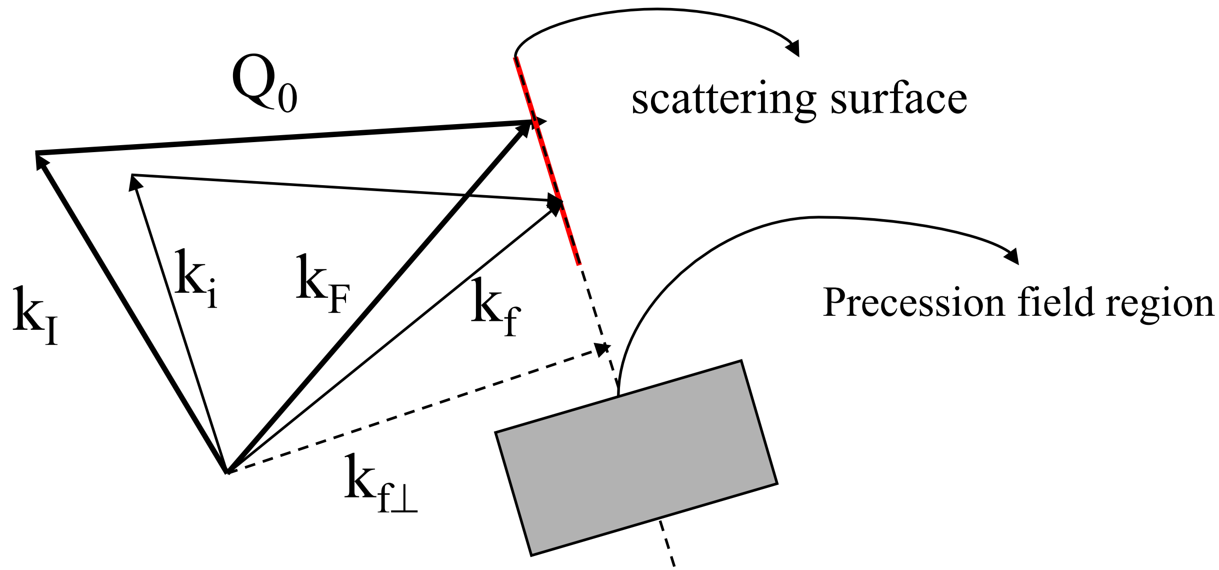
Larmor Labelling of Scattering Angles

Roger Pynn

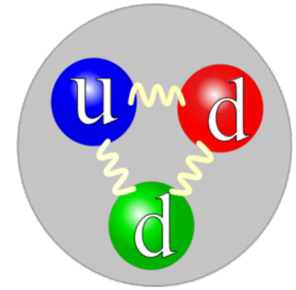
Indiana University, Bloomington &
Oak Ridge National Laboratory

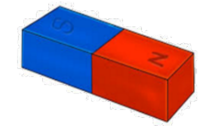
The Picture I made for Reinhard Scherm



But I digress….

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



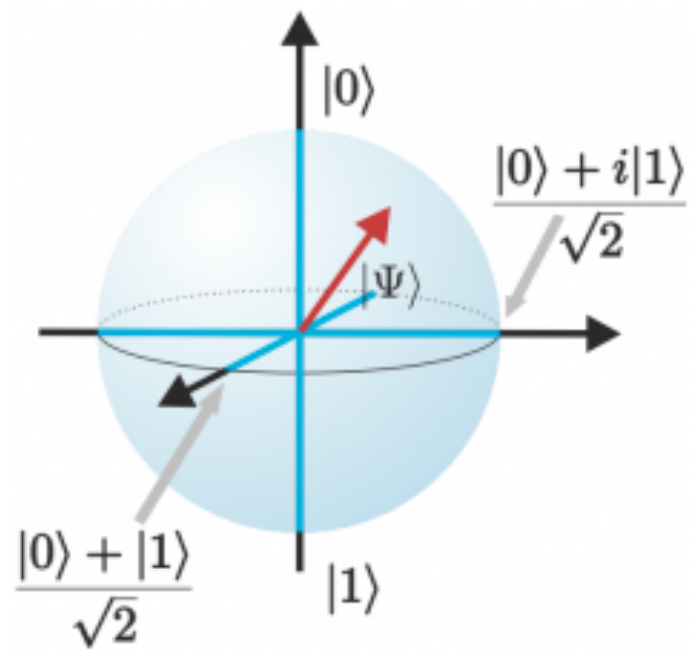


- In an applied magnetic field, B_z, the neutron has two spin eigenstates, “up” and “down” denoted |0> & |1>
  - Magnetic moment either along z or –z (magnetic moment is antiparallel to the spin)
- Quantum mechanics tells us that any neutron spin state is a linear combination of the two eigenstates

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

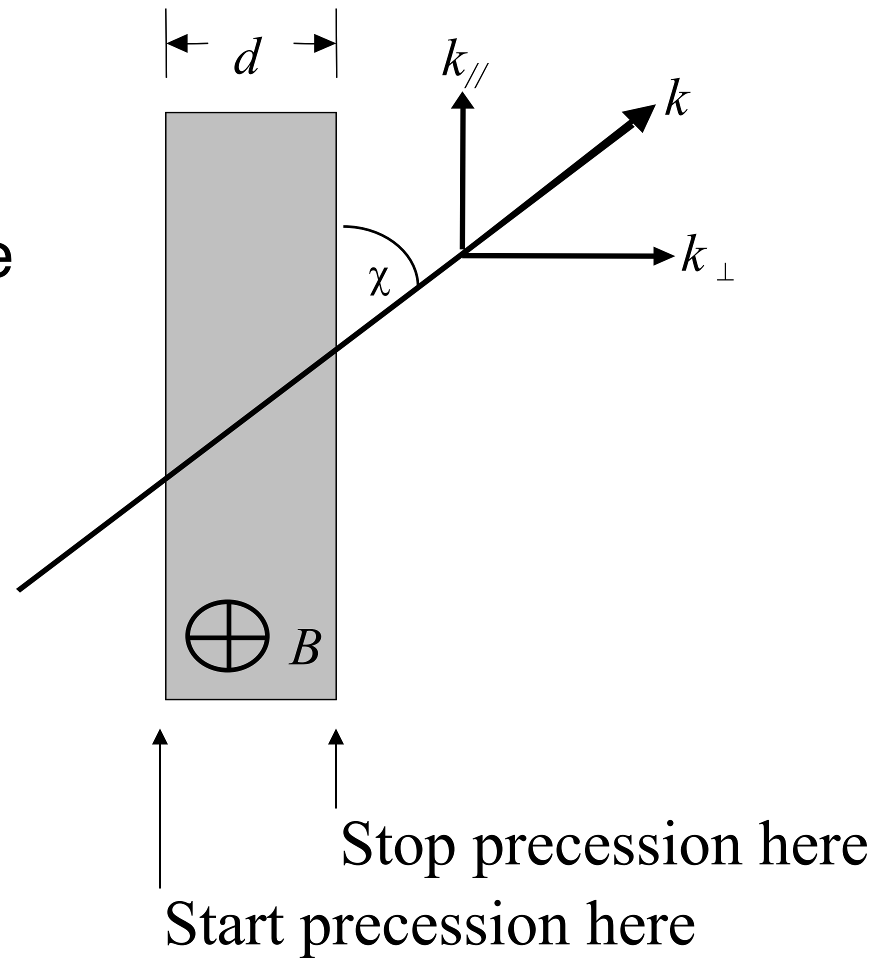
$$\chi = \cos(\theta/2)|0\rangle + e^{i\phi}\sin(\theta/2)|1\rangle$$

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



By "Tilting" the Precession-Field Region, Spin Precession Can Be Used to Code a Specific Component of the Neutron Wavevector

If a neutron passes through a rectangular field region at an angle, its total precession phase will depend only on $k_\perp$.



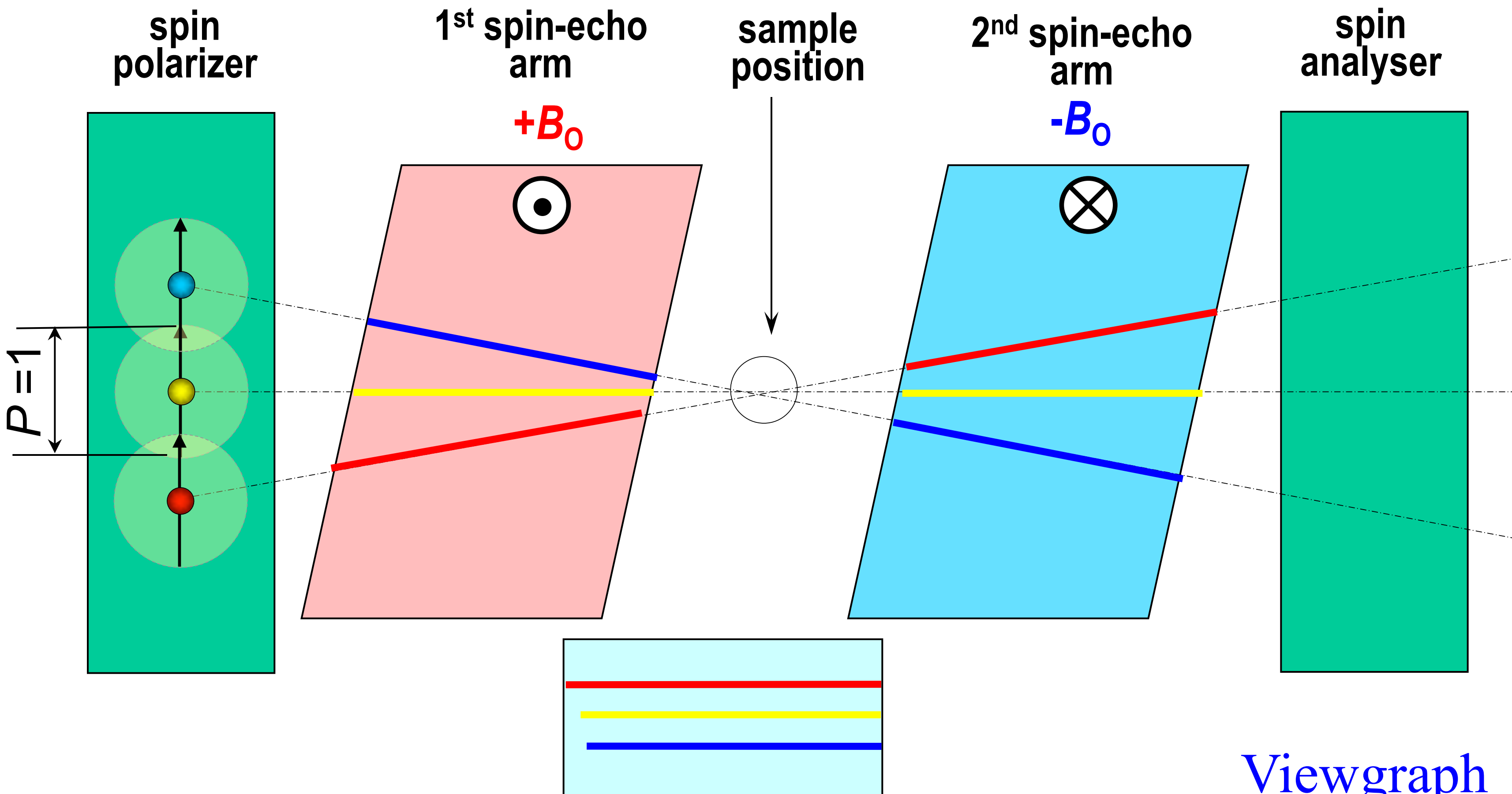
$$\omega_L = \gamma B$$

$$\phi = \omega_L t = \gamma B \frac{d}{\mathrm{v} \sin \chi} = \frac{KBd}{k_\perp}$$

