Larmor Labelling

Part 1: Basics

Robert Dalgliesh

Summary of Part 1

- Some history
- Polarized Neutrons
- Larmor Precession
- Neutron Spin-Echo (A brief explanation)
- Larmor Angle Encoding(A brief explanation)

History

- As early as 1941 Halpern and Holstein presented a detailed general theory of neutron precession in a magnetic field.
- 1950 – Hahn NMR spin echo
- 1972 – Mezei publishes first paper describing Neutron Spin Echo. (Z. PHYSIK A **255**, (1972), 146-160)
- ILL constructs NSE spectrometers
- 1980 – R. Pynn first discusses the use of spin-echo to encode neutron trajectories.
- Late 1990s protype Larmor Labelling instruments are constructed

Polarisation

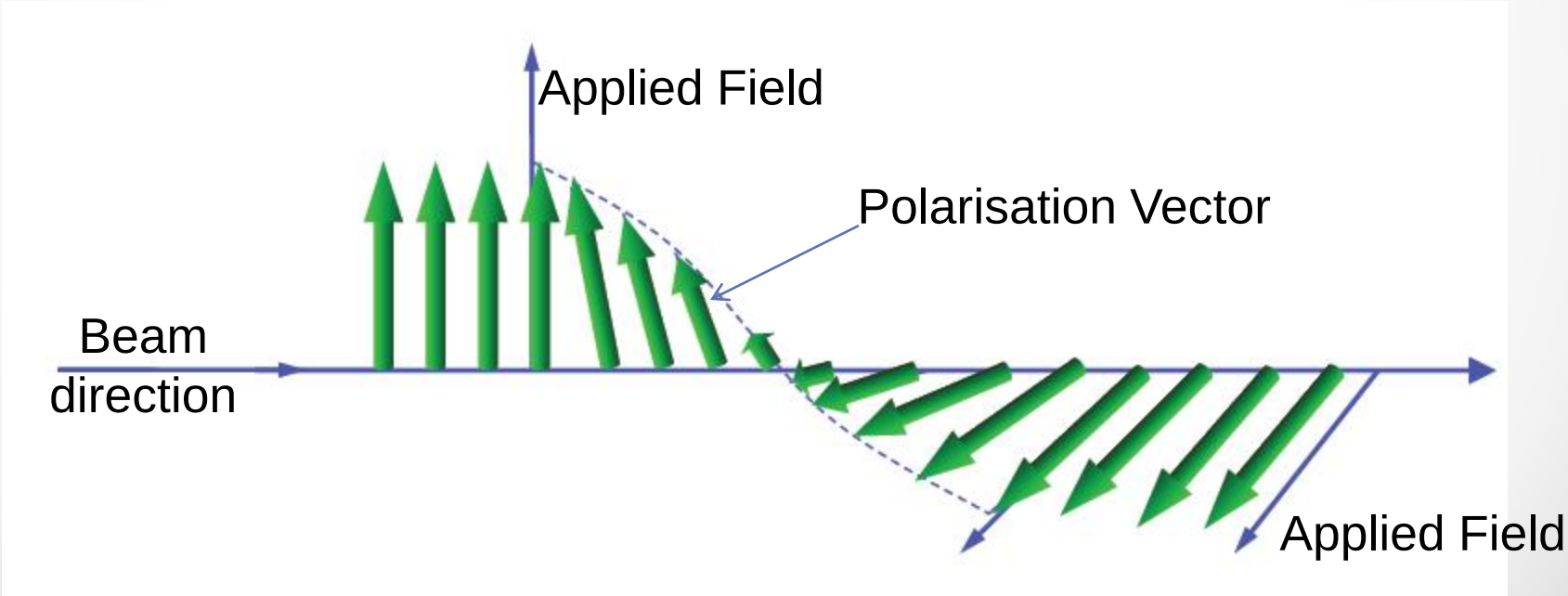
- Neutrons are spin ½ particles
- In an external field the neutrons align parallel (spin up) or anti-parallel (spin down) to the field.
- The polarisation is a scalar that expresses the relative population sizes of the two alignments

$$P(\lambda) = \frac{N_+(\lambda) - N_-(\lambda)}{N_+(\lambda) + N_-(\lambda)}$$

- Polarisation is never perfect and is usually wavelength dependent.

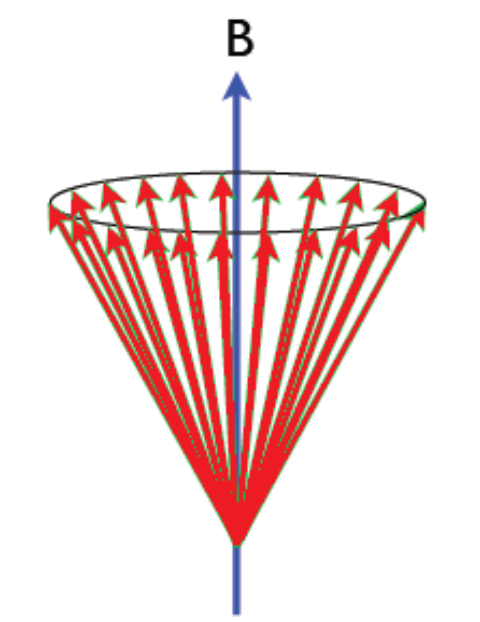
Polarised Neutrons

- Once polarised a beam must be maintained in a field in order to preserve the polarisation.
- Slow changes in applied field allow the polarisation vector to be changed.
- Normally the field and polarisation vector are maintained ~parallel or anti-parallel to each other.



Larmor Precession

- Neutrons precess around an applied field at a frequency determined by the field strength.
- The Larmor precession frequency of the neutron around the applied field vector ω_L is related to the field strength (B) by the ratio of the magnetic moment to the angular momentum γ_n.



$$\omega_L = \gamma_n B$$

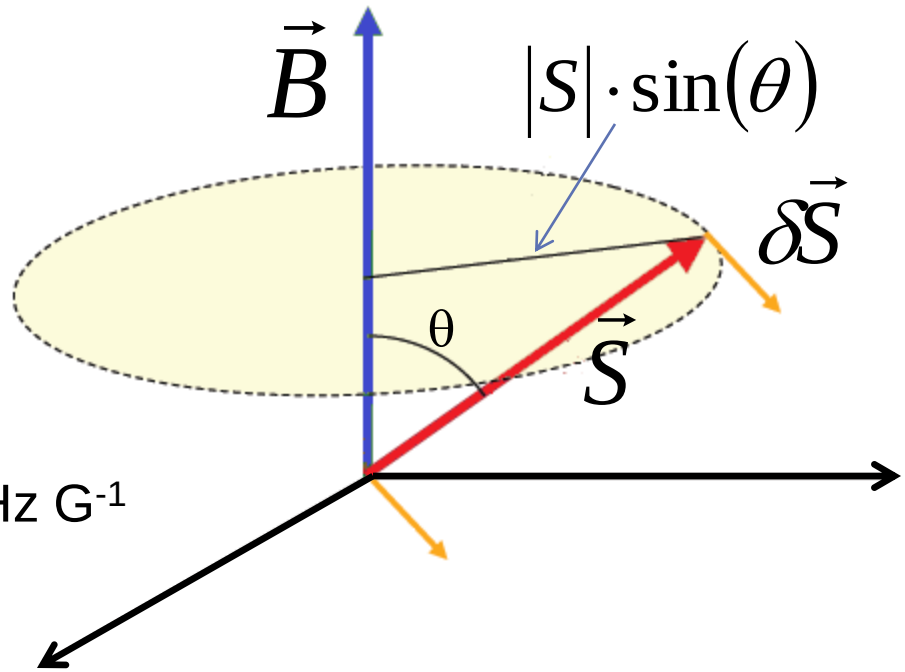
- The total number of rotations around the field can be calculated from the time the neutron spends in the field and is referred to as the Larmor phase

$$\phi(t) = \omega_L t$$

Larmor Precession

$$\frac{\delta\vec{S}}{\delta t} = \gamma\left(\vec{S}\times\vec{B}\right)$$

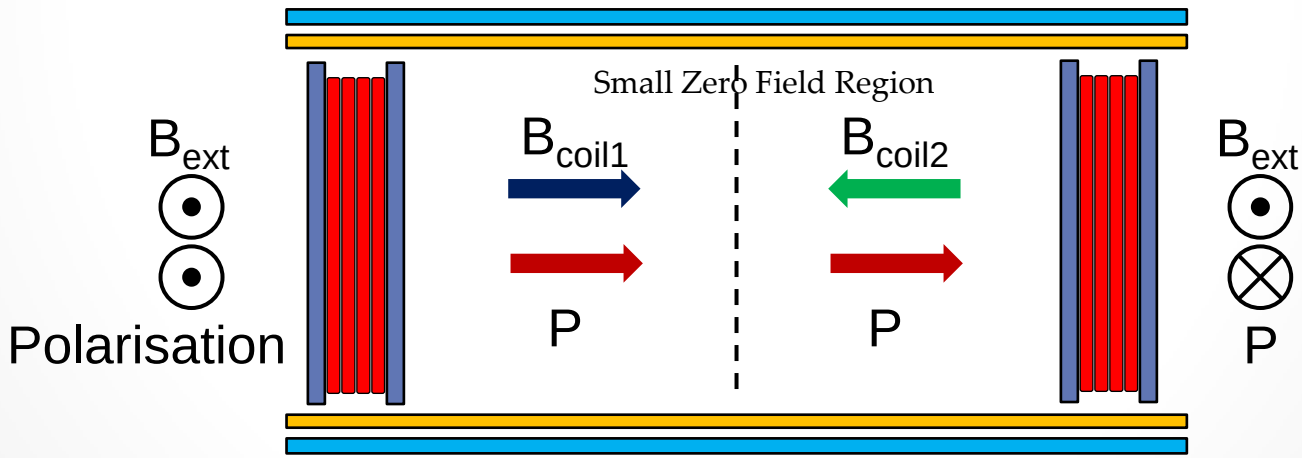
γ, the gyromagnetic ratio = 2.9kHz G^{-1}



- The neutron precesses around the applied field at a rate that is independent of the angle θ.
- This is crucial for inelastic spin-echo and Larmor labelling

Spin Flippers

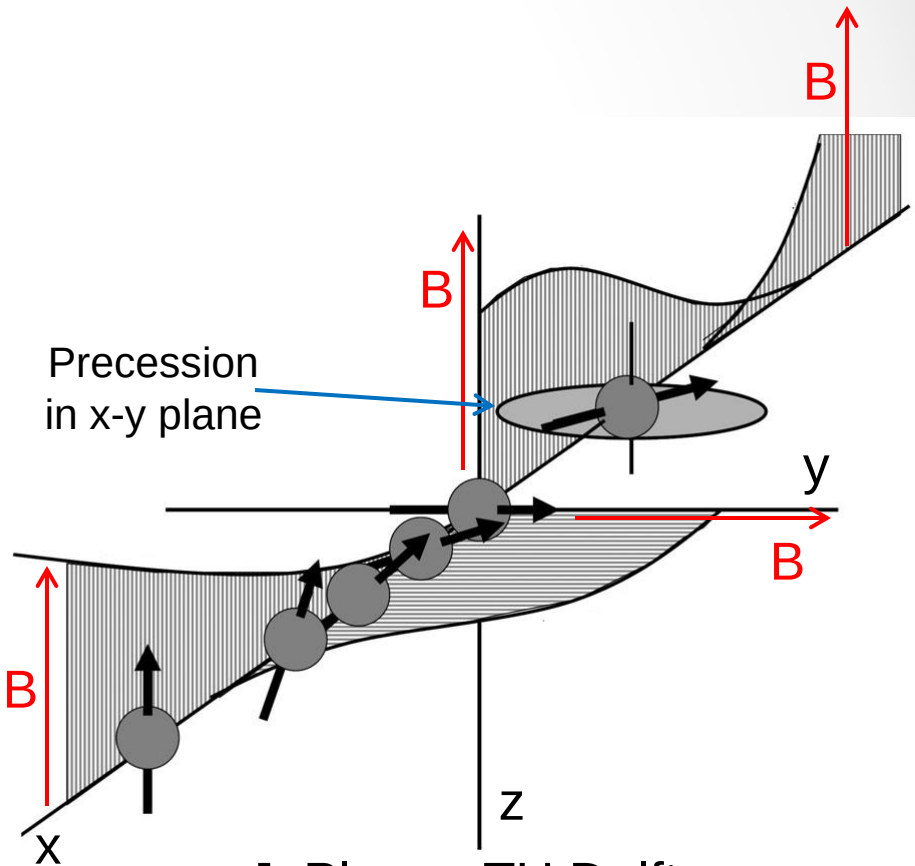
- Many different designs are available, some only work with monochromatic beams.
- Some spin flippers operate by changing the direction of the applied field so rapidly that the neutron does not have a chance to precess and follow the change.
- Relatively straight forward for 180° (π) flip.



•Drabkin Flipper

π/2 Flipper example V-coil design

- Allows a neutron to precess in the plane perpendicular to the field vector. (x-y plane illustrated)
- External field is rotated slowly up to a pair of current screens at 90° to each other.
- Current screen thickness ~0.5mm.
- ~1mm gap between the screens where the fields are ill defined.
- Rapid transition back to a vertical field.



