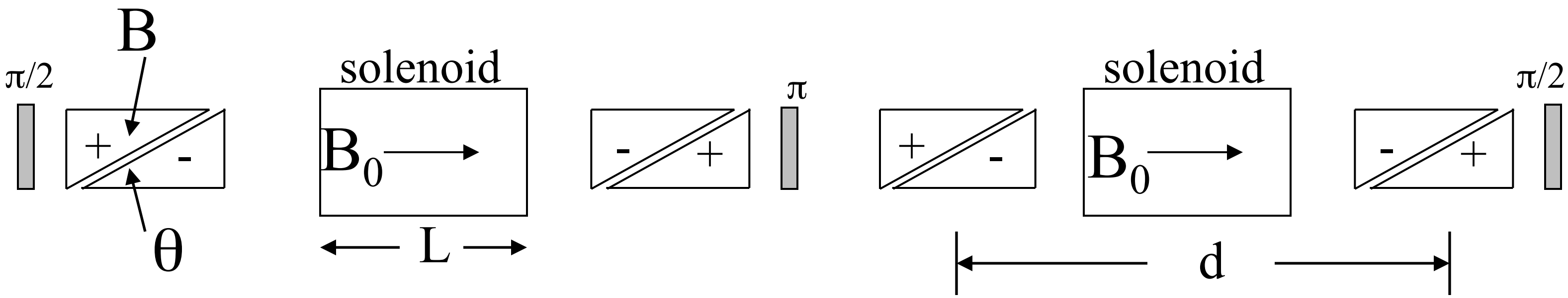
Spin Echo Length, $\zeta = KBd \cos \chi / (k \sin \chi)^2$

How Large is the Spin Echo Length for SESANS?

$Bd/\sin\chi$ (Gauss.cm)	λ(Angstroms)	χ(degrees)	ζ (Angstroms)
3,000	4	20	1,000
5,000	4	20	1,500
5,000	6	20	3,500
5,000	6	10	7,500

It is relatively straightforward to probe length scales of ~ 1 micron

Measuring Correlations in Space & Time



- Use space and time resolving NSE simultaneously
- Large (~ 1 T.m) solenoidal field: $\tau = 186\ B_0.L.\ \lambda^3$ ns (λ in nm)
- Triangular solenoids (~ 0.01 T): $r = 147{,}000$ B.d cot θ. λ^2 nm
- At a reactor source, we could set r and τ independently
- At a pulsed source we could keep B λ^2 constant by varying B with time after a pulse: TOF then maps out τ (might be hard)

- This would be a completely different way of measuring structural correlations using scattering – (r,τ) instead of (Q,ω)