with K = 0.291 (Gauss.cm.Å)$^{-1}$

Roughly 1 turn per 30 Gauss.cm for a 4 Å neutron

The Classical Picture of SESANS

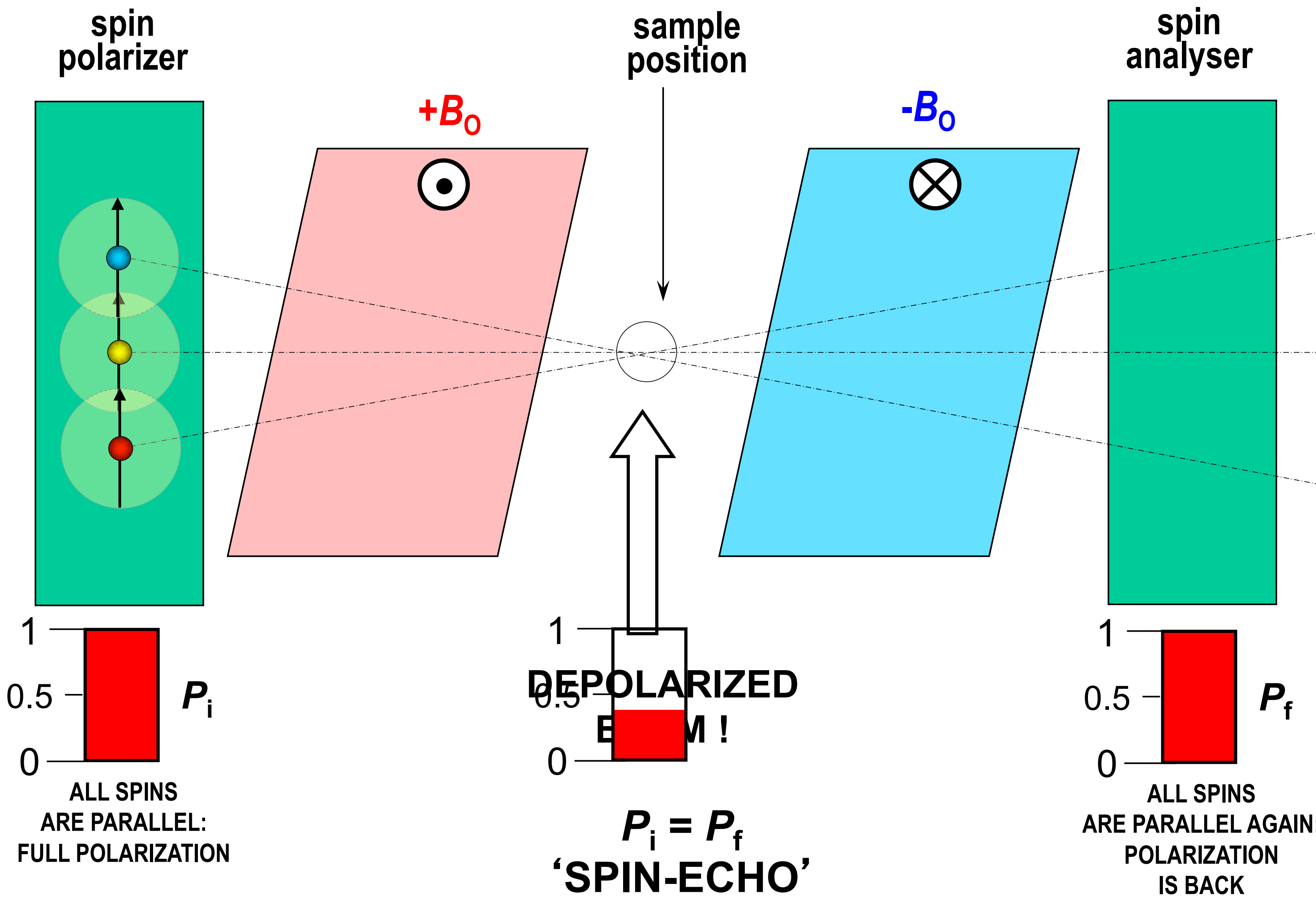


DIFFERENT PATH LENGTHS FOR THE DIFFERENT TRAJECTORIES !

Viewgraph sequence by A. Vorobiev

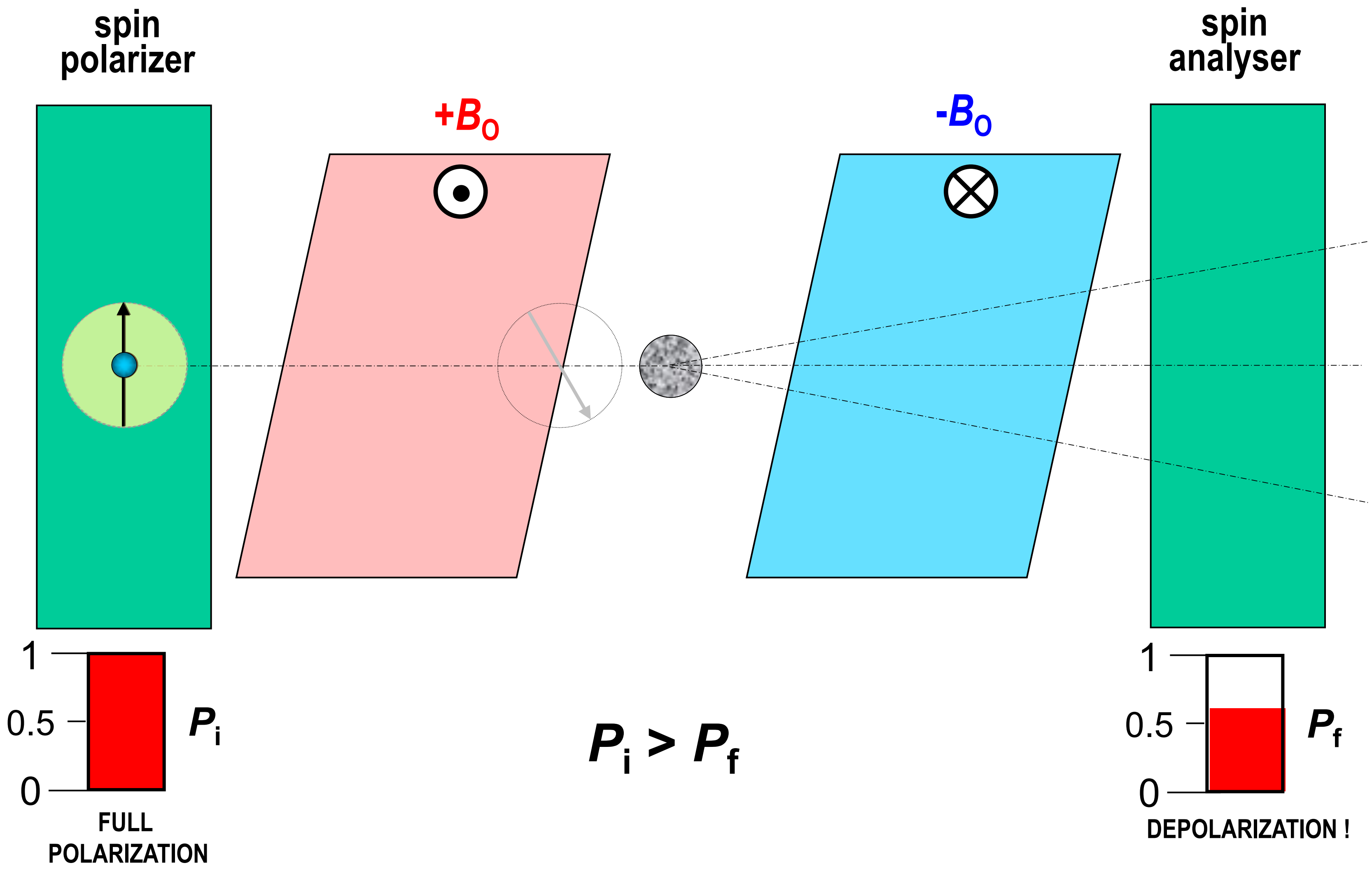
Spin Echo Scattering Angle Measurement (SESAME)

No Sample in Beam

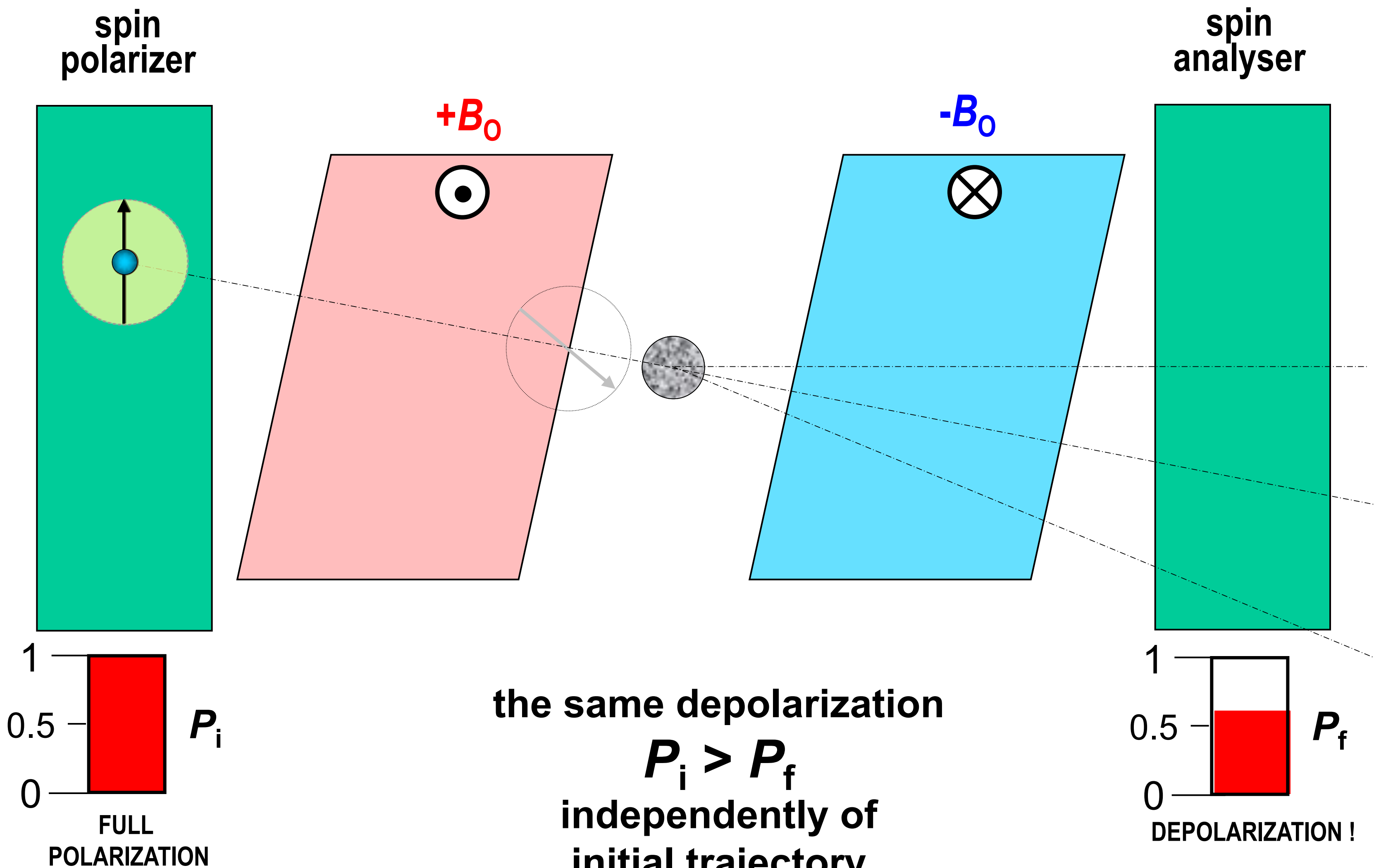


Spin Echo Scattering Angle Measurement (SESAME)