J. Plomp, TU Delft

Neutron Spin Echo or Larmor Labelling

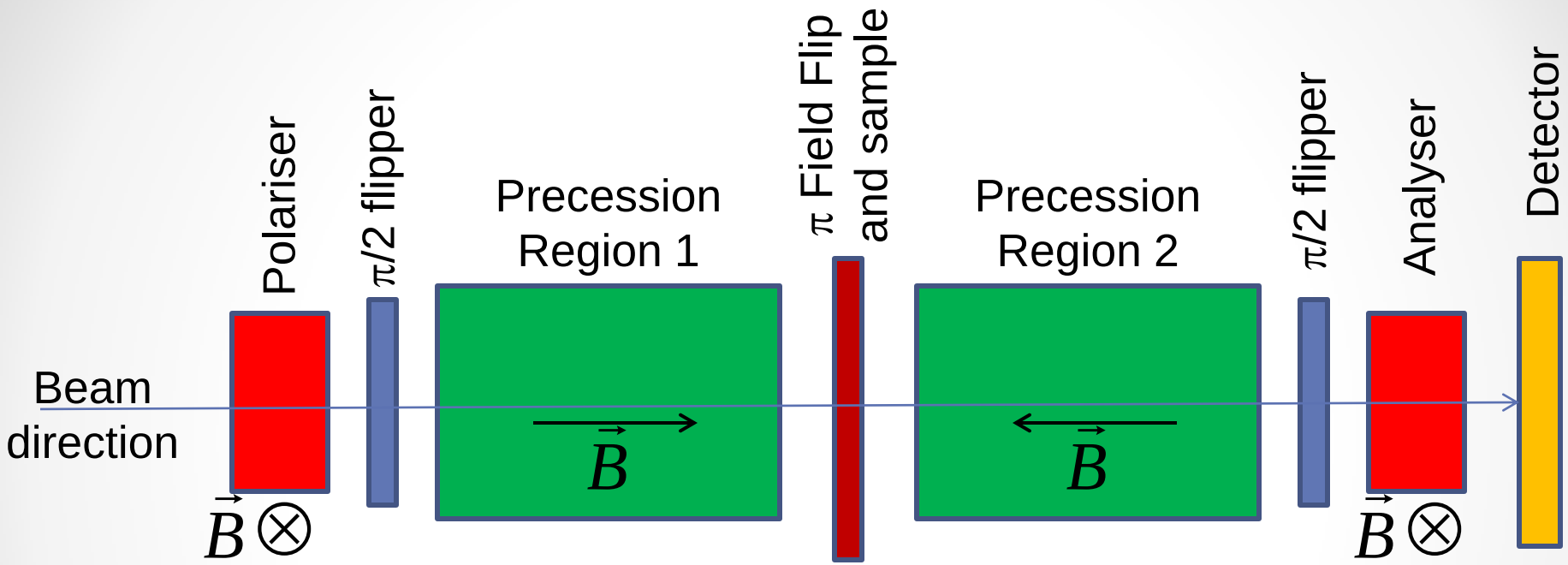
- Through the careful manipulation of the neutron spin neutron spin-echo or Larmor labelling provides a mechanism that allows measurements of either small changes in neutron energy or small angular deflections.
- The measurements break the requirement to strongly collimate the beam in order to achieve high resolution.

Neutron Spin Echo

To avoid confusion.

- Neutron spin echo will be used to refer to the use of Larmor precession to measure inelastic scattering processes.
- Larmor labelling will be used to refer to the use of Larmor precession to label the path that a neutron takes through an instrument.
- However, both terms are used interchangeably.

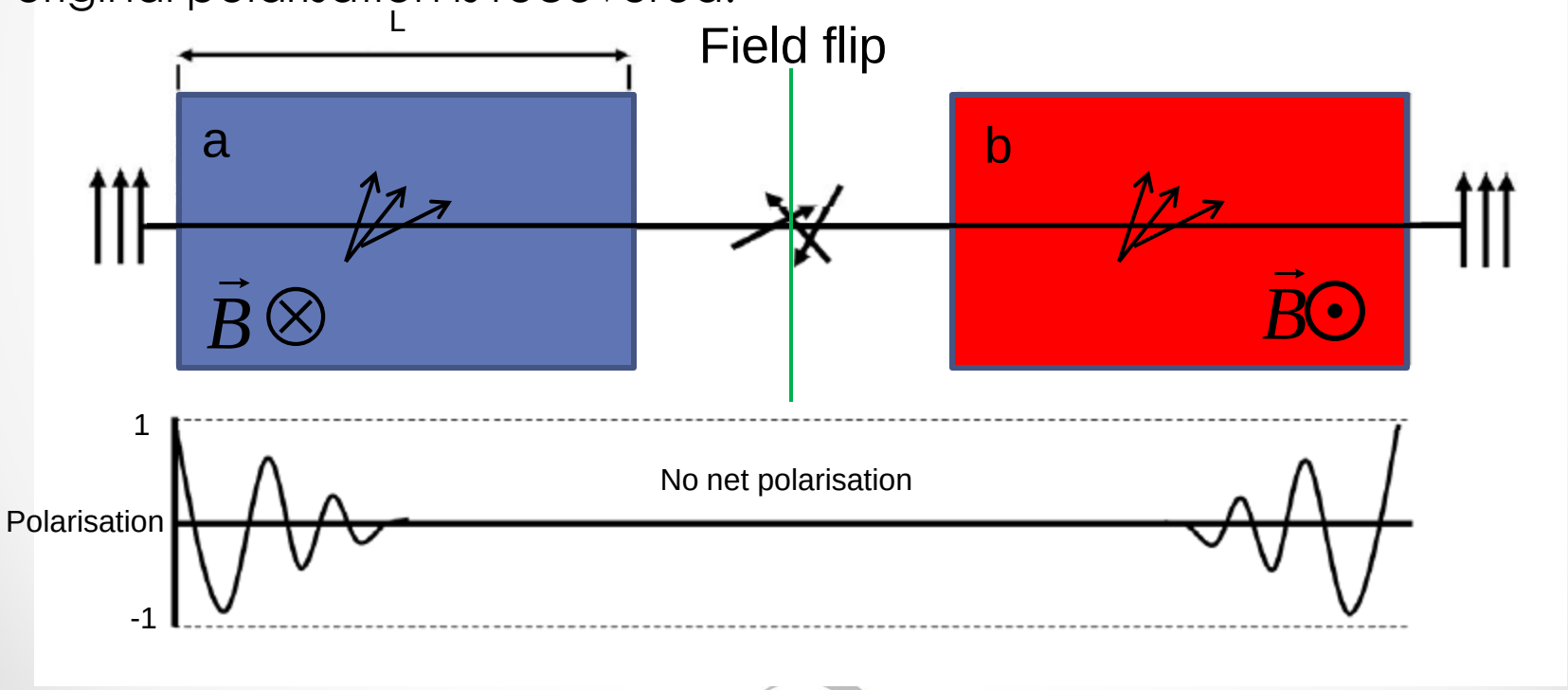
NSE Layout



- 2 precession field regions provided by large solenoids.
- Field regions must provide an extremely homogeneous field.
- Correction elements are always required.

NSE Principal (Classical View)

- A polarised beam with relaxed wavelength resolution travels through a precession region a.
- The different wavelengths precess at varying frequencies
- Between the two coils an abrupt 180° field flip reverses the precession direction
- If the instrument is tuned the precessions are unwound in b and the original polarisation is recovered.



NSE Principal (Classical View)

- Sample is placed as close as possible to the field flip position.
- If scattering from the sample is purely elastic and the correction coils have done their job then the measured polarisation is not perturbed.
- If inelastic scattering occurs the neutrons change velocity by a small amount.
- This change in velocity can be related to a Larmor phase ϕ. τ is referred to as the spin-echo time.

$$\phi_{tot} = \gamma_L BL \left(\frac{1}{v_0} - \frac{1}{v_0 + \frac{\hbar\omega}{m v_0}} \right) \approx \frac{\gamma_L BL\hbar}{m v_0^3} \omega = \tau\omega$$

NSE Principal (Classical View)

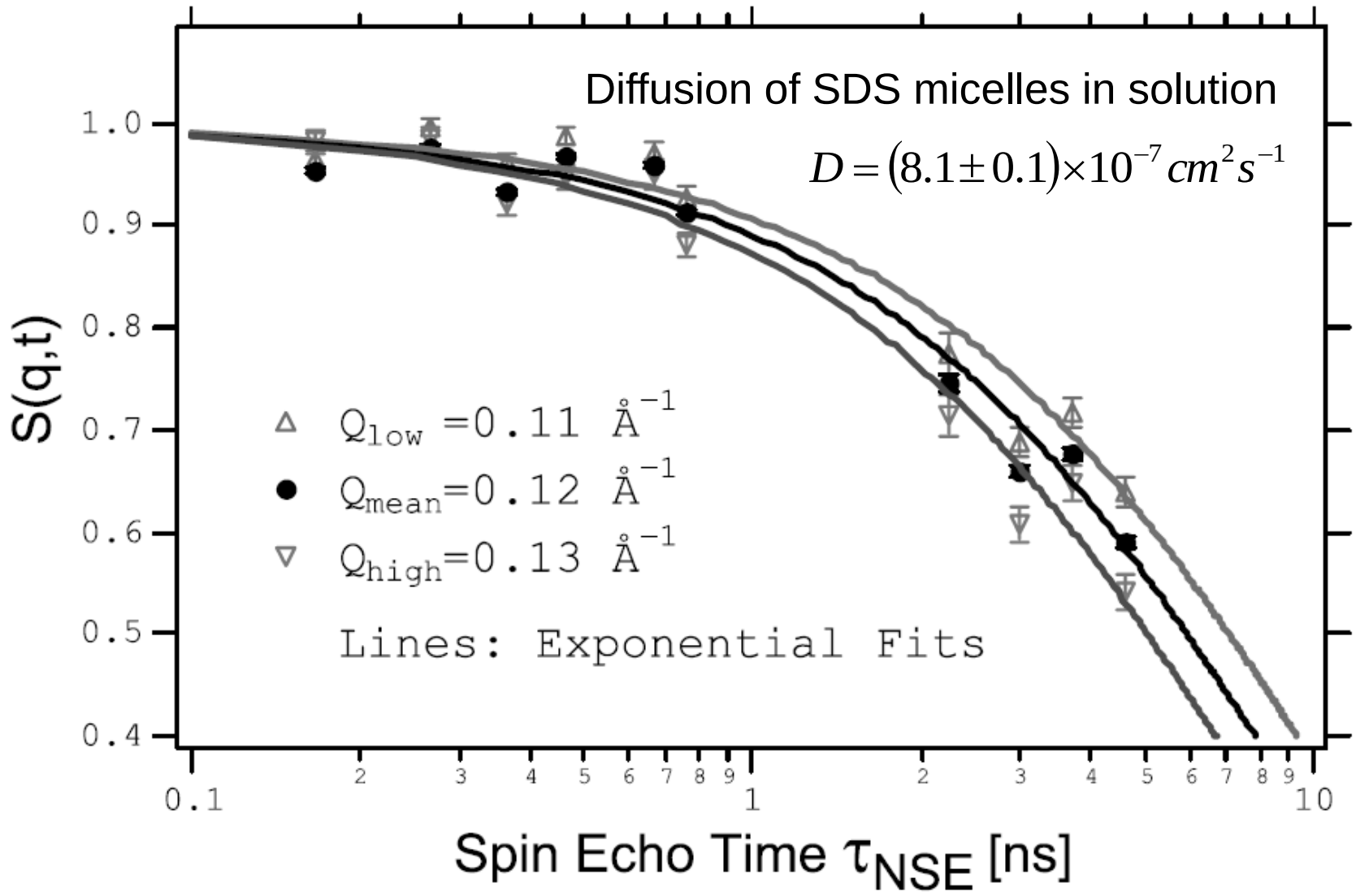
- The spin echo time may also be expressed as a function of neutron wavelength.

$$\tau_{NSE} = \frac{\gamma_L BL\hbar}{m v_0^3} = \frac{m^2 \gamma_L BL \lambda^3}{2\pi h^2}$$

- Note the λ^3 dependence, this explains the large range of dynamics that can be probed (ps-μs)
- By measuring the Larmor phase as a function of both q and spin-echo time the intermediate scattering function (F(q,τ)) is measured. This is related to S(q,ω) by a Fourier Transform.

$$\left\langle \cos\left(\varphi_{L\,tot}\right)\right\rangle = \frac{\int \cos(\tau\omega) S(\vec{q},\omega)\, d\omega}{\int S(\vec{q},\omega)\, d\omega} = F(\vec{q},\tau)$$

W. Häussler et al. / Chemical Physics 292 (2003) 501–510



Further Details

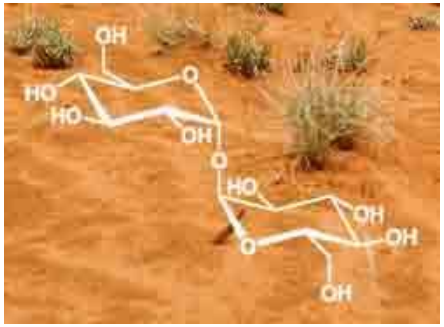
- More detailed explanations and discussion can be found in numerous places.
- http://www.indiana.edu/~neutron/ (Roger Pynn's Lectures)
- http://www.oxfordneutronschool.org/2011/lectures/teaching%20materials_2011.htm (C. Pappas's Lecture)
- Neutron Spin Echo Spectroscopy Basics, trends and applications, F. Mezei, C. Pappas, T. Gutberlet (Eds.), Springer Verlag Lecture notes in Physics Series

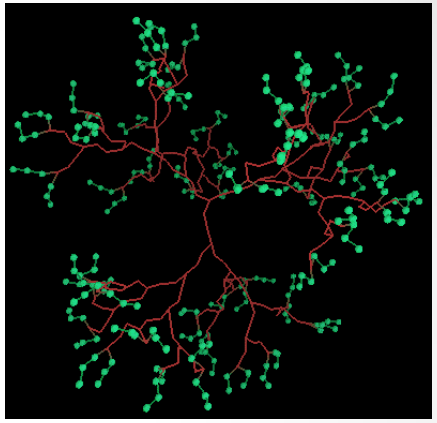
NSE Science

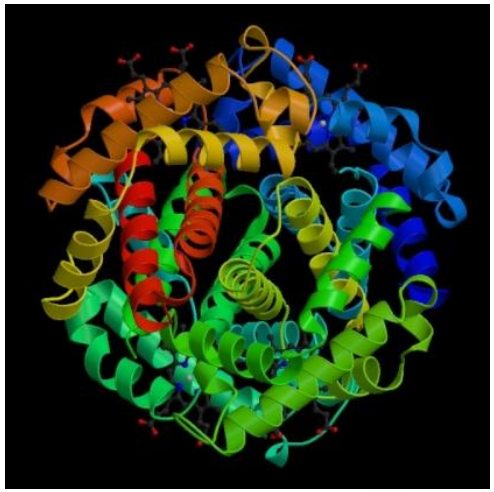
Broad range of macro scale dynamics may be studied

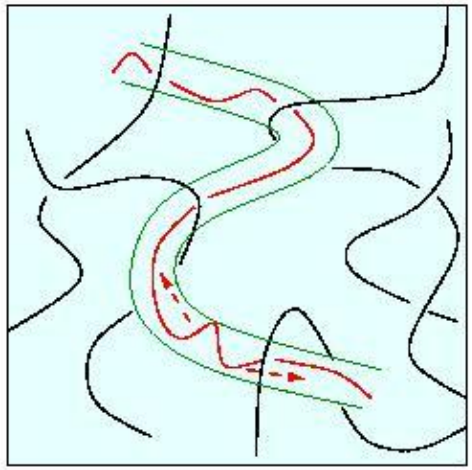
- Collective diffusion of aggregate structures e.g. Micelles
- Polymer dynamics such as reptation
- Biomolecular dynamics

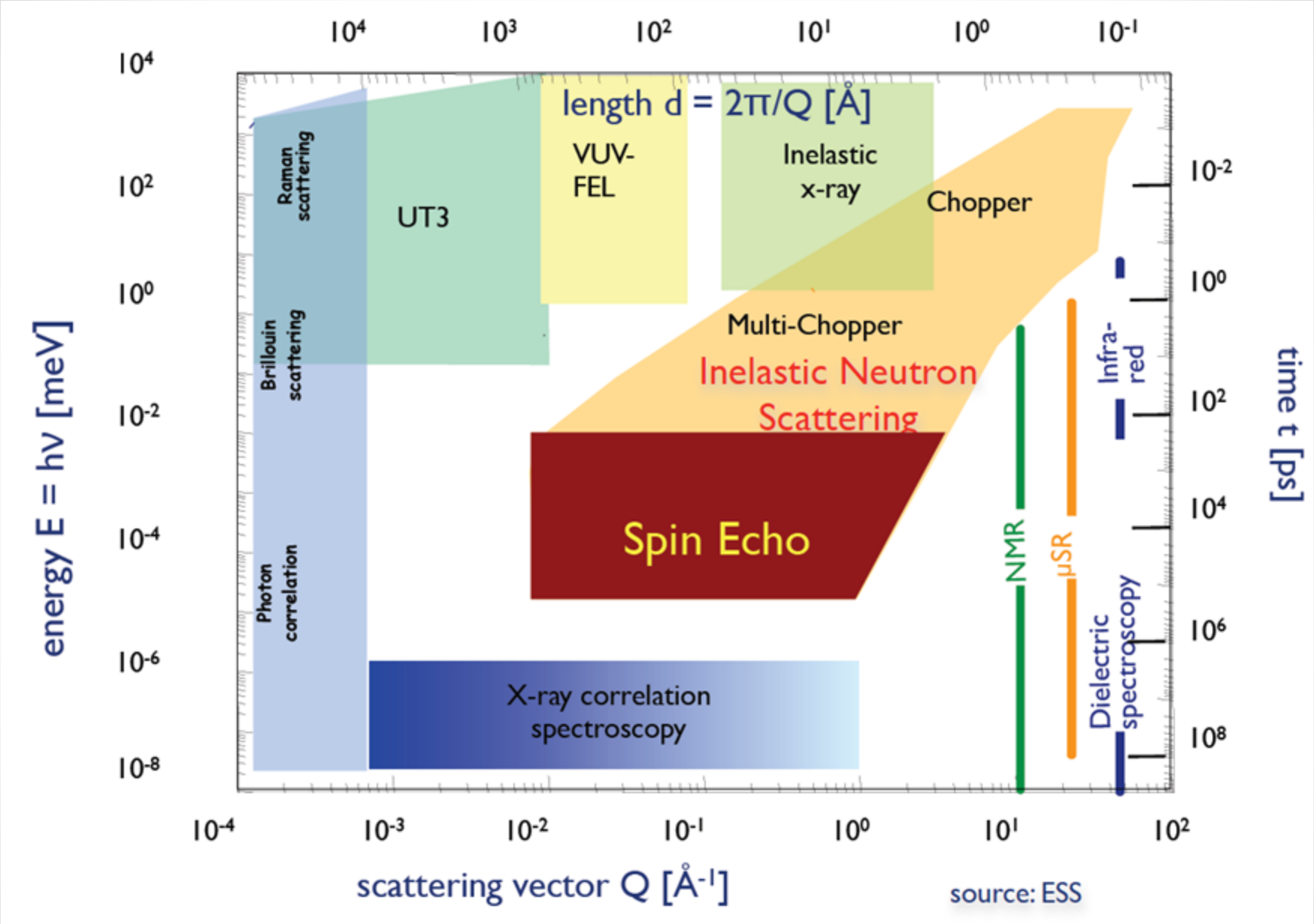
Through additional options magnetism and phonon dispersions may also be measured.





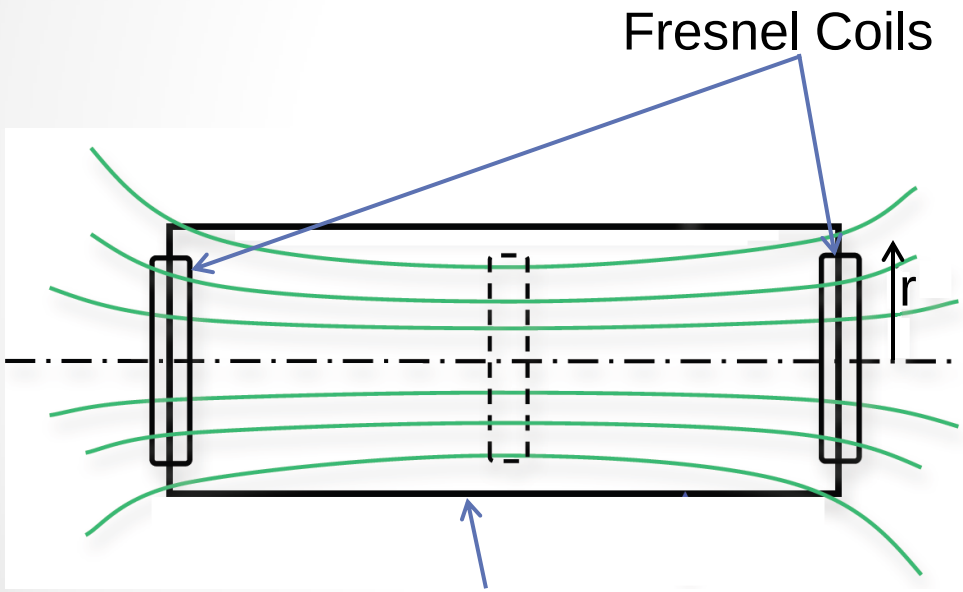


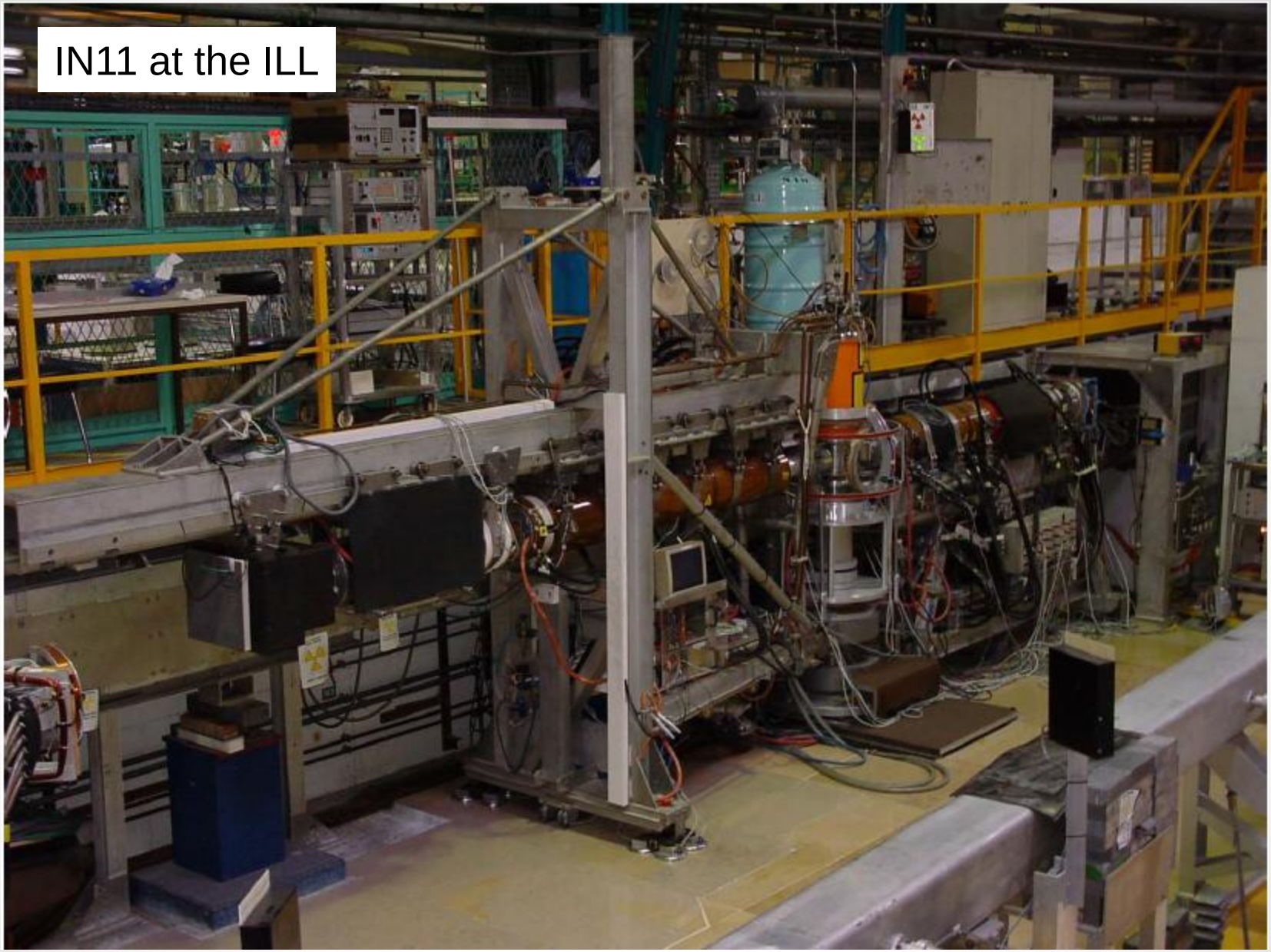



length d = 2π/Q [Å]
VUV-FEL
Inelastic x-ray
Chopper
UT3
Raman scattering
Brillouin scattering
Photon correlation
Multi-Chopper
Inelastic Neutron Scattering
Spin Echo
NMR
μSR
Infra-red
Dielectric spectroscopy
X-ray correlation spectroscopy
energy E = hν [meV]
time t [ps]
scattering vector Q [Å⁻¹]
source: ESS

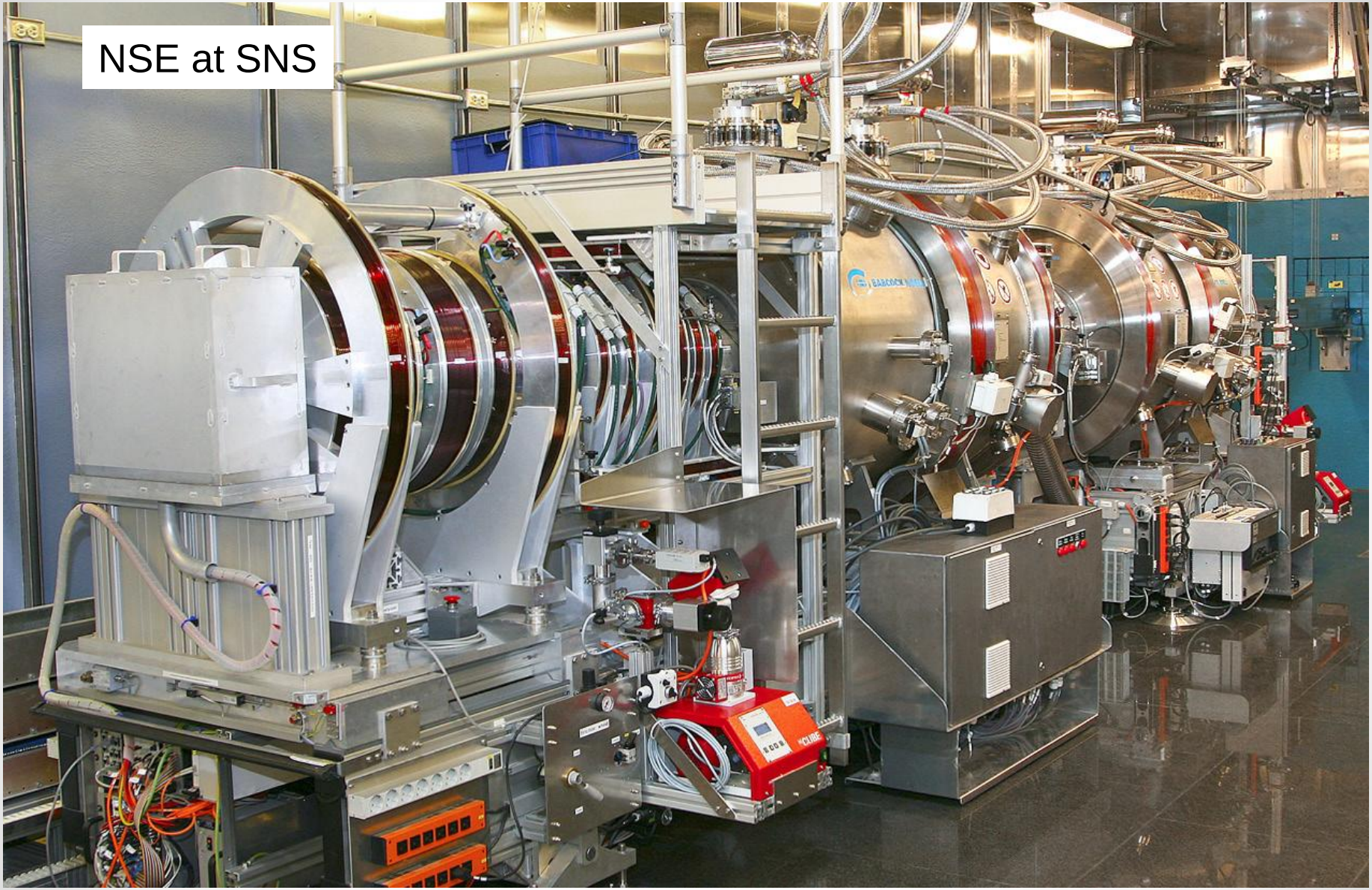
Correction Elements

- Solenoid field weakens at the limit of the device.
- Fresnel correction coils are often used.
  - Current Density varies as r^2
  - Large amounts of material in the beam as very high currents are required