Scattering by the Sample



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

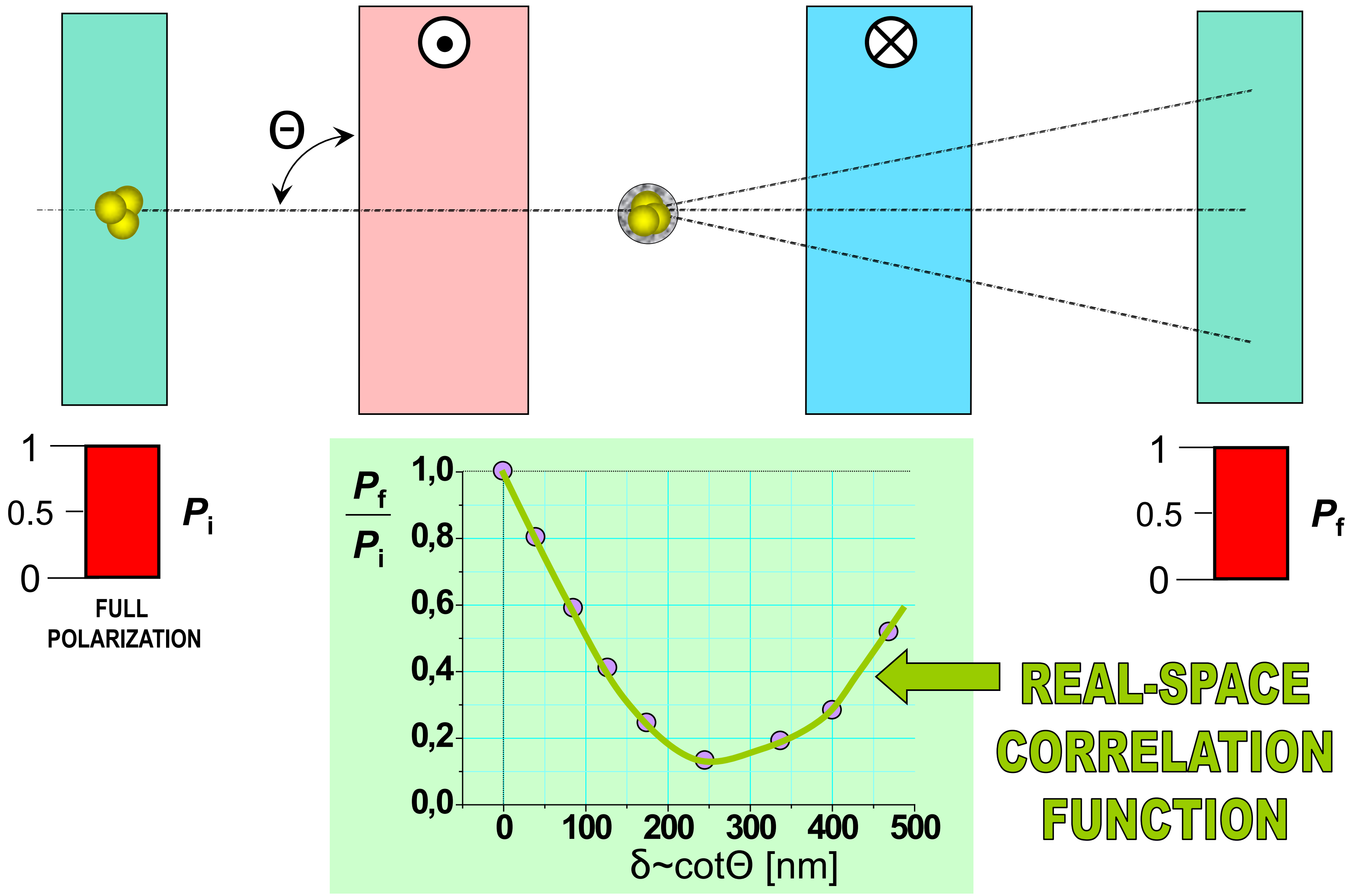


the same depolarization

$$P_i > P_f$$

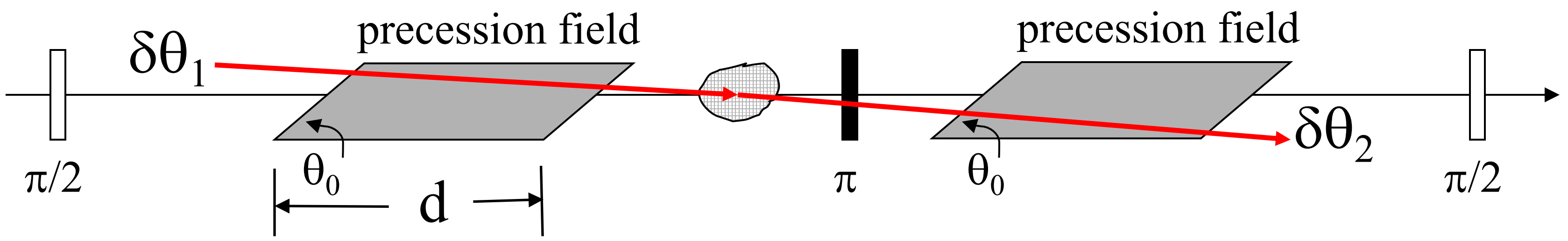
independently of initial trajectory

Spin-echo angular encoding: the experiment



NSE Angle Coding Illustrated for SESANS

- Make the number of spin precessions depend on the neutron's direction of travel instead of (only) its speed…..



$$\omega_L = \gamma B$$

$$\phi_1 = \omega_L t_1 = cBd\lambda(1 - cot\theta_0 . \delta\theta_1) \;\; where \;\; c = \frac{\gamma m}{h} = 4.635 \; x \; 10^{14} T^{-1} m^{-2}$$

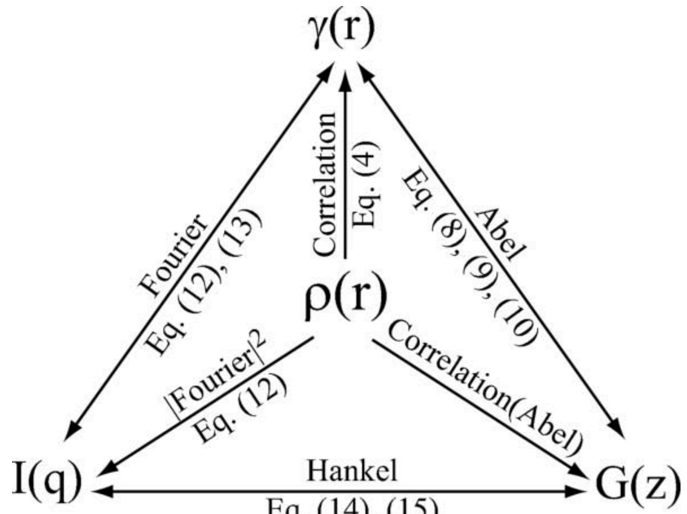
$$\phi_1 - \phi_2 = \mathrm{Q} \; \frac{cBd}{4\pi} \lambda^2 cot\theta_0 = Q.\zeta$$

$$\frac{P}{P_0}(Z) = \frac{\int_0^\infty dQ_y \int_0^\infty dQ_z \frac{d\sigma}{d\Omega} \cos(Q_z Z)}{\int_0^\infty dQ_y \int_0^\infty dQ_y \frac{d\sigma}{d\Omega}} = e^{\Sigma[G(Z)-1]}$$

ζ is called the spin echo length

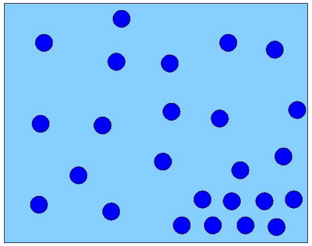
Σ = total probability for single scattering; G(Z) is a correlation function

Correlation Function



Andersson et al J. Appl. Cryst. (2008). 41, 868–885

Local Density Distribution $\rho(\vec{r})$



Debye Correlation Function

$$\gamma(r) = \frac{1}{V}\left\langle \int_V \rho(\vec{r}\,')\rho(\vec{r}\,' + \vec{r})d^3\vec{r}\,' \right\rangle$$

SANS

SESANS

$$I(Q) = \frac{d\Sigma}{d\Omega}(Q) = 4\pi\int_0^\infty \gamma(r) J_0(Qr) r^2 dr$$

Fourier

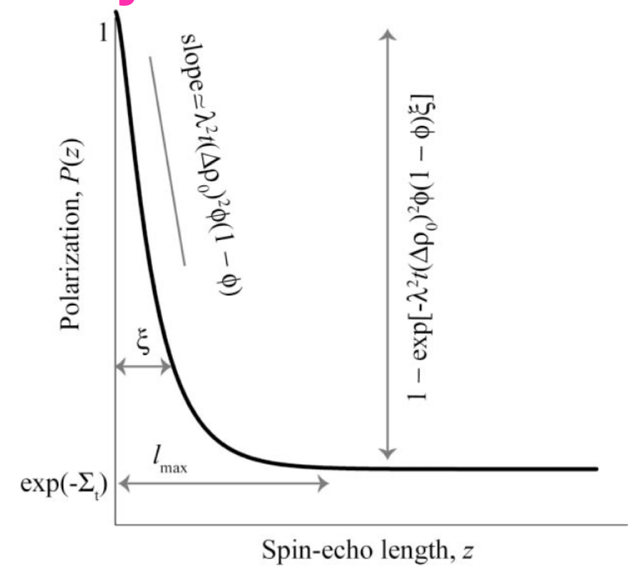
$$G(z) = 2\int_z^\infty \gamma(r)\frac{r}{\sqrt{r^2 - z^2}}dr$$

Abel

What we Actually Measure with SESANS for a Bulk Two-Phase System

$$\frac{1}{\lambda^2}\ln\left(\frac{P(z)}{P_0}\right)=\Sigma_t[G(z)-1]$$

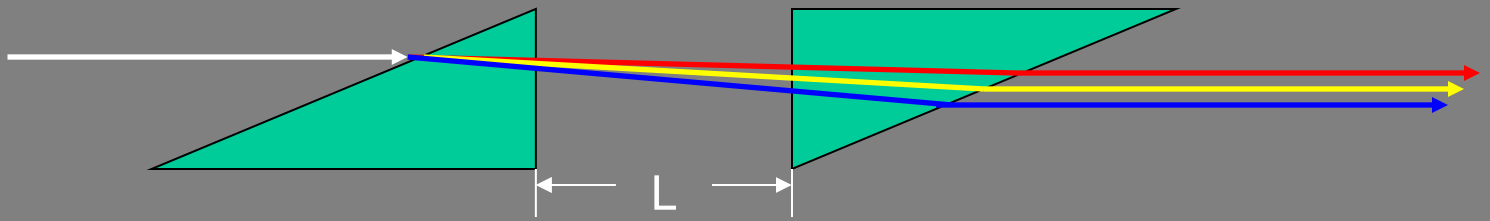
since $G(\infty)=0$, $\quad \Sigma_t=-\dfrac{1}{\lambda^2}\ln\left(\dfrac{P(\infty)}{P_0}\right)$



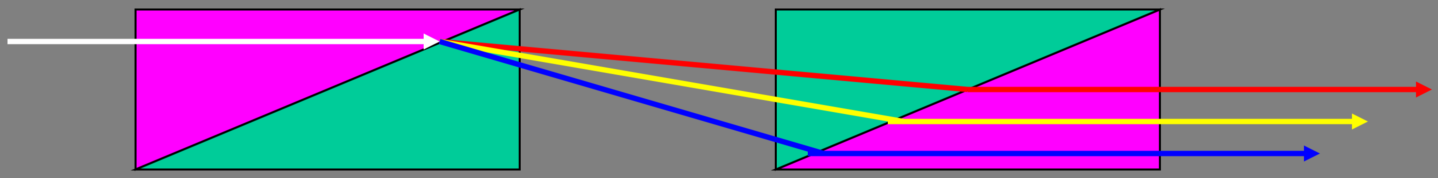
also, $\Sigma_t=t\Delta\rho^2\phi(1-\phi)\xi$ where the correlation length $\xi=2\int\gamma(r)\,dr$

- G(z) is a projection of the usual Debye density-density correlation function: it is a **real space correlation function**
- The value of z where $\ln(P)/\lambda^2$ becomes flat tells us the maximum size of density correlations in the system (particle diameter for dilute spherical objects)
- The value of $\ln(P)/\lambda^2$ at $z\to\infty$ measures the total scattering