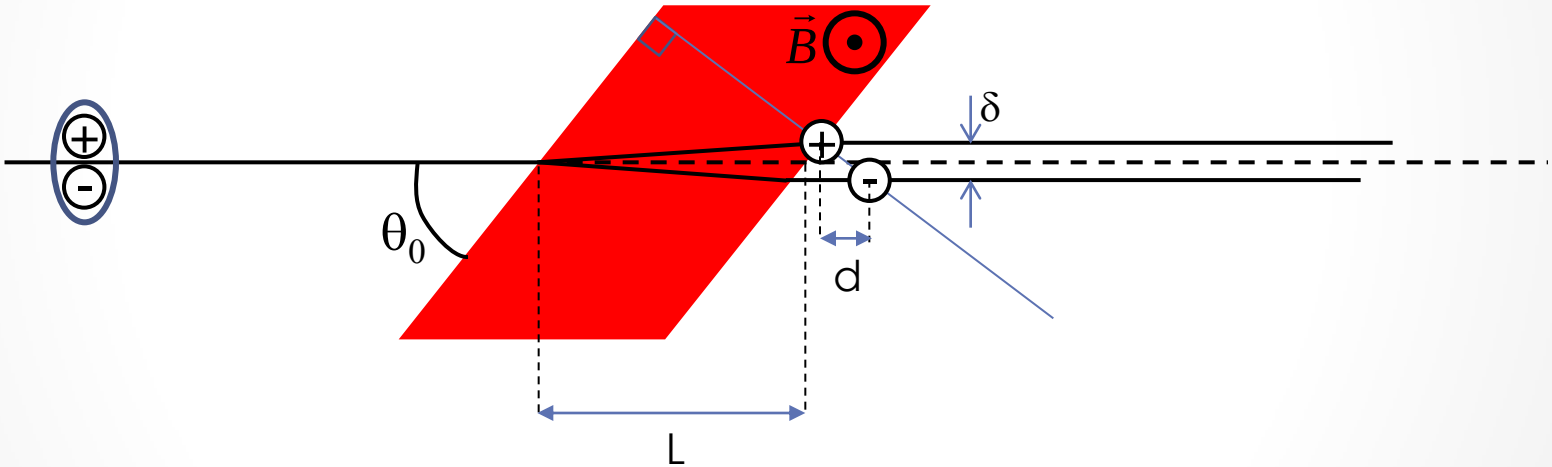

IN11 at the ILL

NSE at SNS



Angle Encoding (Semi-Classical View)

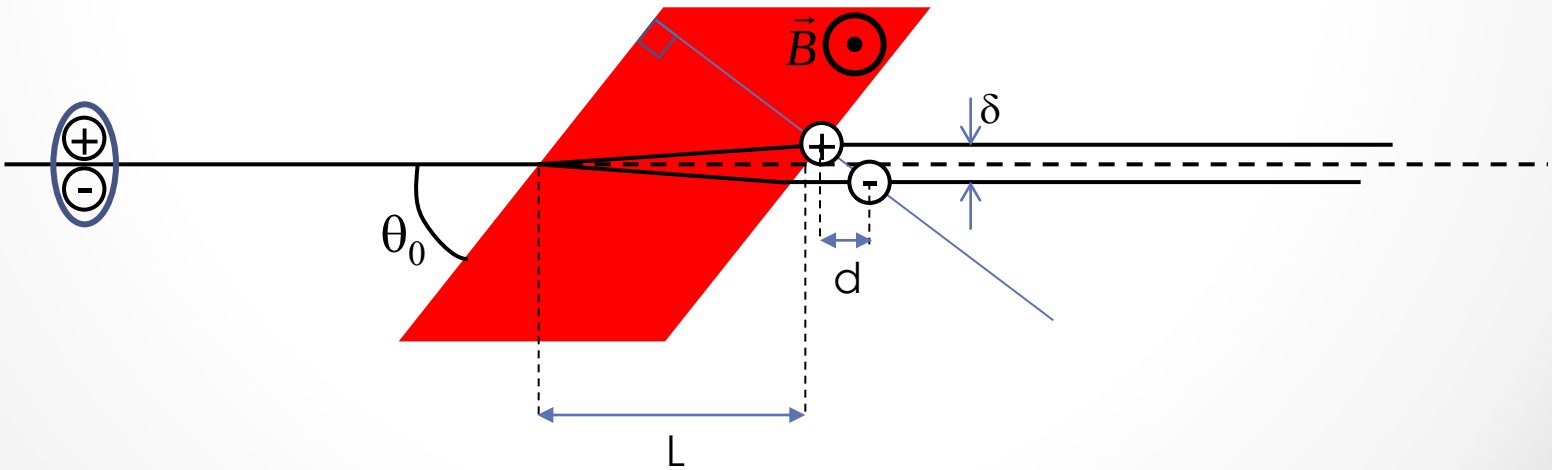
- If the simple solenoid is replaced with a field region with an inclined faces the path of the neutron through an instrument can be encoded. (Pynn 1980)
  - Semi-Classical view After Gahler et. al. (Gahler R, *Physica B 1996;**229:1-17***)



- Consider the precessing neutron as a superposition of the up and down spin states.

Angle Encoding (Semi-Classical View)

- On entering the field region where B<0
  - Spin up state looses energy
  - Spin down state gains energy
- Hence, the kinetic energy of the two states are shifted by $\Delta E = \pm \mu_n B$ and the relative velocities change.
- In an analogous way to light passing through a prism the two states are refracted differently within the field region and become shifted with respect to each other in space in two directions.



Angle Encoding (Semi-Classical View)

- The shift along the beam path corresponds to a spin-echo time τ which is the same as that described earlier.

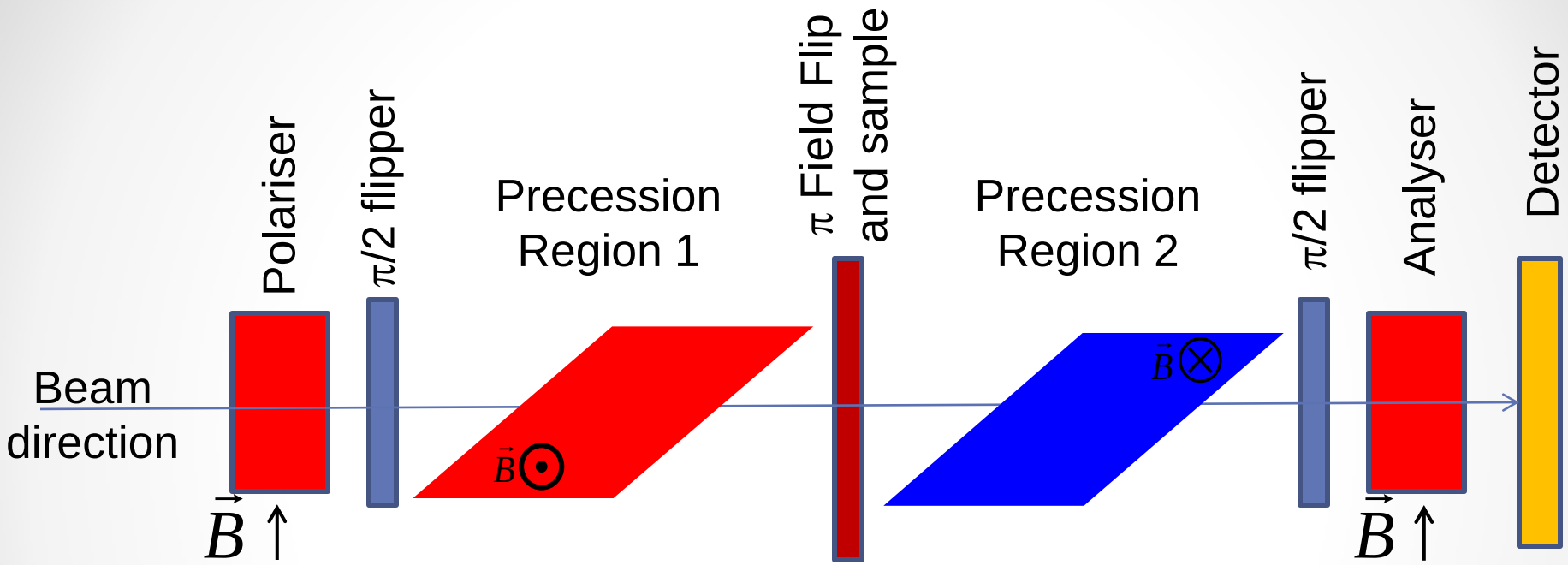
$$\tau = \frac{d}{v} = d\frac{m\lambda}{h} = \frac{m^2\gamma_L BL\lambda^3}{2\pi h^2}$$

- The shift perpendicular to the beam path is called the spin-echo length.

$$\delta = \frac{m\gamma_L BL\lambda^2 \cot\theta_0}{2\pi h}$$

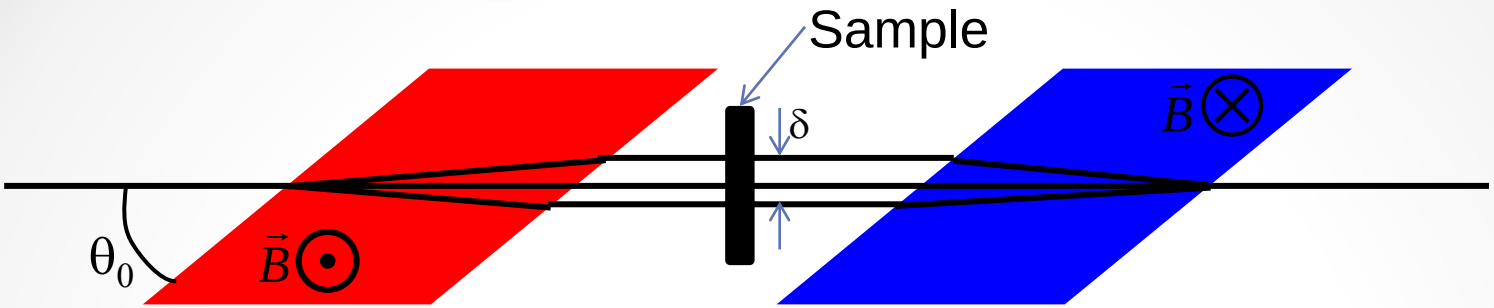
- It is clear from these two equations that a number of options exist for probing either spin-echo length or time. B,L,θ_0 or λ may all be varied and the choice will typically depend on the neutron source

Spin-Echo SANS Layout



- 2 tilted precession field regions
- Field regions must provide an homogeneous field.
- Manufacture the precession regions to cancel aberrations as correction coils are not easily possible.

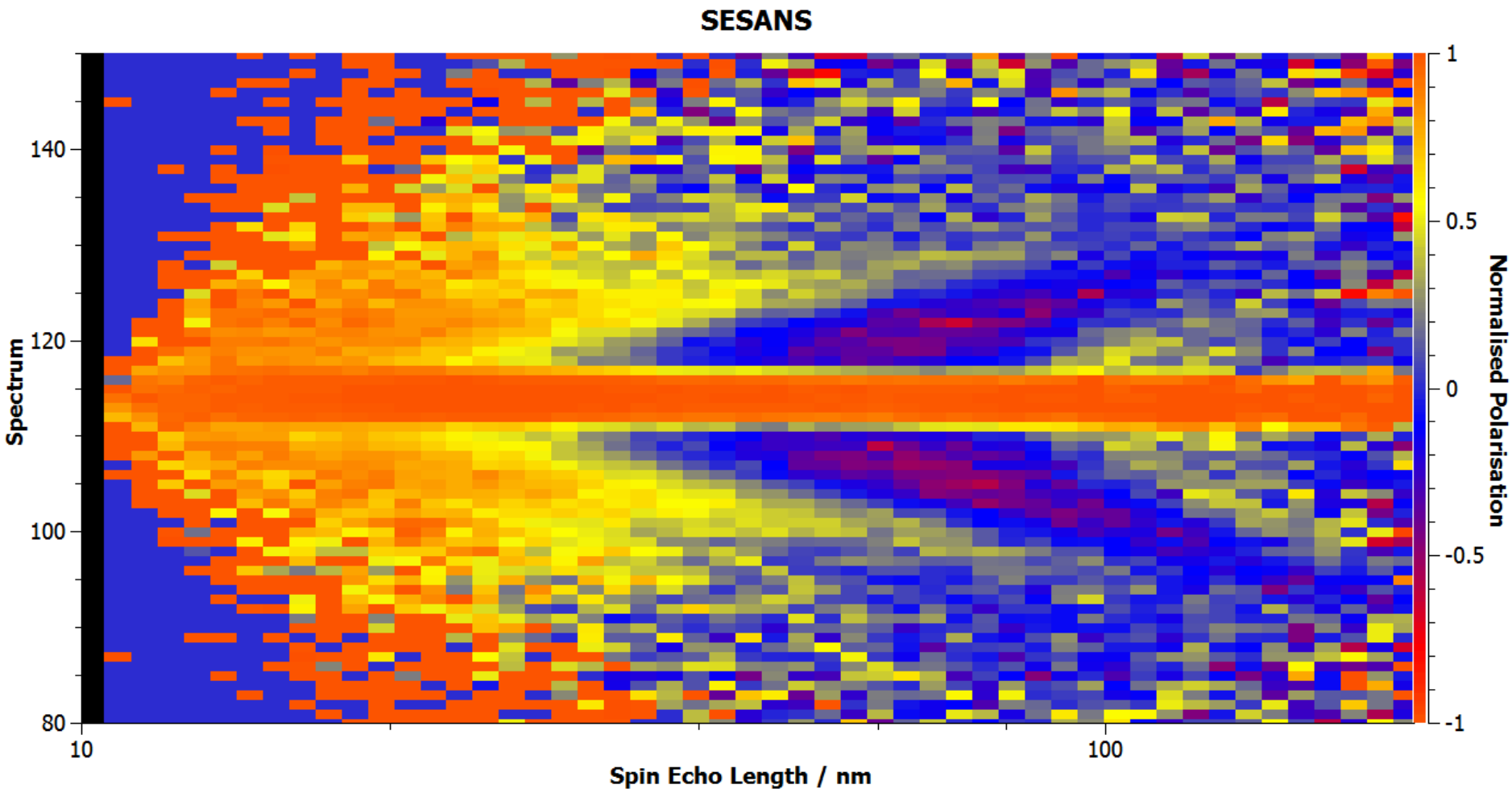
Spin-Echo SANS



- Use spin echo measure small deflections of the neutron beam normally ignored in SANS as they fall within the beamstop.
- Corresponds to larger length scales than normally measured in SANS (50nm-50μm)
- Measure a real space correlation function integrated over the measurable q range.