Increasing the Spin Echo Length

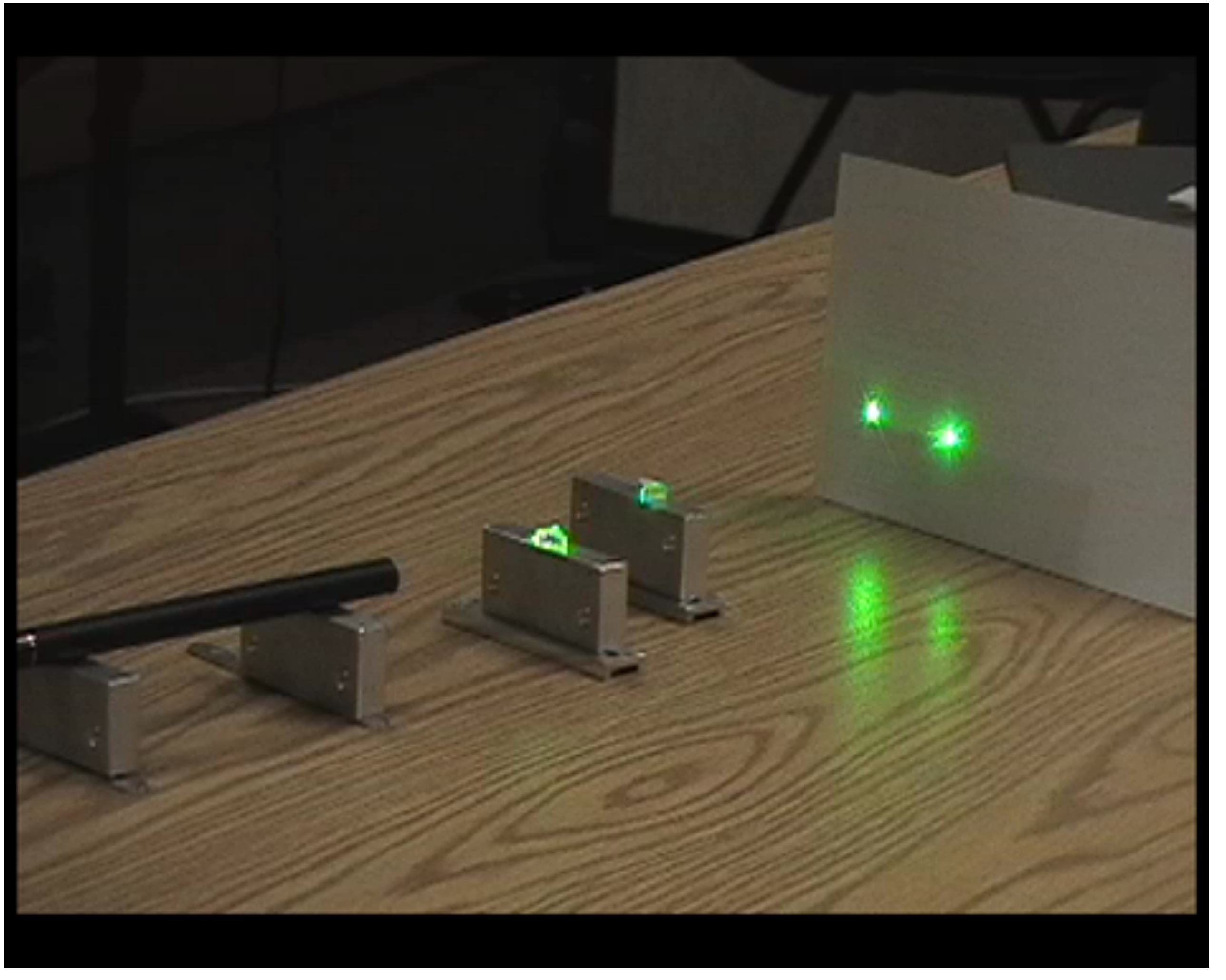


Cutting the parallelogram into prisms increases the length of the parallelogram (d in the previous VG) and adds a rectangular region that does no angle coding. In QM language, it allows the spatial separation up (red) and down (blue) states to be controlled by the separation of the prisms

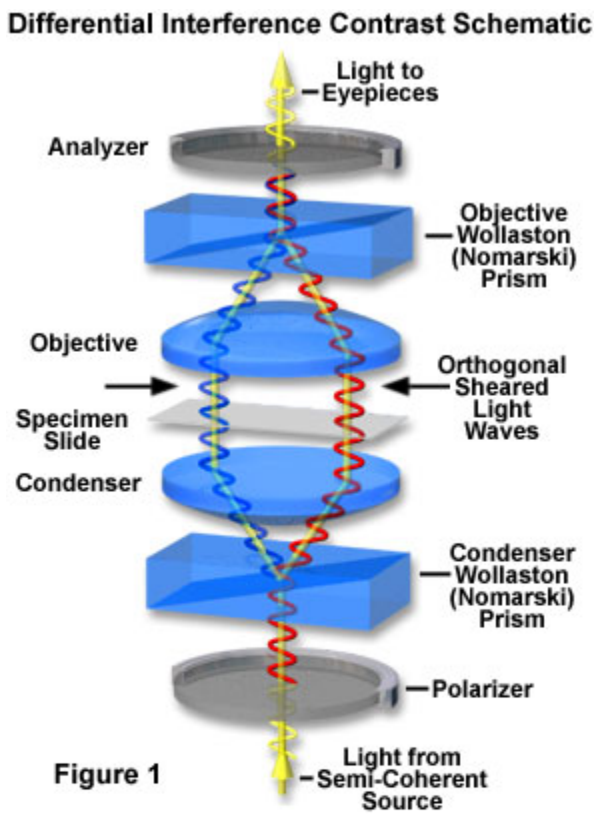


Adding prisms with the opposite dispersion (i.e. opposite magnetic fields) doubles the separation of the red and blue rays (i.e. doubles the spin echo length)

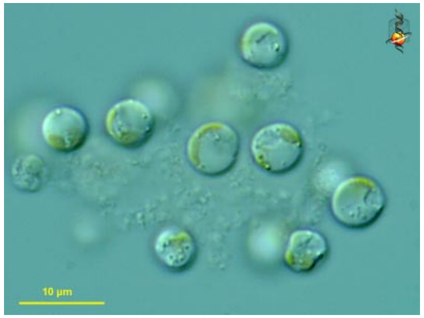
The Quantum View



Differential Interference Contrast Microscopy



on Prism

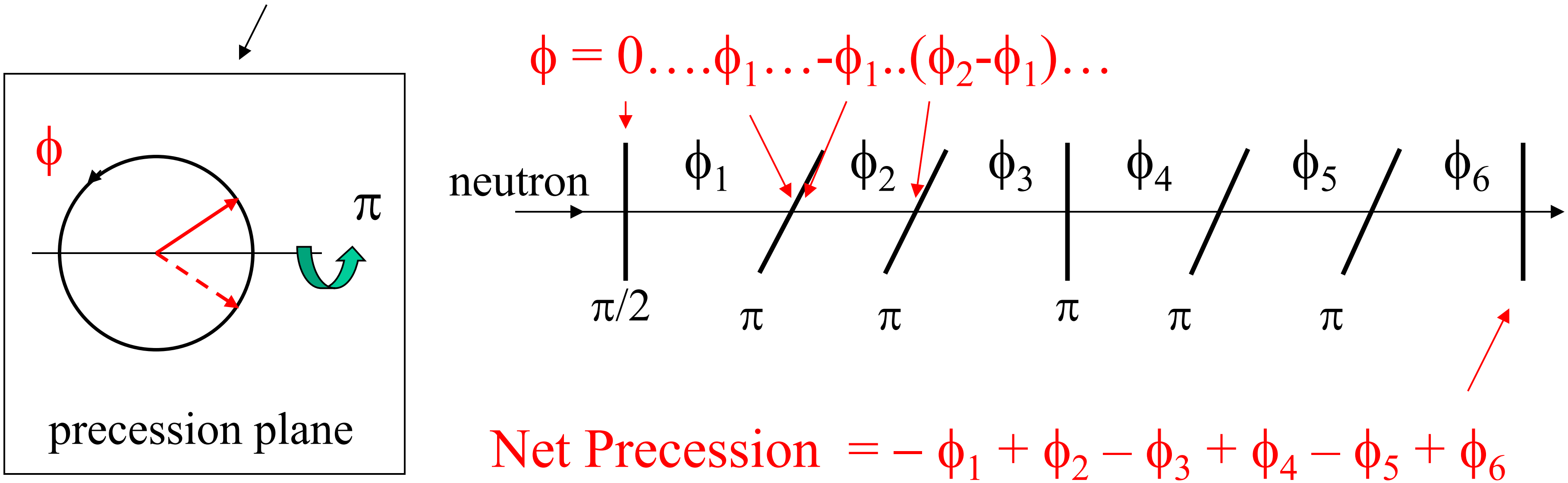


Eukaryotic Algae

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.

Using Sequential π Flippers to Implement SESANS

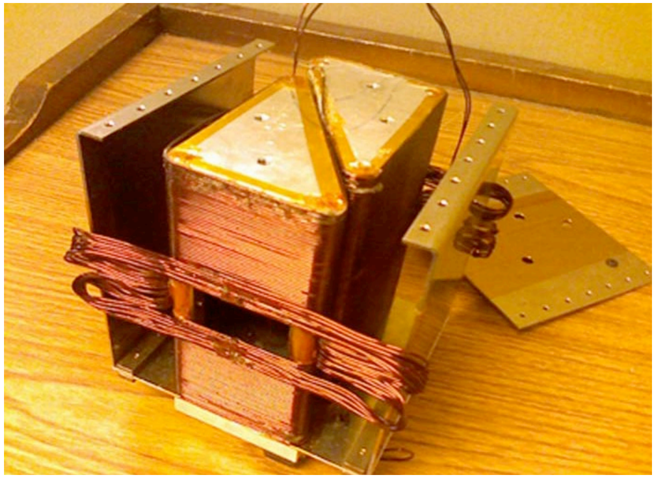
An element that performs a π rotation about an axis in the precession plane changes the sign of prior precession angles



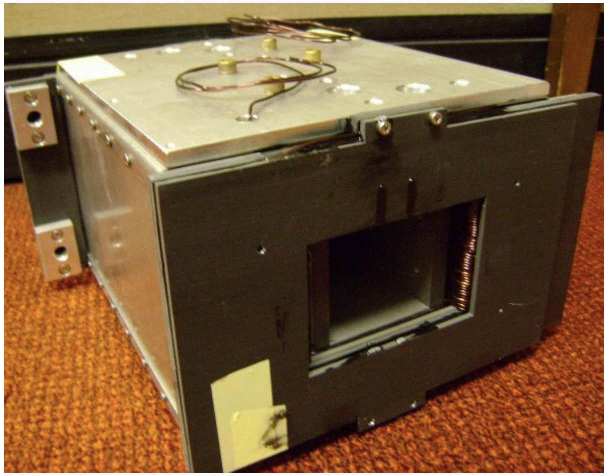
- Total net precession angle = $(\phi_4 + \phi_5 + \phi_6) - (\phi_1 + \phi_2 + \phi_3) - 2(\phi_5 - \phi_2)$
- The first two terms are the familiar spin-echo terms
  - Depend only on neutron velocity as shown here
- Last term depends on velocity *and* angle of neutron trajectory
  - Picks up an extra factor of 2 compared to using a parallelogram-shaped field
  - The Wollaston prism is a pi-flipper

How can we realize the magnetic Wollaston prism in practice?

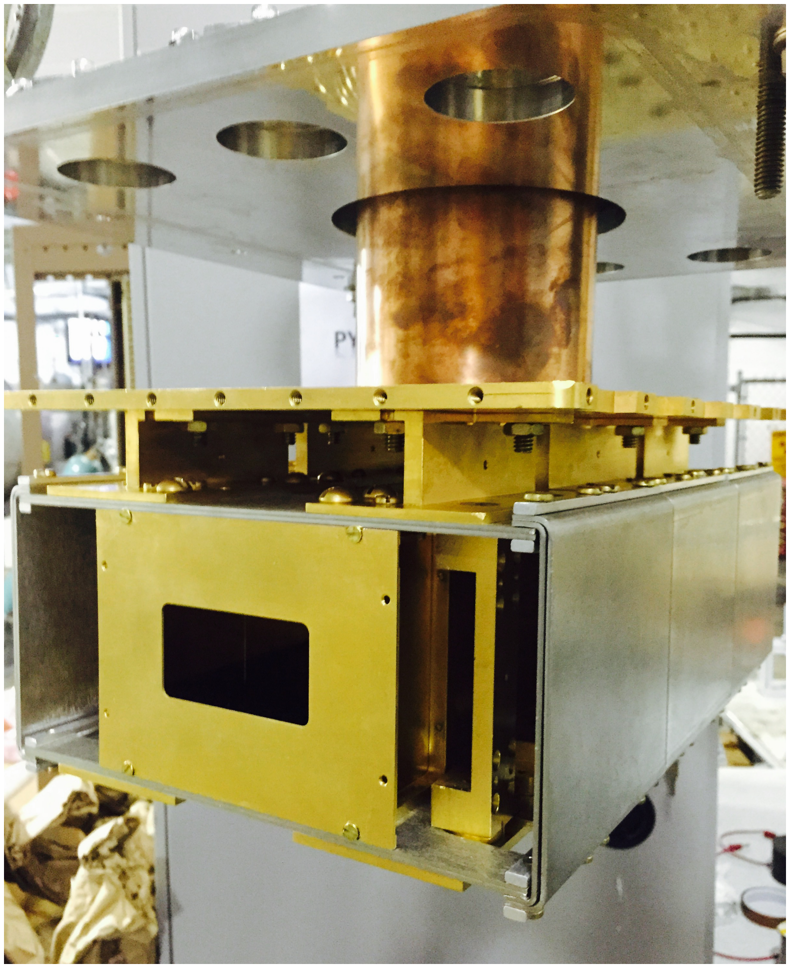
- We built several generations of air- and water-cooled WPs and (re)discovered the importance of using symmetry (pi flip) to cancel decoherence effects (Larmor phase aberrations)
- Ultimately, room temperature coils are limited by:
  - Flatness of coils (waviness, discrete wires)
  - Neutron absorption by wires
  - Maximum achievable magnetic field

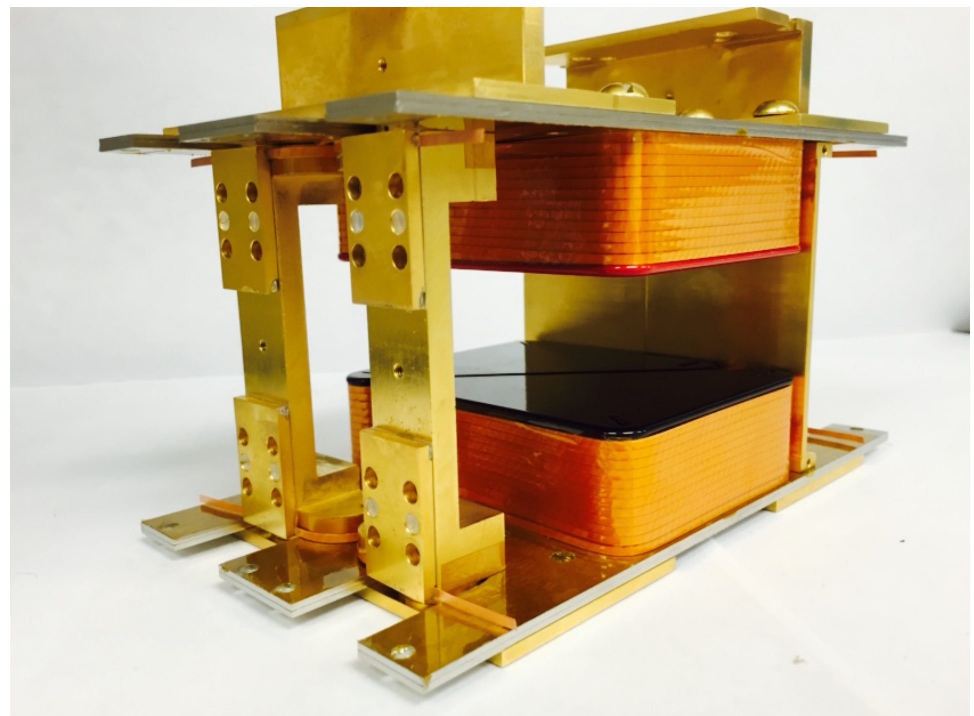


Paul Stonaha

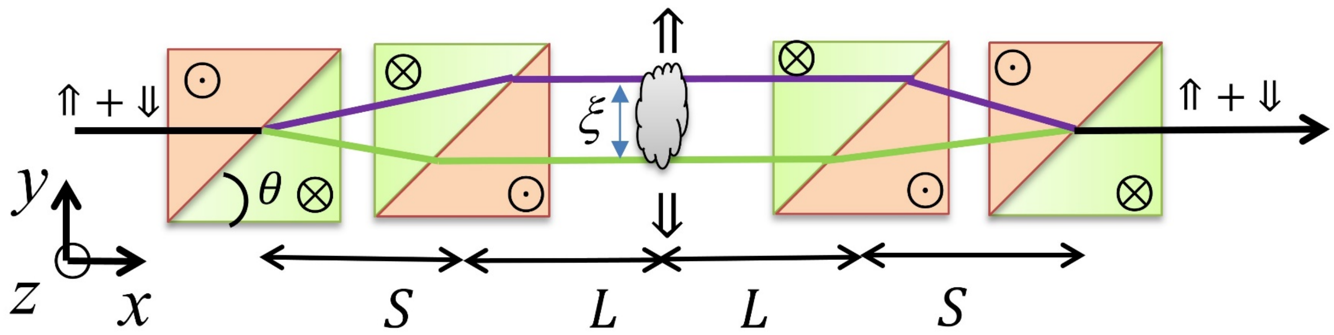


Finally we built Wollaston prisms using HTS tape and HTS films surrounding the field regions to achieve high fields & good uniformity





Spin echo small angle neutron scattering- a polarized neutron interferometer



$$\zeta = c\lambda^2 BS\cot\theta/2\pi \qquad c = 1.47\times10^{14}\,T^{-1}m^{-2}$$

$$\zeta = 590 \text{ nm for } \lambda = 4\,\text{Å},\ \text{B} = 0.1\text{ T}\ \&\ \text{S} = 0.5\text{ m}$$

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

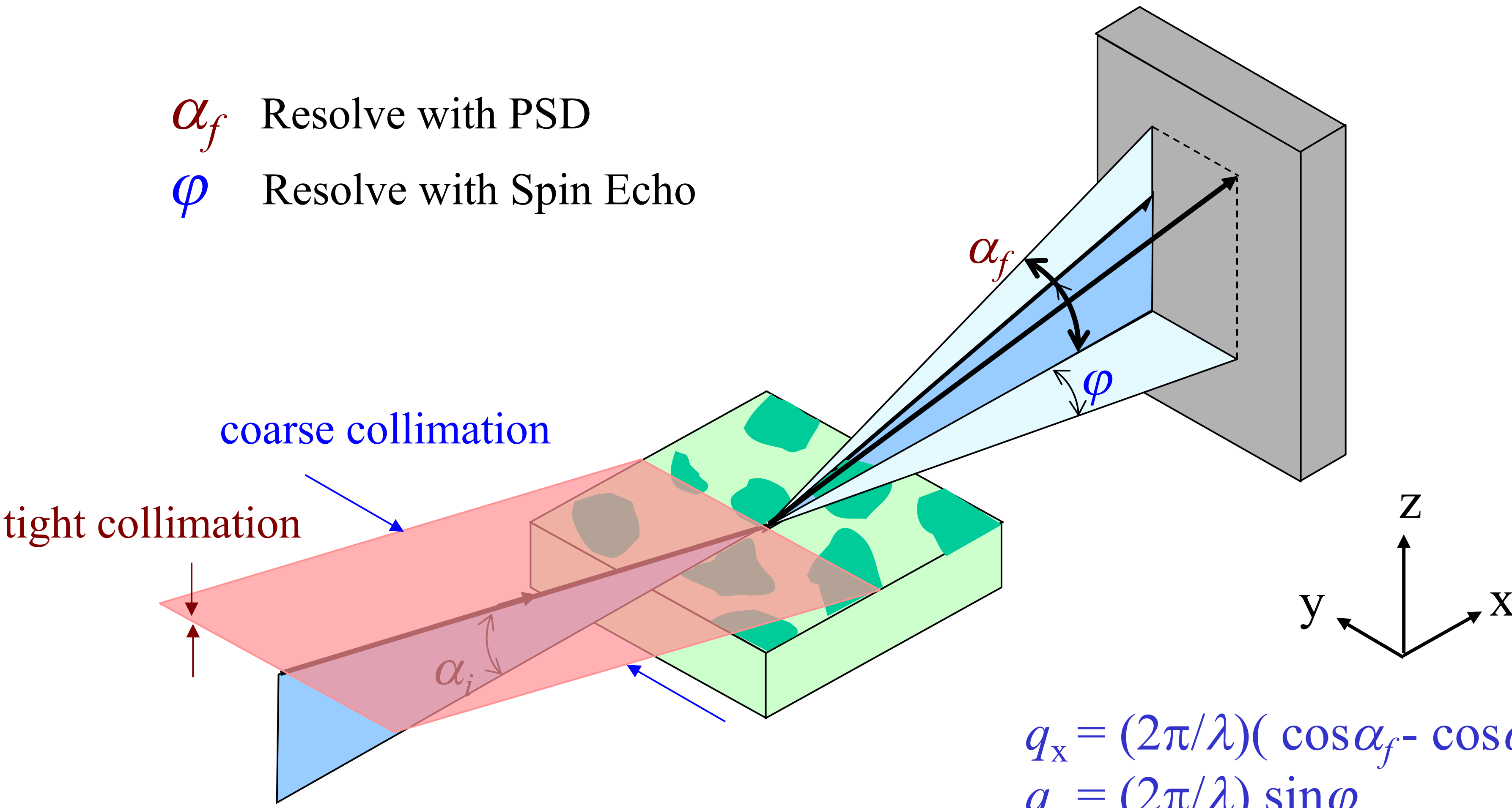
- The separation of the two devices, S
- Inclination angle, θ
- Field intensity