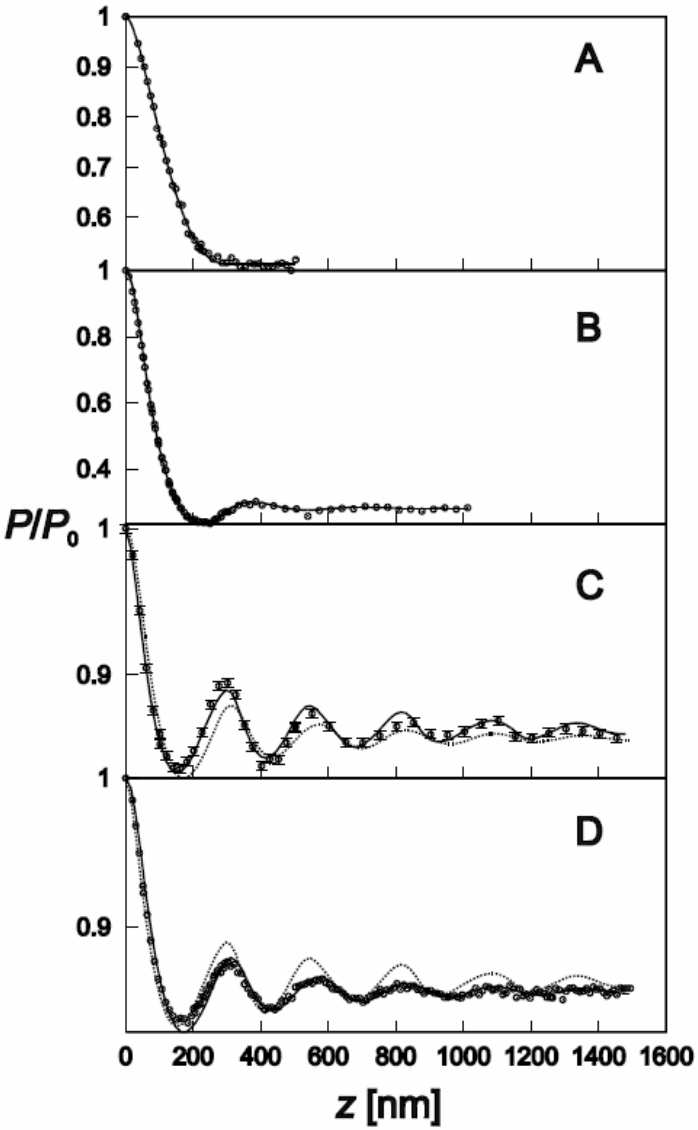
$$\frac{P}{P_0} = \frac{\int_{-\infty}^{\infty} S(q)\cos(q\delta)dq}{\int_{-\infty}^{\infty} S(q)dq}$$

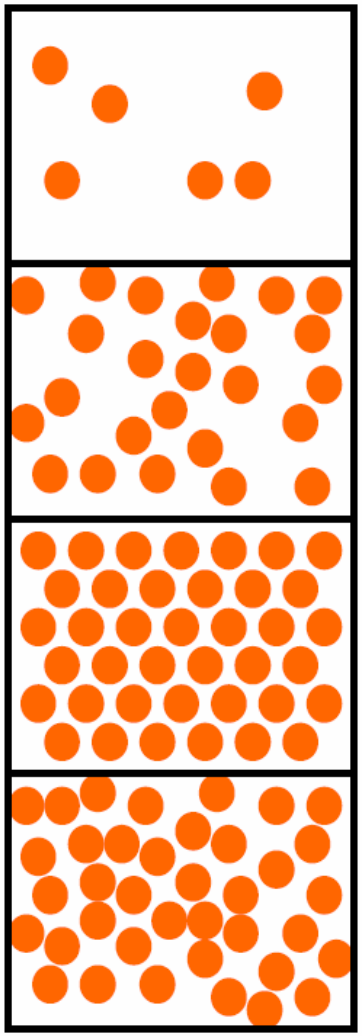
Spin-echo single before integration on a linear detector



Wim Bouwman
http://www.tnw.tudelft.nl/en/about-faculty/departments/radiation-radionuclides-reactors/research/research-groups/npm2/research-projects/sesans/

Colloidal phase transitions as function of concentration





gas

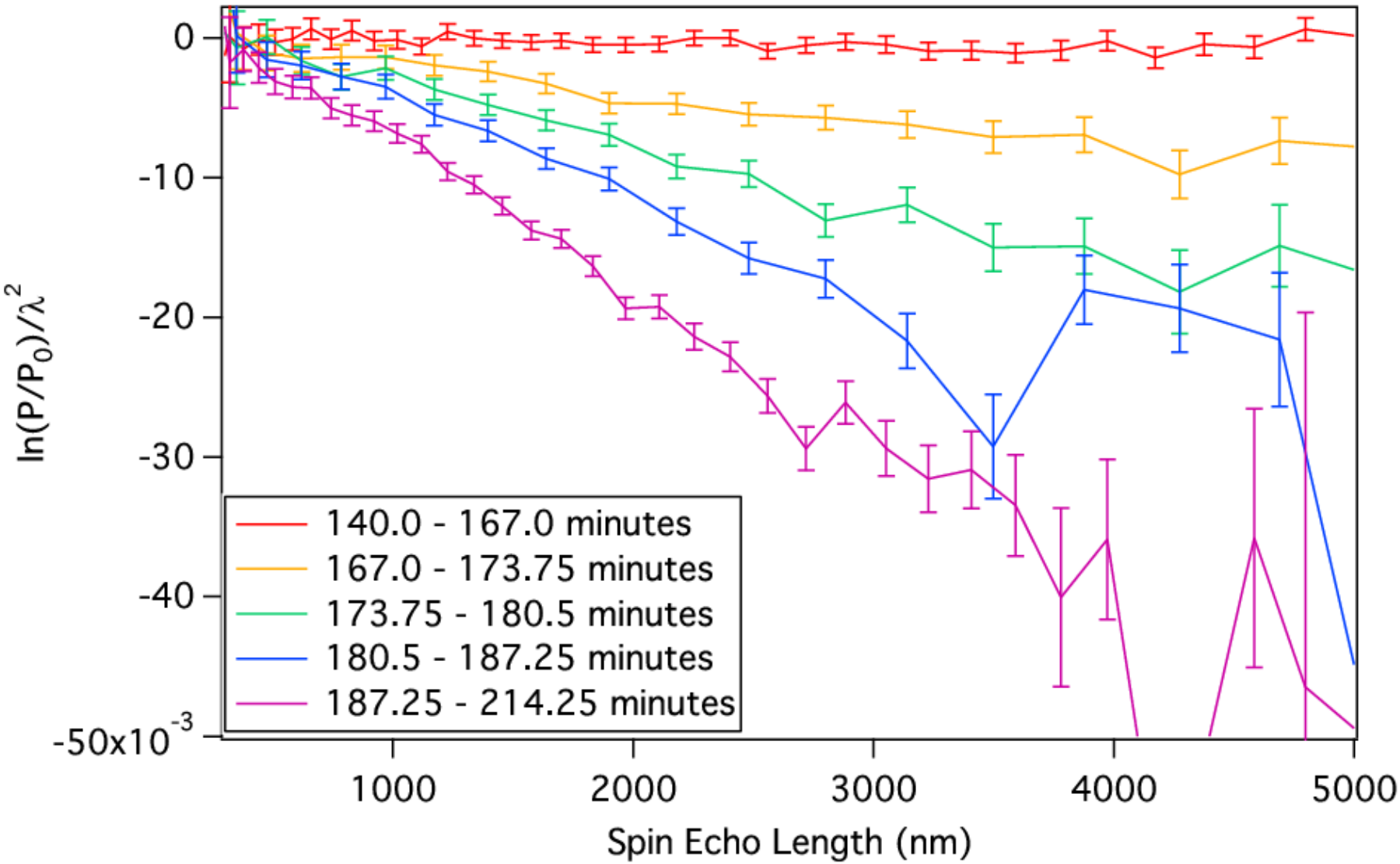
liquid

crystal

glass

TU Delft

SESANS from surfactant aggregation



Further Reading

- Analysis of spin-echo small-angle neutron scattering

  R. Andersson, L.F. van Heijkamp, I.M. de Schepper, W.G. Bouwman

  J. Appl. Cryst.. 41, 868-885 (2008)

  http://dx.doi.org/10.1107/S0021889808026770
- Real-space neutron scattering methods

  W.G. Bouwman, J. Plomp, V.O. de Haan, W.H. Kraan, A.A. van Well, K. Habicht, T. Keller, M.T. Rekveldt

  Nuclear Instruments and Methods in Physics Research A 586 9–14 (2008)

  http://dx.doi.org/10.1016/j.nima.2007.11.045
- Structural transitions of hard-sphere colloids studied by spin-echo small-angle neutron scattering

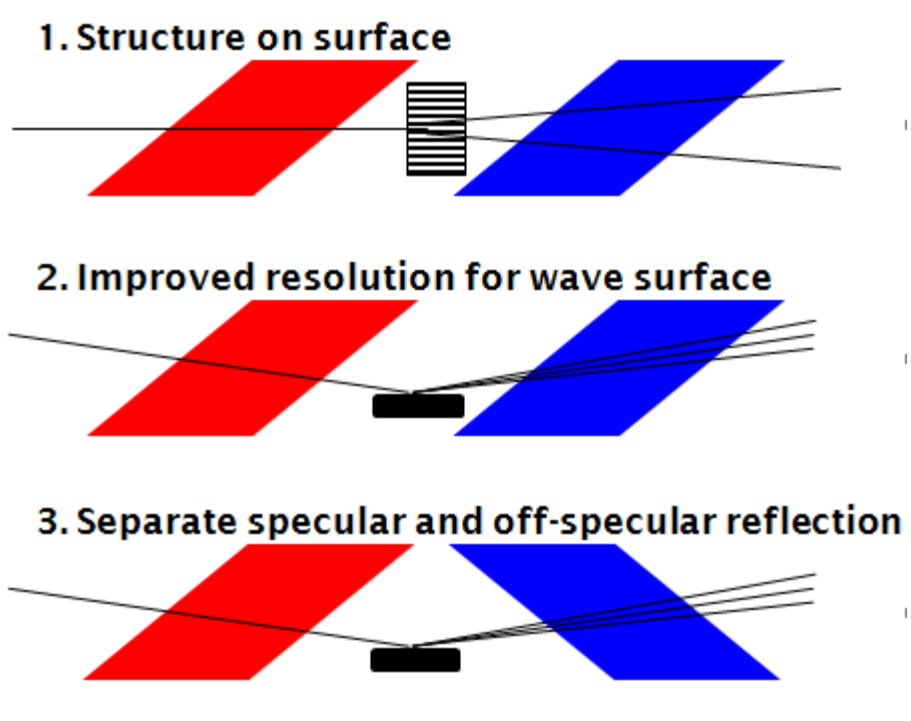
  T.Krouglov, W.G. Bouwman, J. Plomp, M.Th. Rekveldt, G.J. Vroege, A.V. Petukhov and D.M.E. Thies-Weesie

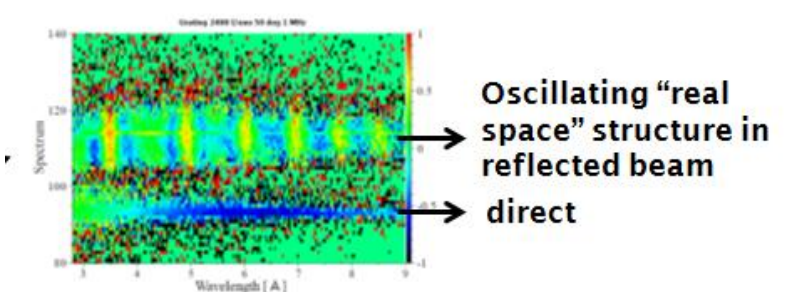
  J. Appl. Cryst. 36, 1417-1423 (2003)

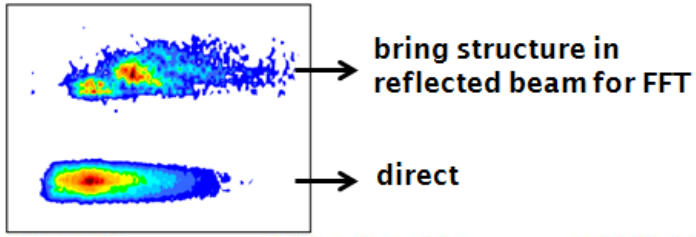
  http://dx.doi.org/10.1107/S0021889803021216

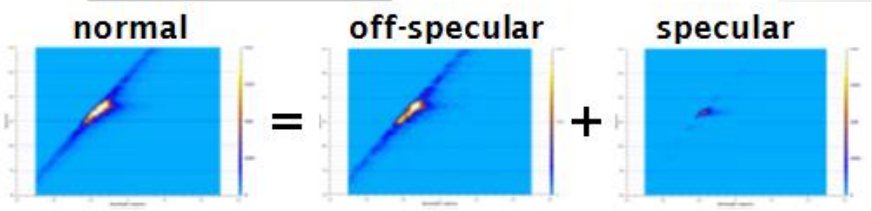
Further Angle Encoding options

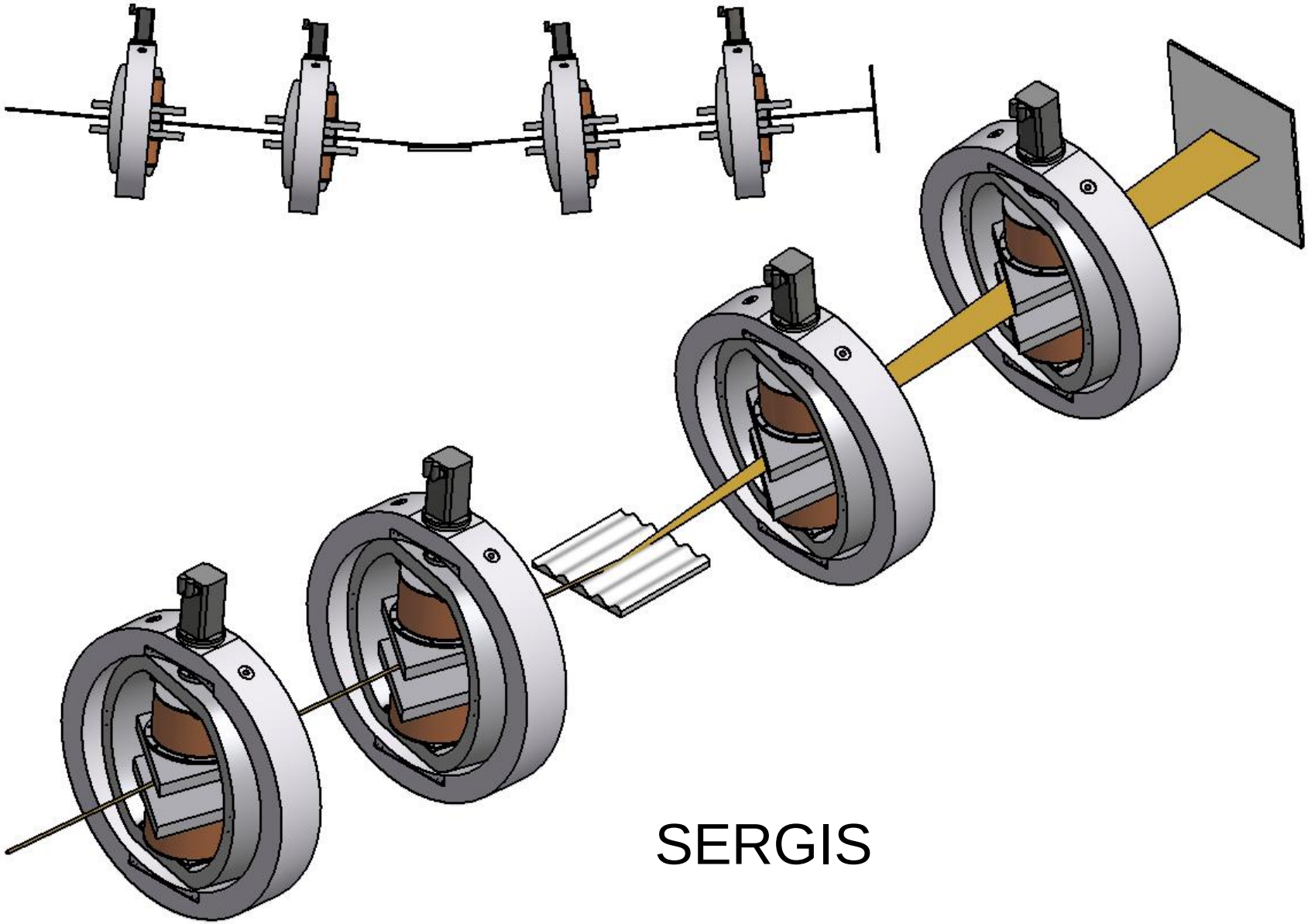
- Applying the same field geometry to surface scattering allows in plane structure to be probed.