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

SESANS in reflectometry - SERGIS

α_f Resolve with PSD

φ Resolve with Spin Echo

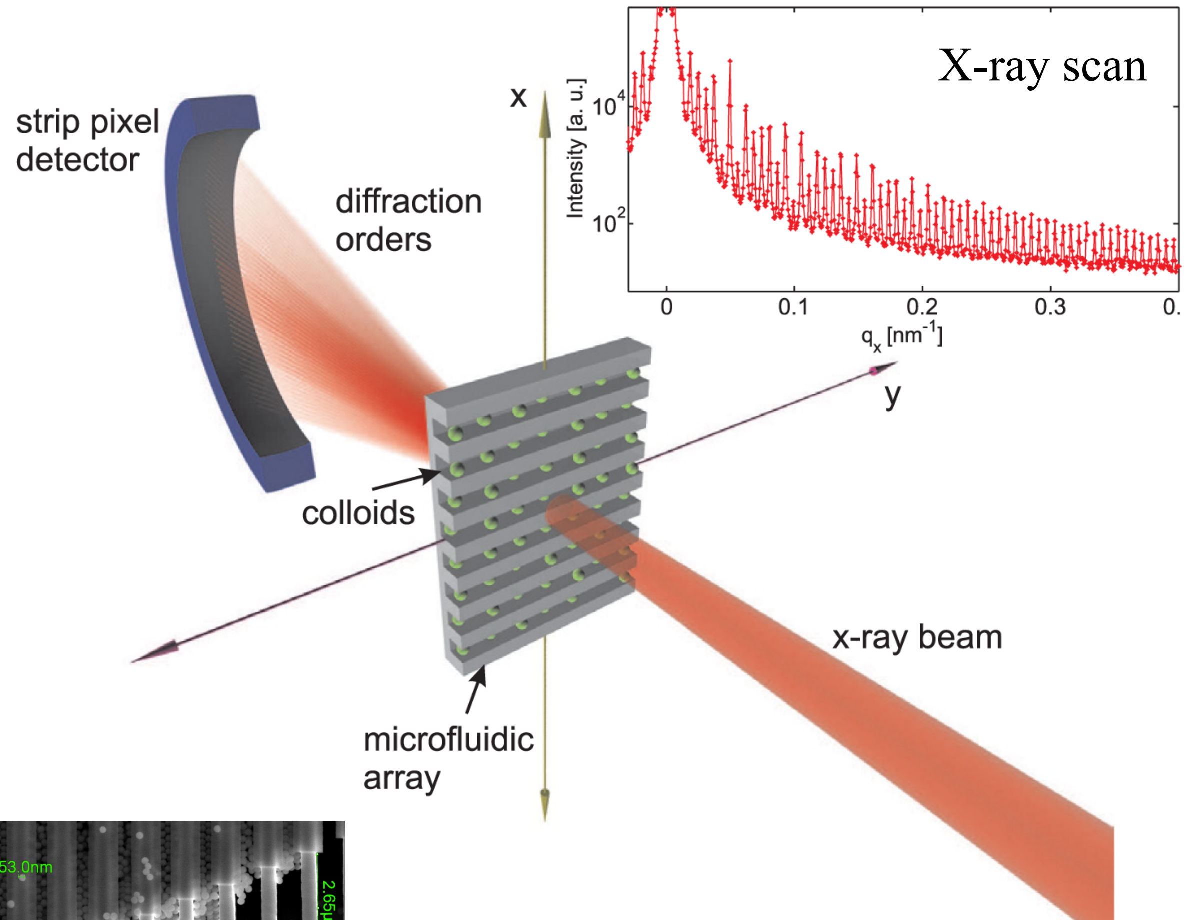


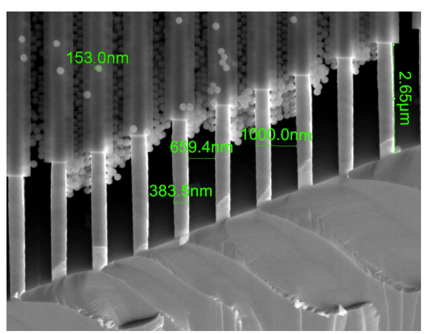
$$q_x = (2\pi/\lambda)(\cos\alpha_f - \cos\alpha_i)$$
$$q_y = (2\pi/\lambda)\sin\varphi$$

Note that scattering at a particular value of q is spread over larger angles α_f than ϕ

X-Ray & Neutron Grating Holography

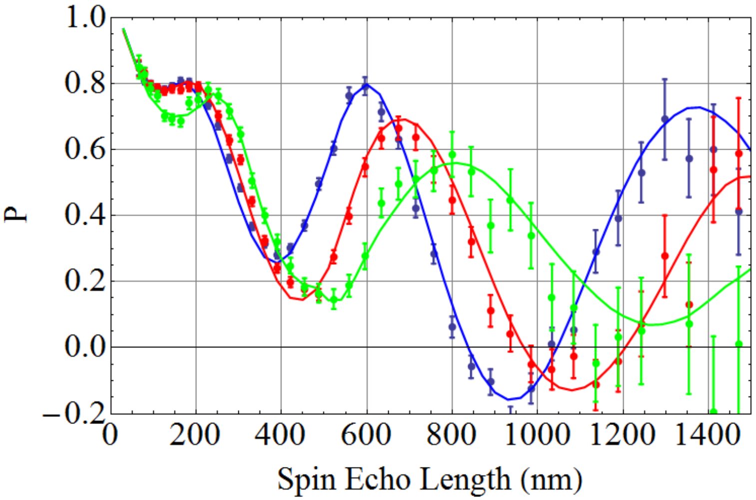
- Grating creates near-field modulation of amplitude & phase
- Contents of grooves act as a weak-phase scattering object
- Deconvolve pattern produced by bare grating to deduce structure in grooves
- X-rays measure Bragg peaks & calculate Patterson function
- Neutrons measure Patterson function using SESANS

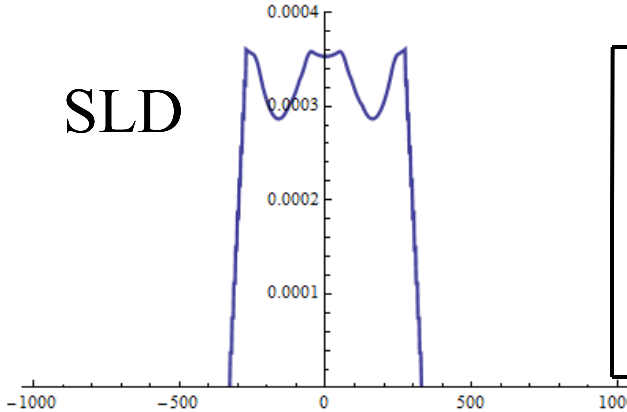


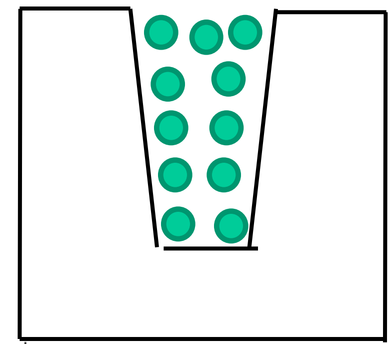


SEM of grating with silica particles after neutron experiment

Neutron Holography Results

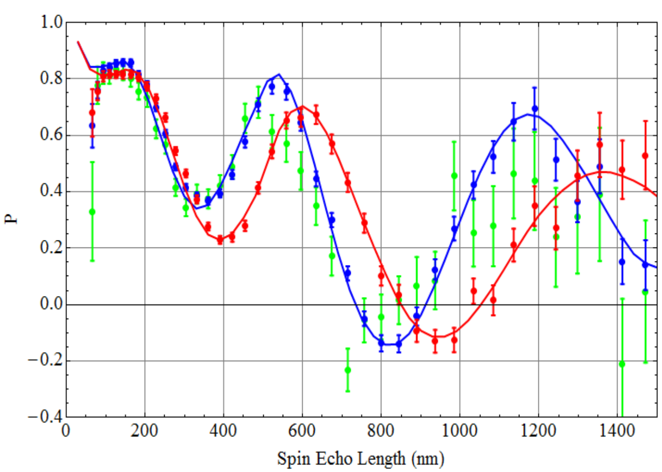


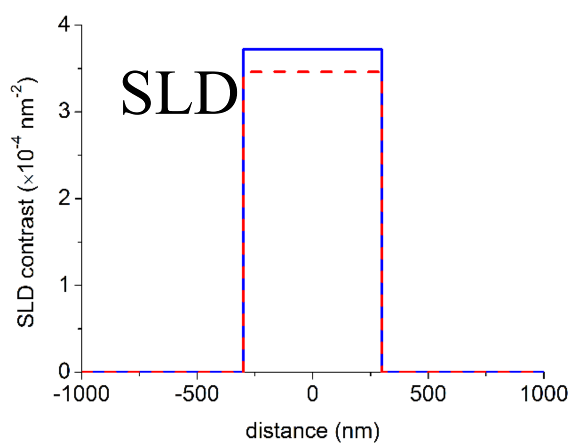




Above: Blue: carrier fluid only; Red: with colloid at pH = 9.7; Green: same as red 24 hours later

Below: Blue: carrier fluid only; Red: with colloid ar pH = 10.6



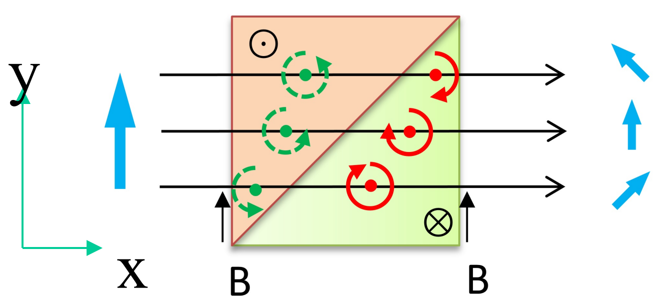


Magnetic Wollaston Prisms

- Label a neutron's distance from the beam center *$(y\text{-}y_0)$* by its Larmor precession angle
- Each prism consists of two triangular magnetic-field regions with oppositely directed B fields and sharp boundaries

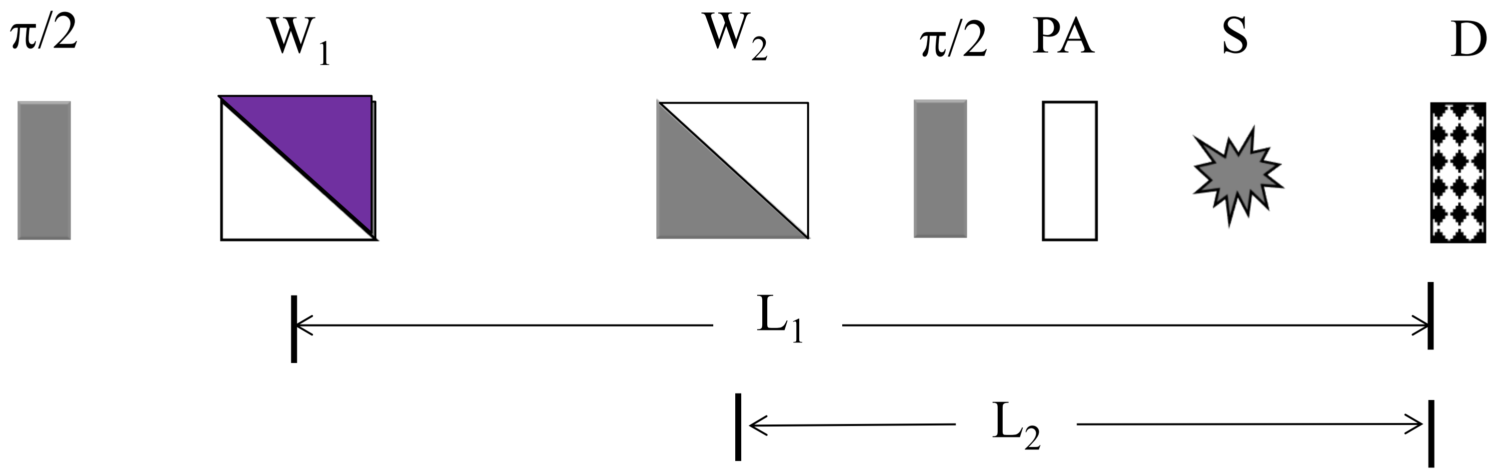
$$\phi_{total} = 2cB\lambda(y - y_0)cot\theta$$

~4 rads/mm for 4 Å neutrons and B = 100 G



If a polarization analyzer is placed after the prism, the measured intensity varies as $1+\cos(\phi_{total})$

Spin Echo Modulated SANS (SEMSANS)

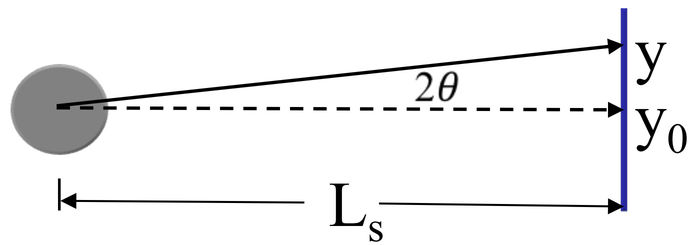


- Consider a neutron arriving at a point on the detector at position *y* (relative to the center) with divergence angle, θ

$$\varphi_1 = c\lambda B_1(y + L_1\theta) \qquad \varphi_2 = -c\lambda B_2(y + L_2\theta)$$

- The total phase is independent of θ if $B_1L_1 = B_2L_2$ so we get a cleanly modulated intensity on the detector **even with a divergent beam**
- Period is: $p = \pi . tan\theta / c\lambda(B_2\text{-}B_1)$
- I.e p ~ 1.7 mm for 4 Å neutrons and B_2-B_1 = 100 G

What do we measure with SEMSANS?