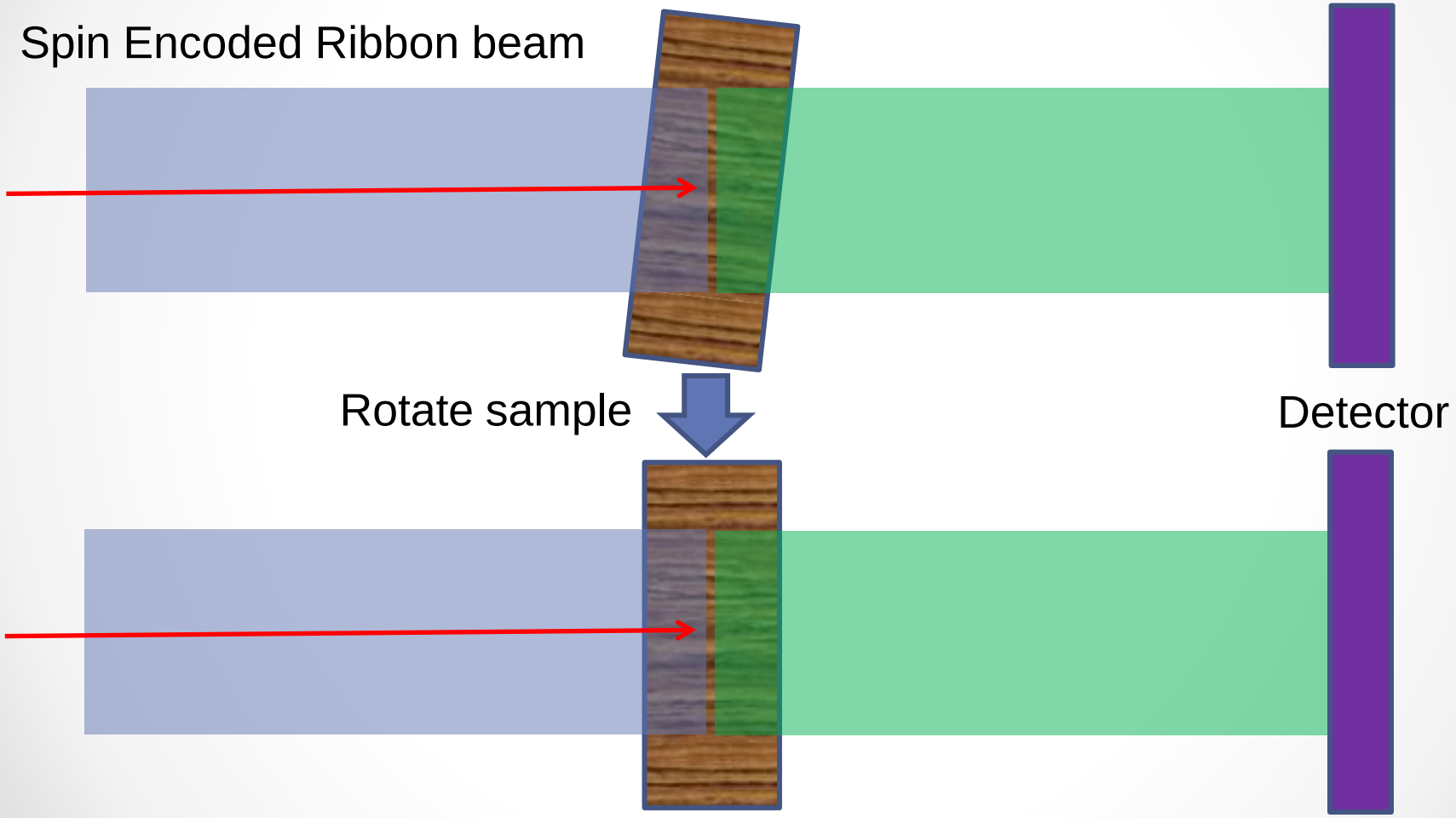


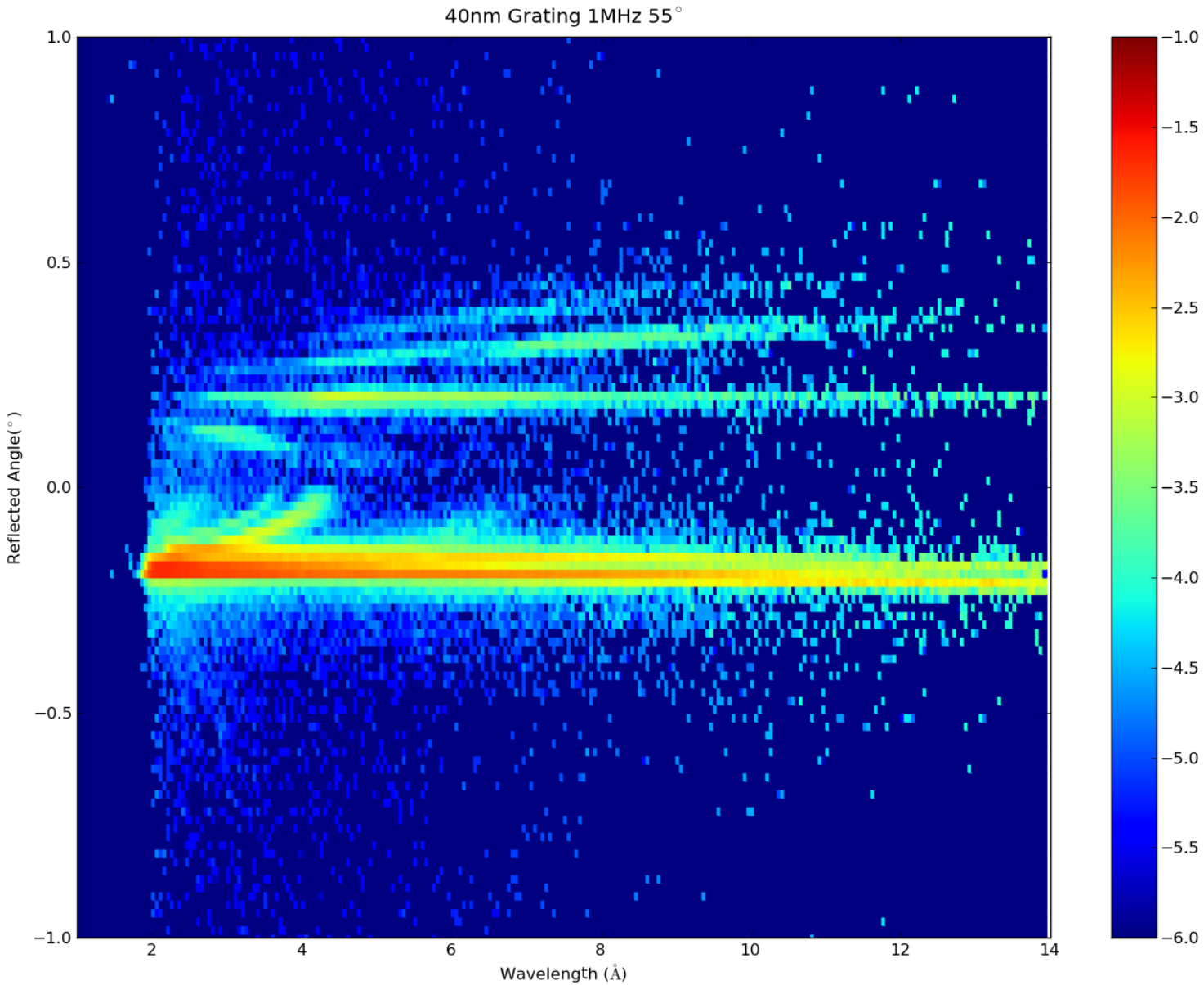
SERGIS

Spin Echo resolved grazing incidence scattering

Reflection from a grating

Grating Sample Rotated off beam axis




40nm Grating 1MHz 55°
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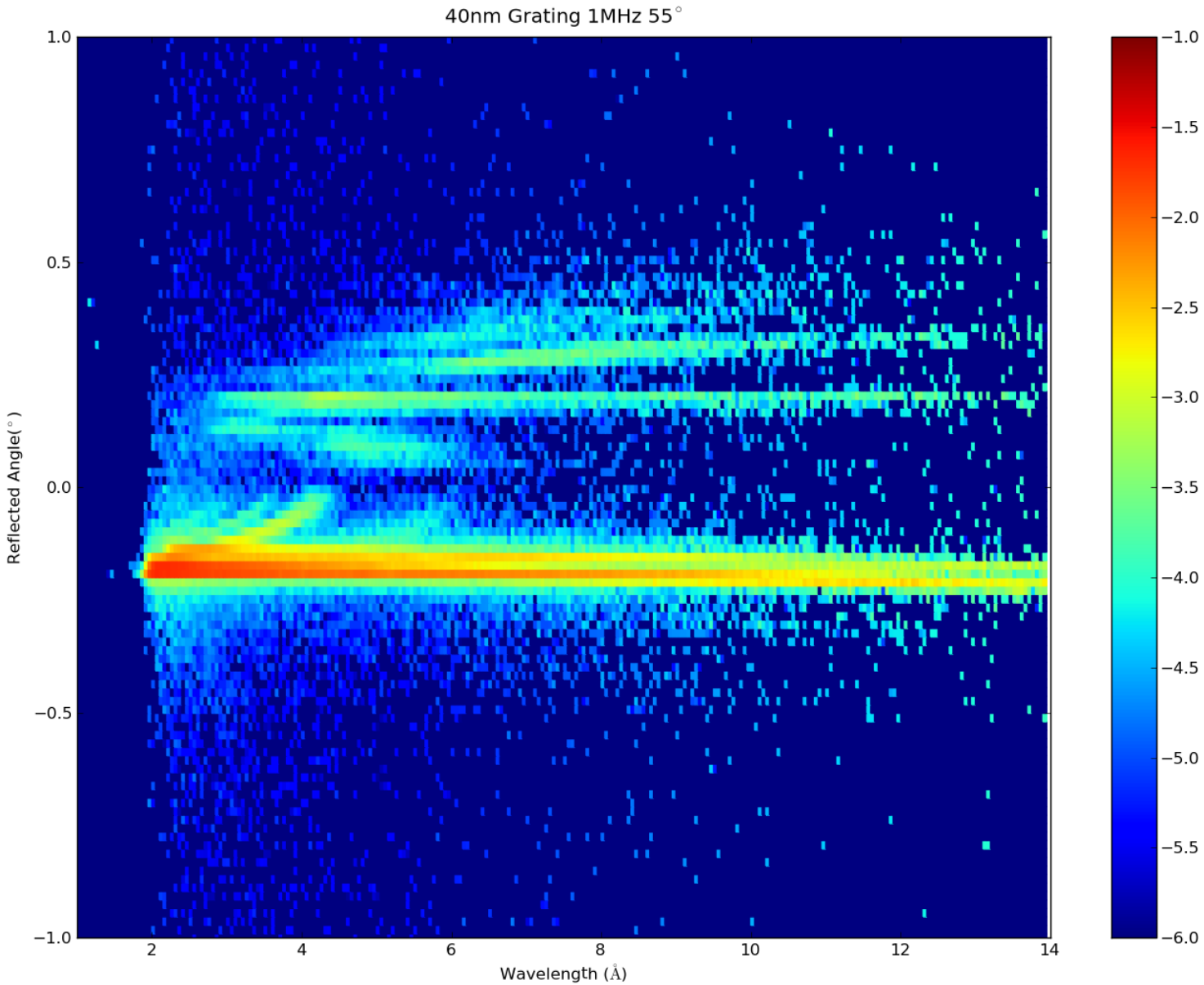
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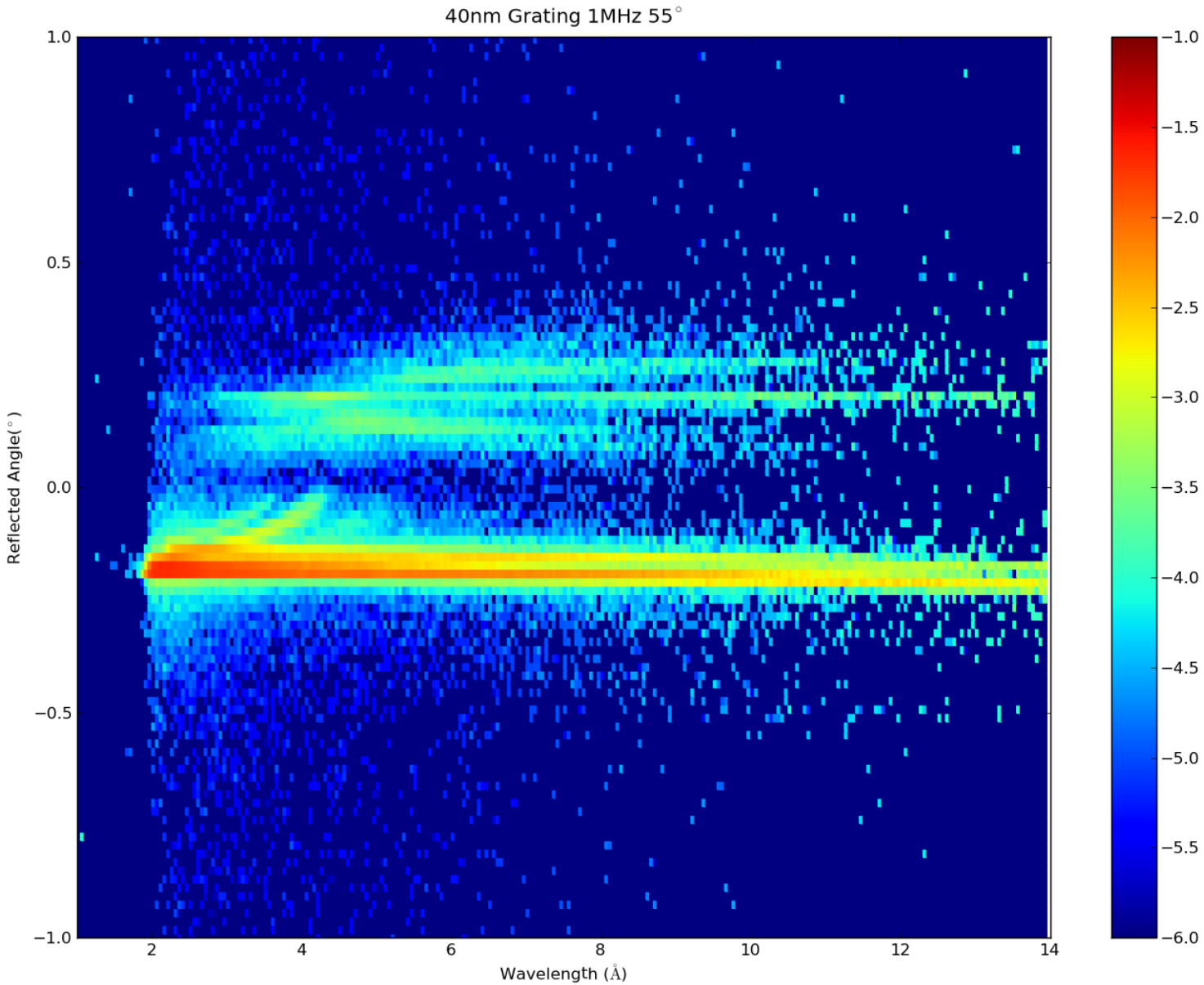
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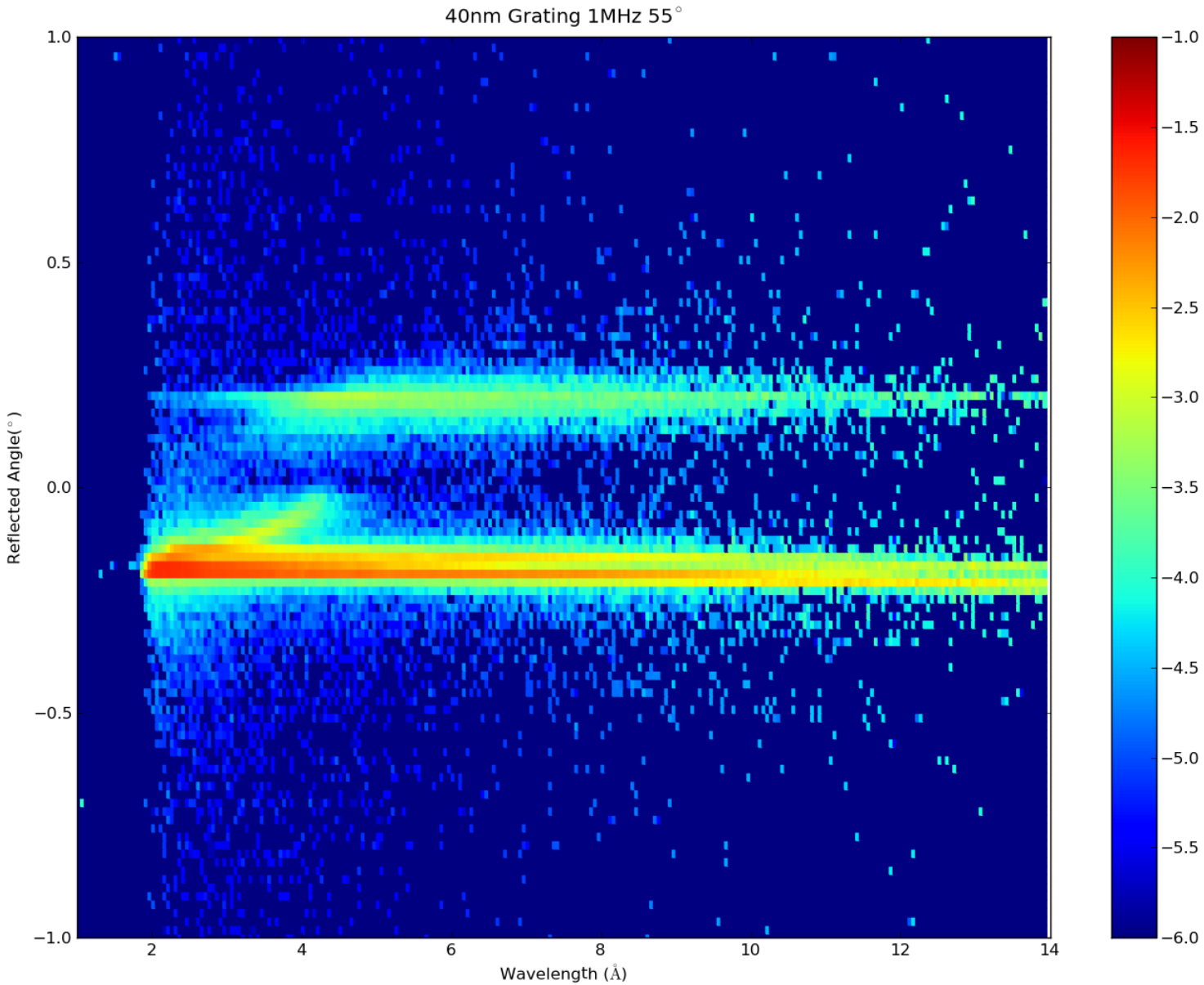
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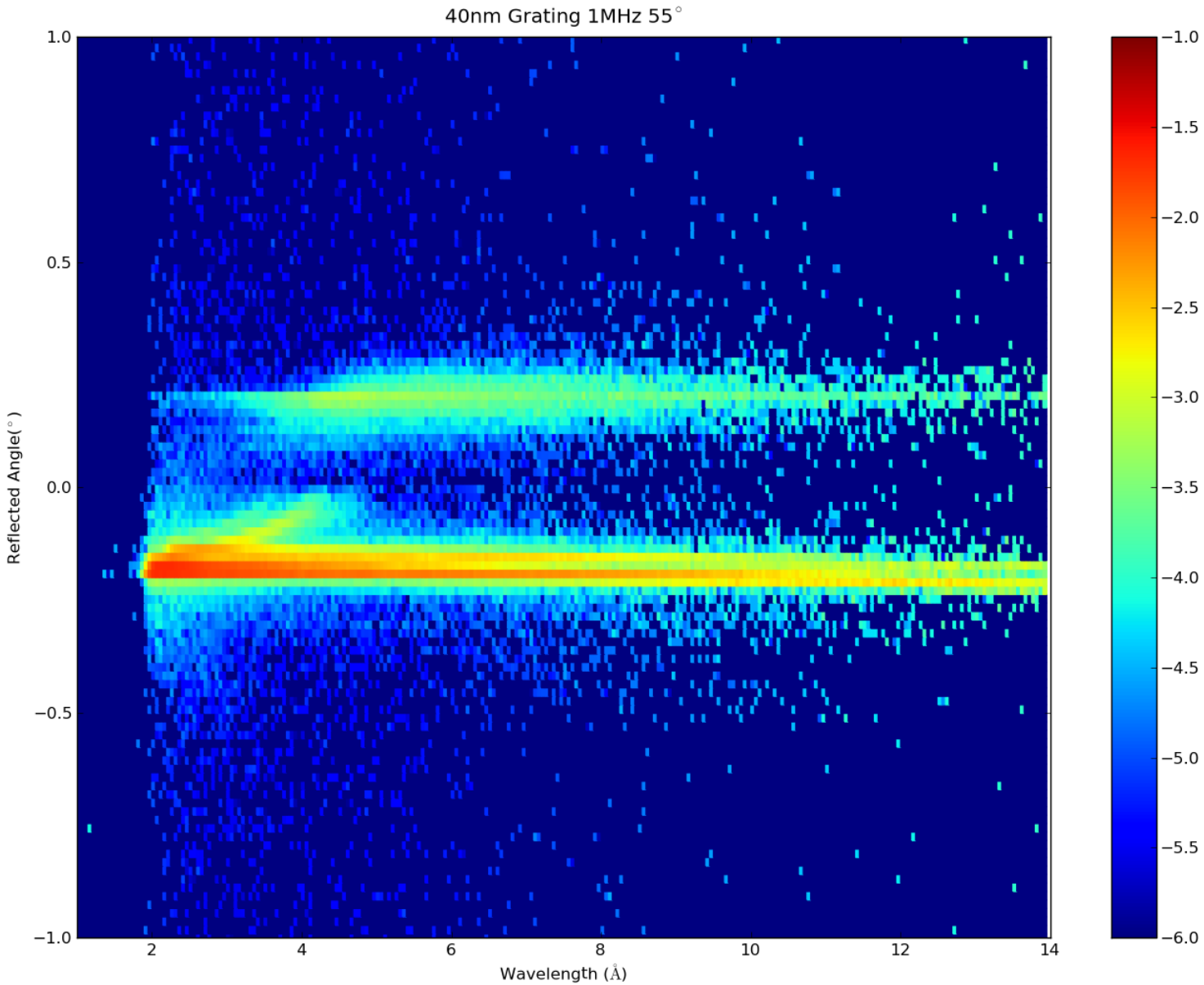
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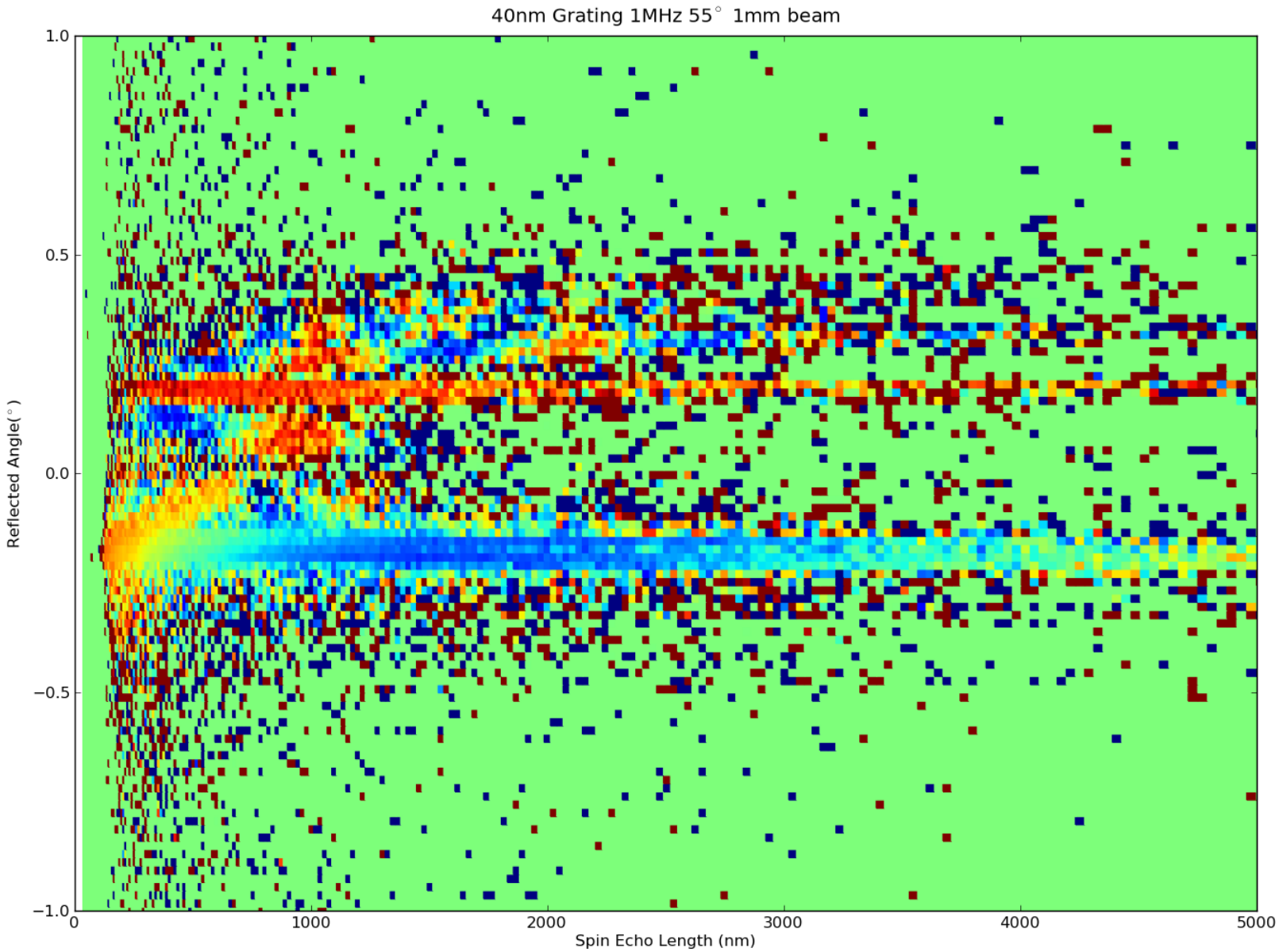
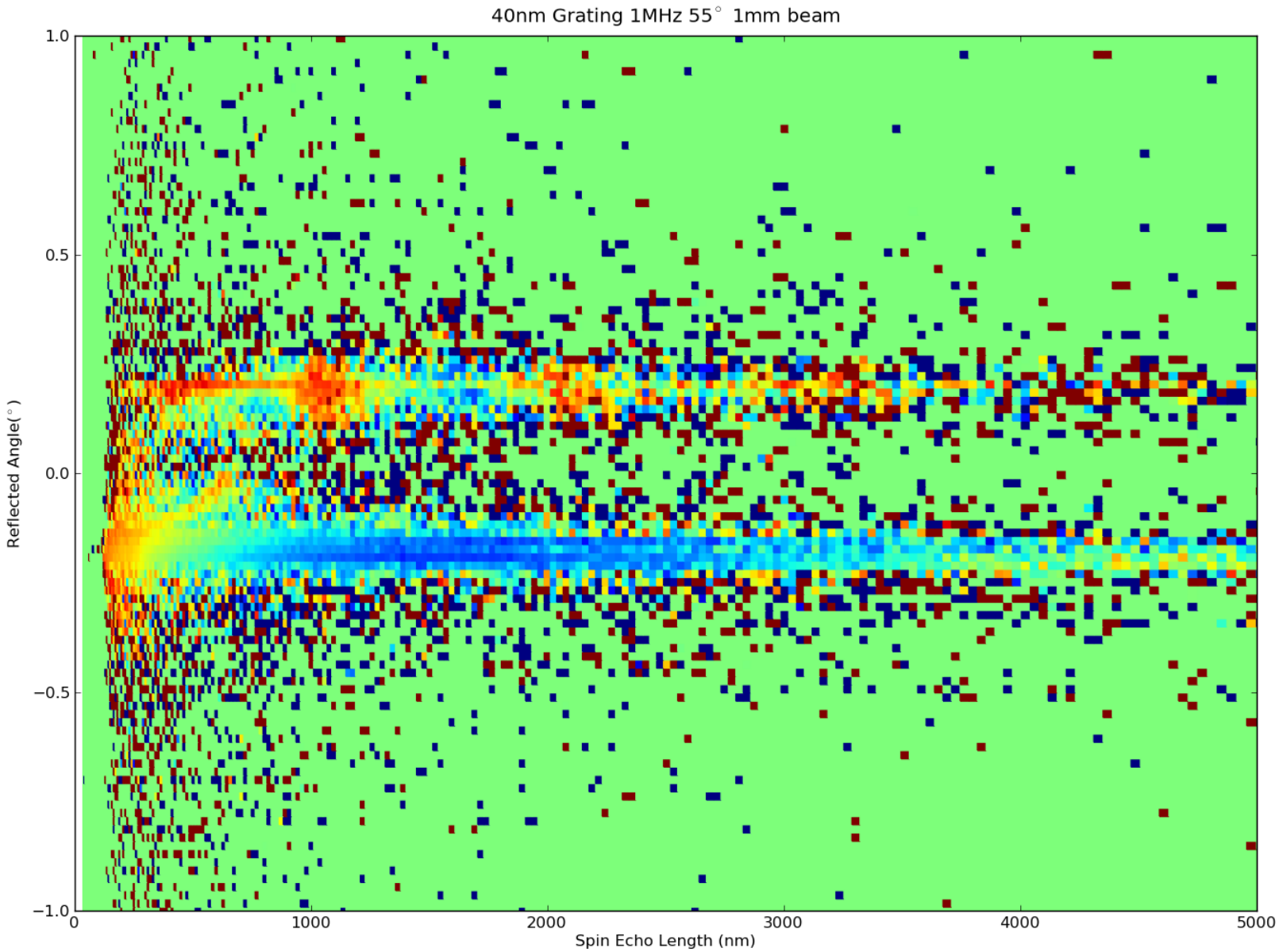
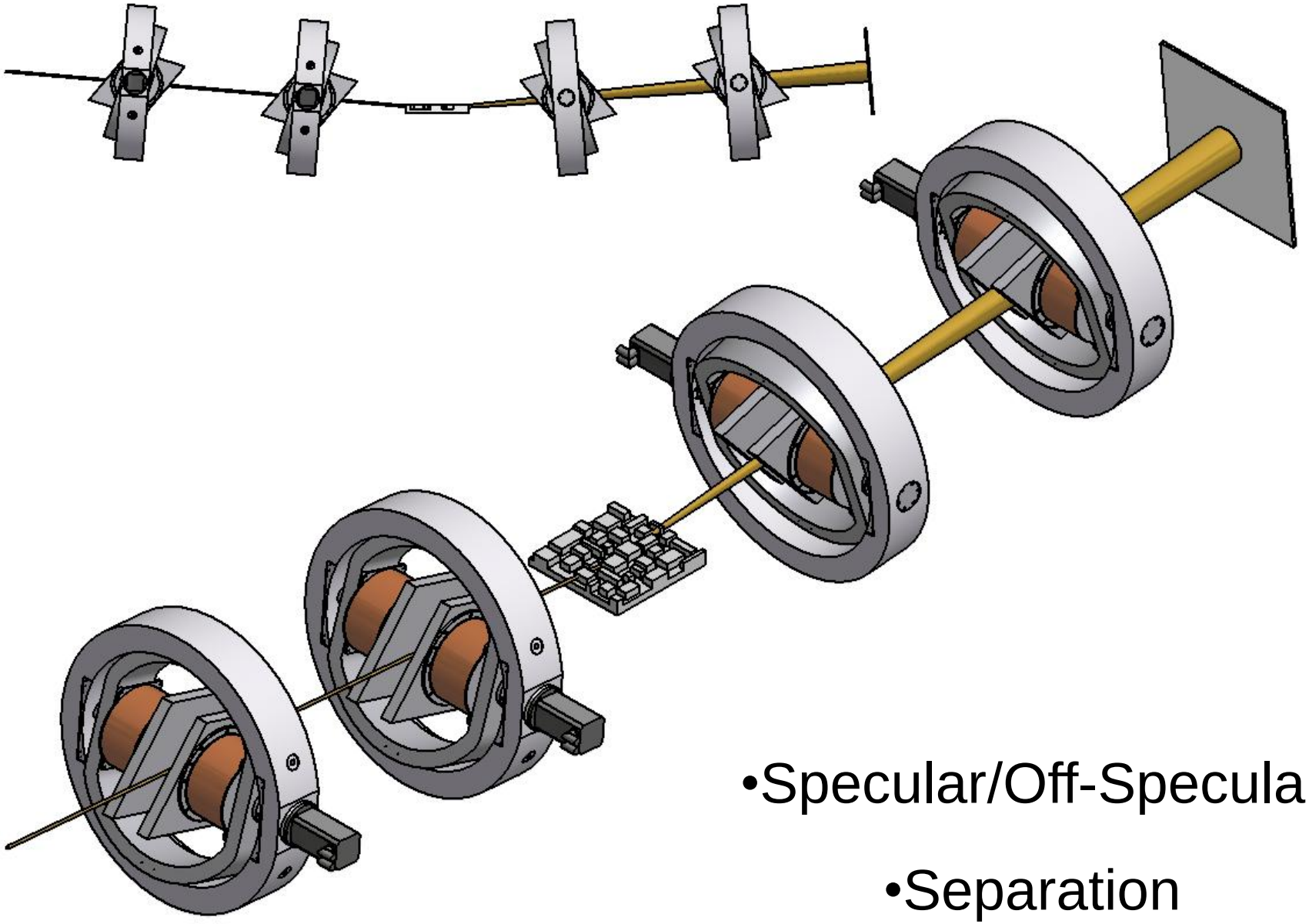
−5.5
−6.0