$$y - y_0 = 2\theta . L_s = \frac{Q\lambda}{2\pi}$$

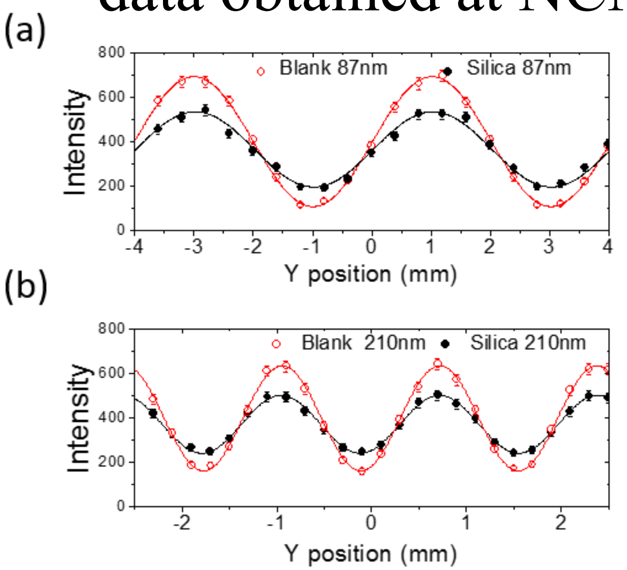
$$P(y) = \int_{det} \cos(\frac{2\pi y_0}{p}) \frac{d\sigma}{d\Omega} dy_0 / \int_{det} \frac{d\sigma}{d\Omega} dy_0$$

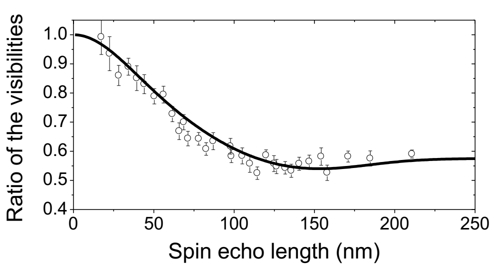
$$P(y) = \frac{\int_{det} \cos\left(\frac{2\pi(y - \frac{Q\lambda L_s}{2\pi})}{p}\right) \frac{d\sigma}{d\Omega} dQ}{\int_{det} \frac{d\sigma}{d\Omega} dQ} = P(y_0) \frac{\int_{det} \cos(Q\,\zeta) \frac{d\sigma}{d\Omega} dQ}{\int_{det} \frac{d\sigma}{d\Omega} dQ}$$

With $\zeta = \frac{\lambda L_s}{p}$ as the spin echo length

SEMSANS for a homogeneous sample: every point on the detector measures the same thing

data obtained at NCNR



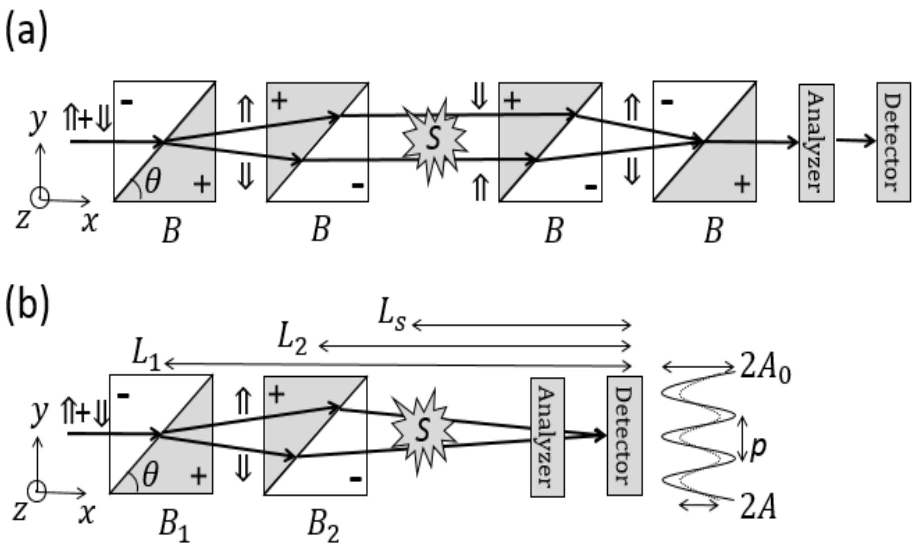


silica colloid (NCNR)

$$\frac{P_s}{P_0} = e^{\Sigma_t (G(\xi)-1)} \qquad \zeta = \lambda L_s / p$$

- P_s = polarization at any point on detector with sample
- P_0 = polarization without sample
- Σ_t is the fraction of the beam (single) scattered by the sample
- G(Z) is a real space correlation function measured at a spin echo length, Z. It is the same function as is measured using SESANS
- Data analysis can be tricky because of zero-crossing of polarization
- Can also implement dark-field radiography this way
  - each point on the detector measures a SESANS curve for a small area of the scattering object

Comparison of SESANS & SEMSANS



SESANS

- ❑ Four Wollaston prism needed
- ❑ All the fields are equal and balanced
- ❑ Paths at the sample are parallel
- ❑ Non-magnetic sample environment
- ❑ Low resolution detector is OK

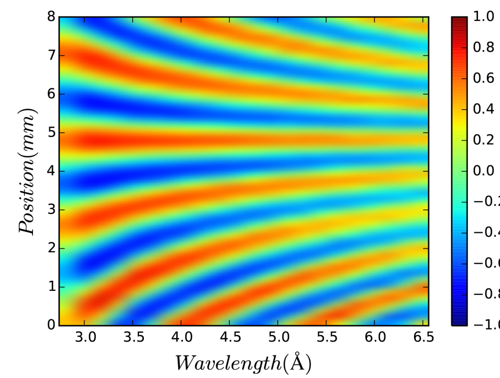
SEMSANS

- ❑ Two Wollaston prism needed
- ❑ The fields are not balanced
- ❑ Focused towards the sample
- ❑ Sample environment relaxed
- ❑ High resolution detector is required

They both measure the correlation function of the sample.

F. Li, *et al*, J. Appl. Cryst. 49, doi:10.1107/S1600576715021573.

Double WP unit on LARMOR with ^{3}He polarization analyzer and 55-μm-pixel, 28x28 mm^2 Tremsin Detector

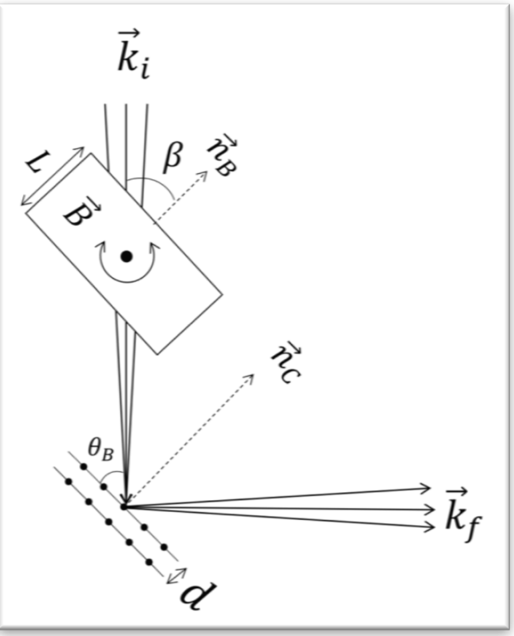


Fringe period is chromatic
Visibility is achromatic
(opposite of grating-based far-field interferometer)

Polarization projected on to horizontal coordinate as a function of position and wavelength.

Independent of beam divergence – would work with a lens

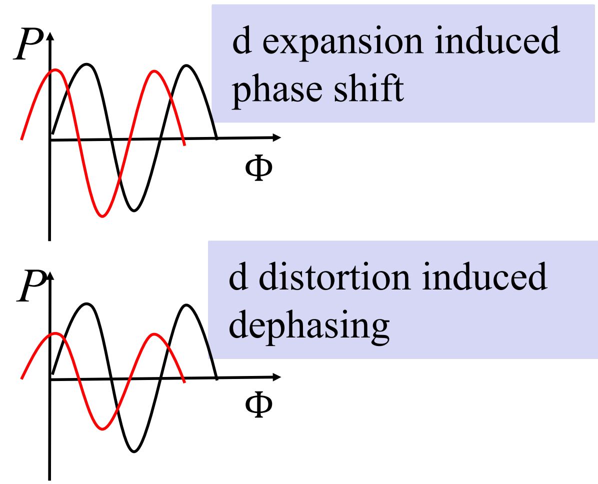
Larmor diffraction to achieve higher resolution



$$\Phi = \frac{\gamma_N m B L}{h k_{i,\perp}}$$

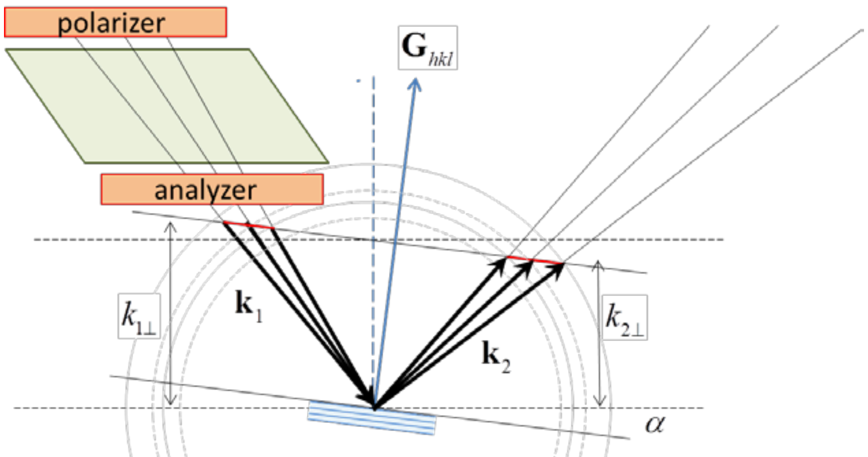
$$k_{i,\perp} = \frac{2\pi}{d}$$

$$\Phi = \frac{\gamma_N m B L d}{2\pi h}$$

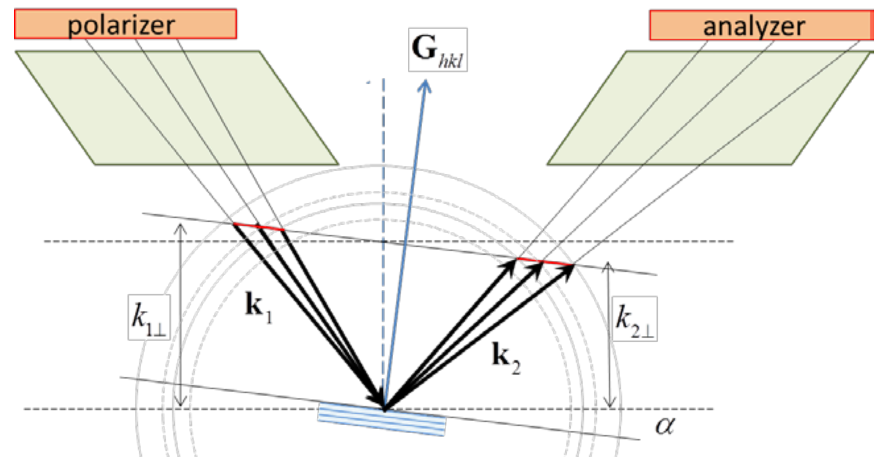


- ❖ The d spacing distribution is encoded into Larmor phase
- ❖ High resolution for d-spacing <u>expansion</u> $\Delta d/d \sim 10^{-6}$.
- ❖ The tilting angle has to match the crystal plane.

Single arm Larmor diffraction



Double arm Larmor diffraction

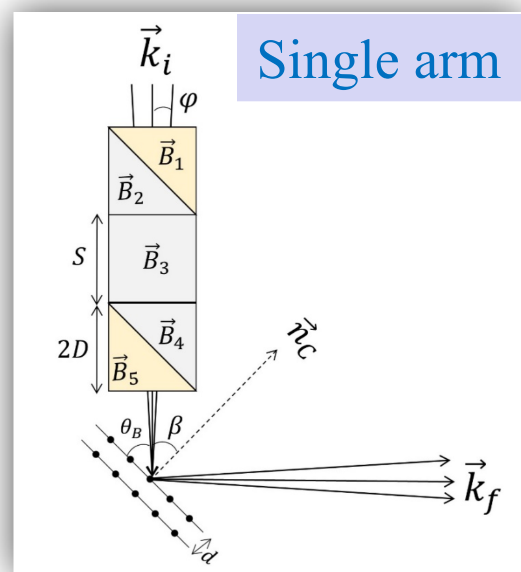


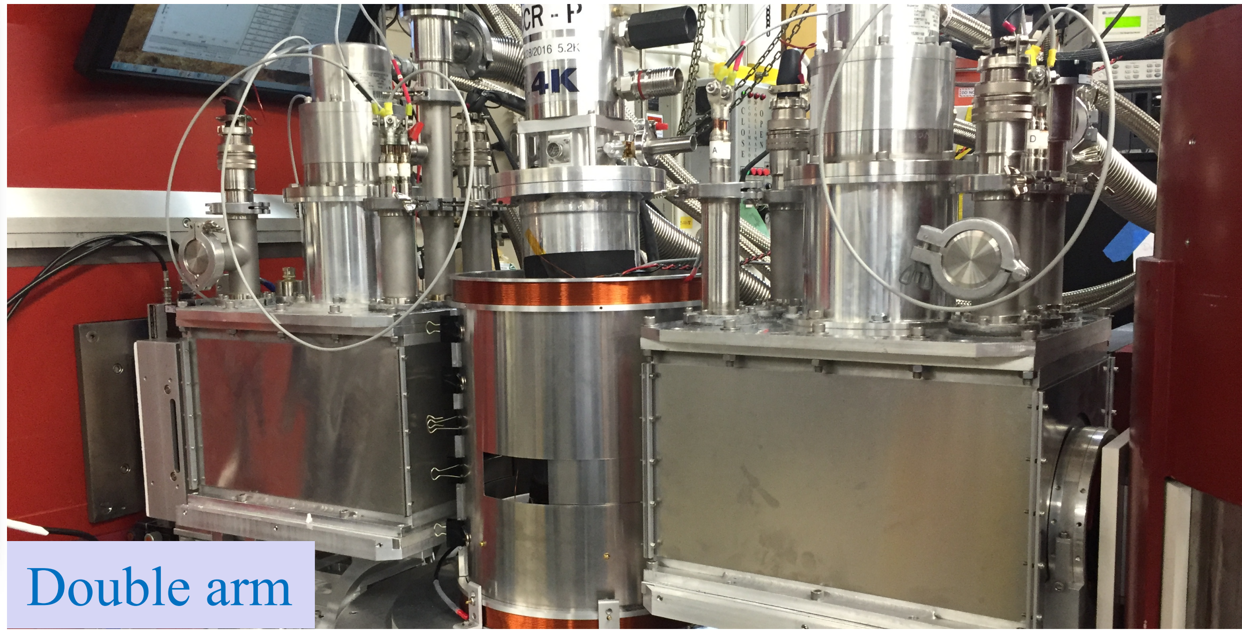
Drawback: Lower resolution and not for mosaic sample
Advantage: OK for magnetic sample

Drawback: problematic for magnetic sample
Advantage: higher resolution, OK with mosaic sample

Courtesy of Dr. Ad van Well

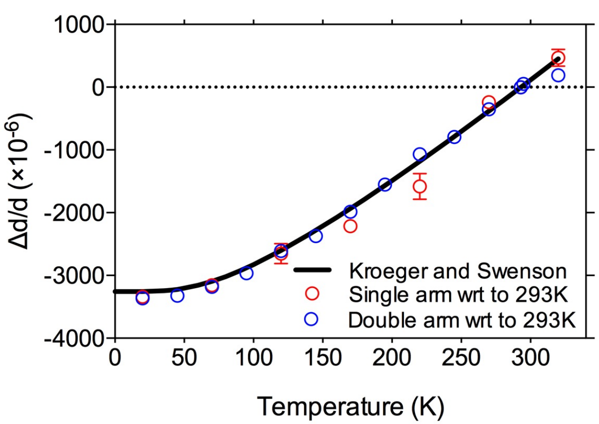
Larmor diffraction with Wollaston Prisms: all electromagnetic tuning





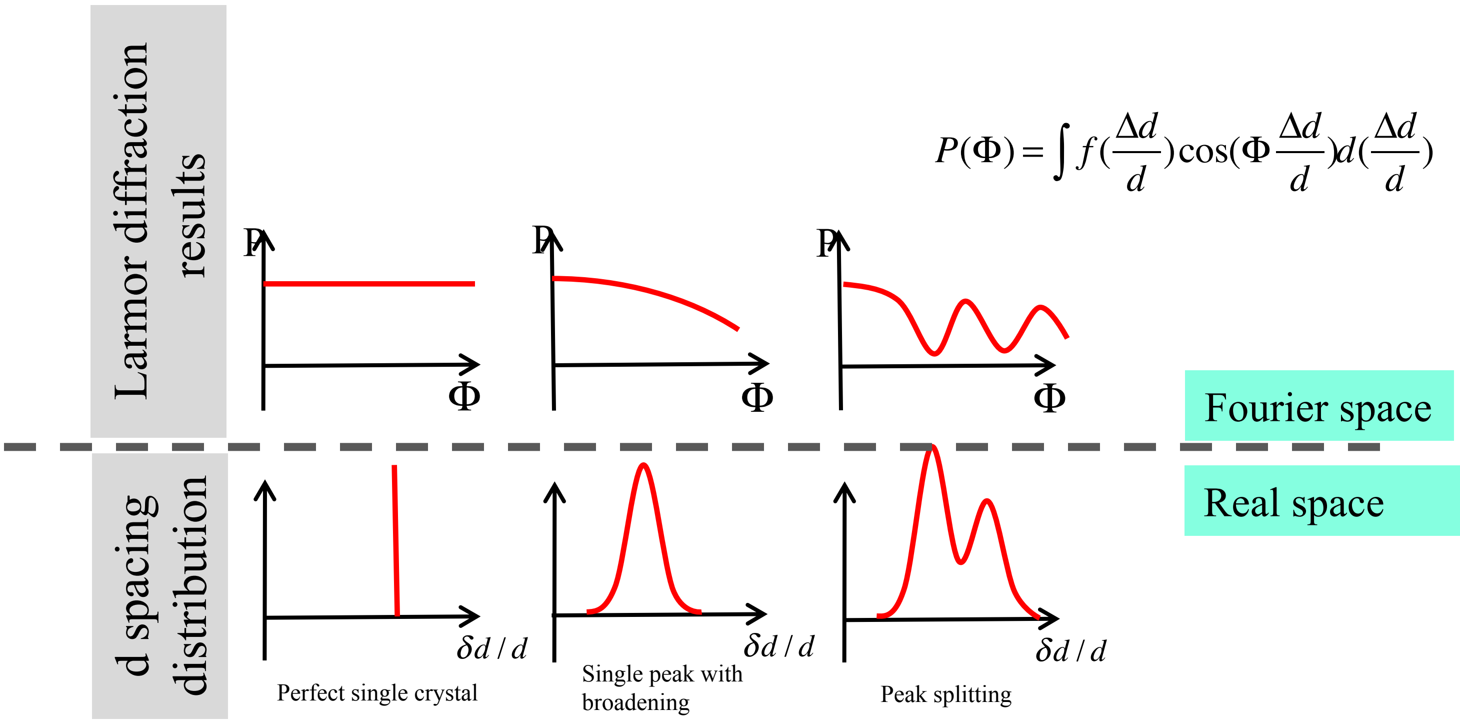
HB-1 @ ORNL
Feb. 2017

Thermal expansion of silicon



F. Li et al. J. Appl. Cryst. (2014). 47

Rules of thumb for Larmor diffraction



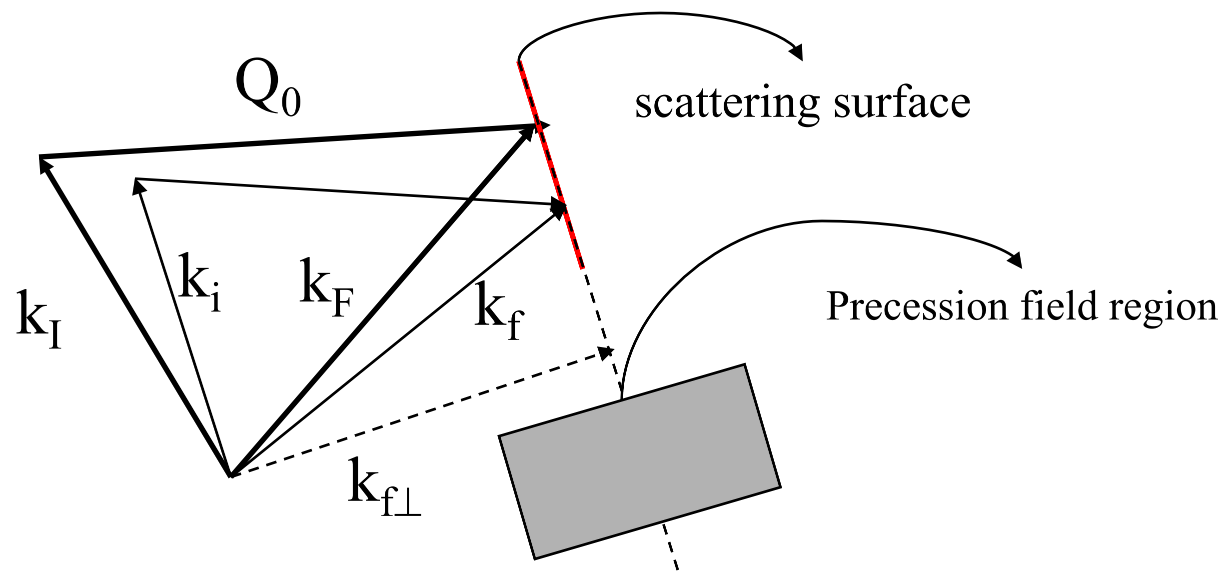
❖The maximum range of the total Larmor phase denotes the capability of the setup to see small lattice splitting.

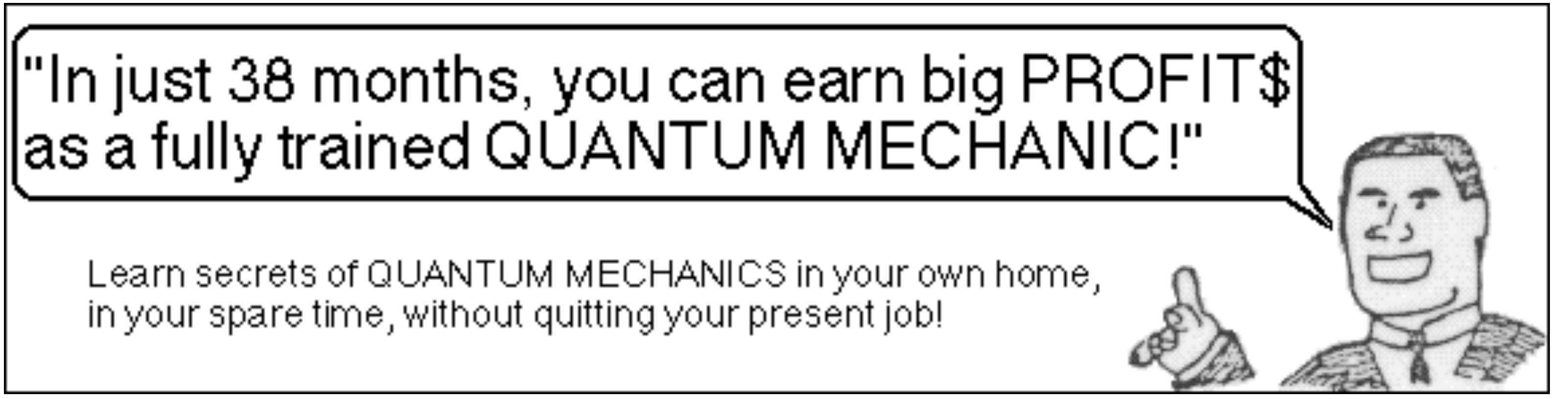
Viewgraph from Fankang Li

"Phonon Focusing"

- For a single incident neutron wavevector, $\boldsymbol{k_I}$, neutrons are scattered to $\boldsymbol{k_F}$ by a phonon of frequency ω_o . The "scattering surface" is the locus in Q space of all phonons of frequency ω_o .

- Provided the edges of the NSE precession field region are parallel to the scattering surface, all neutrons with scattering wavevectors on the scattering surface will have equal spin-echo phase





Thank You

Questions?