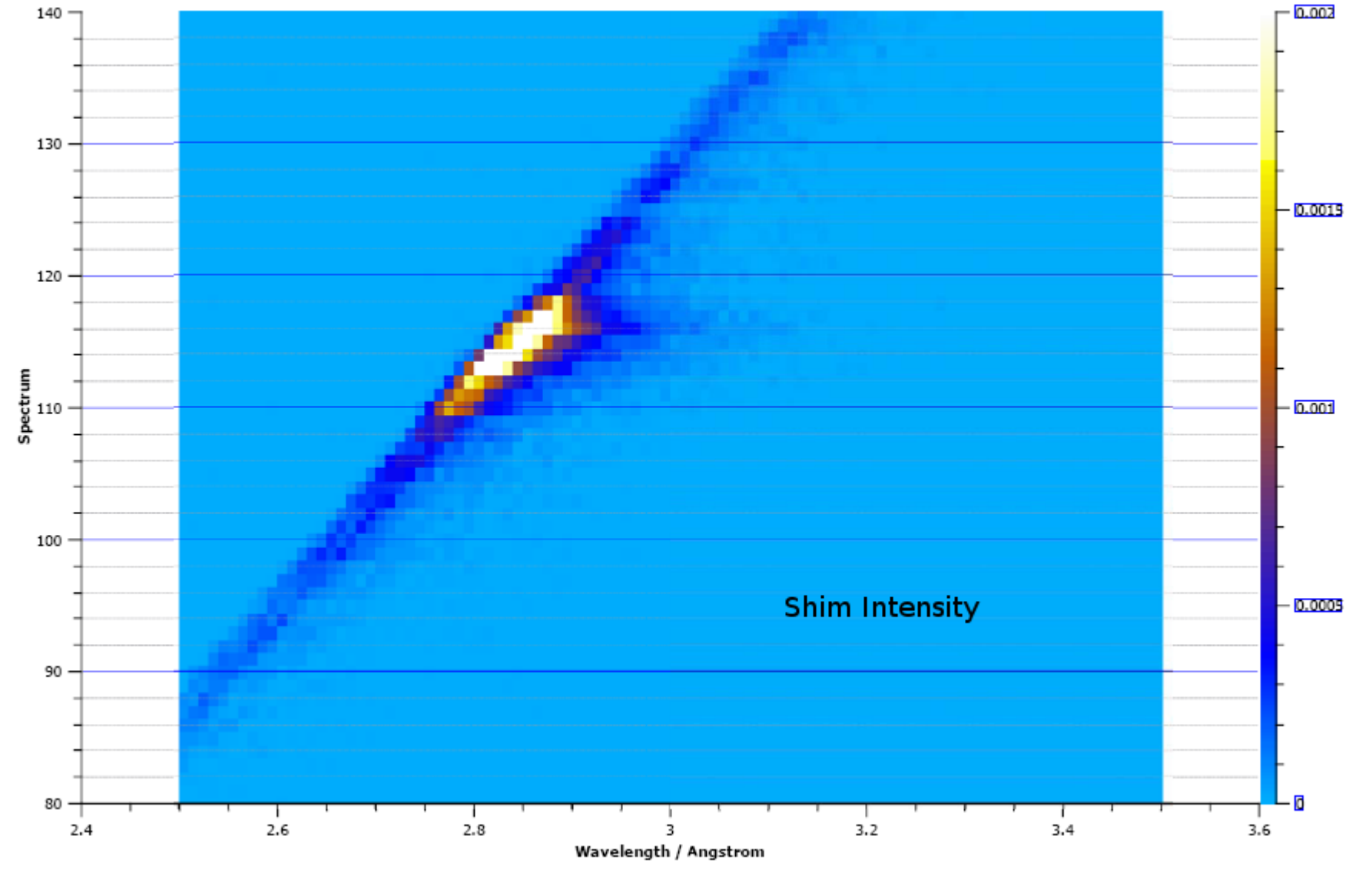

40nm Grating 1MHz 55° 1mm beam
Reflected Angle(°)
1.0
0.5
0.0
−0.5
−1.0
0
1000
2000
3000
4000
5000
Spin Echo Length (nm)

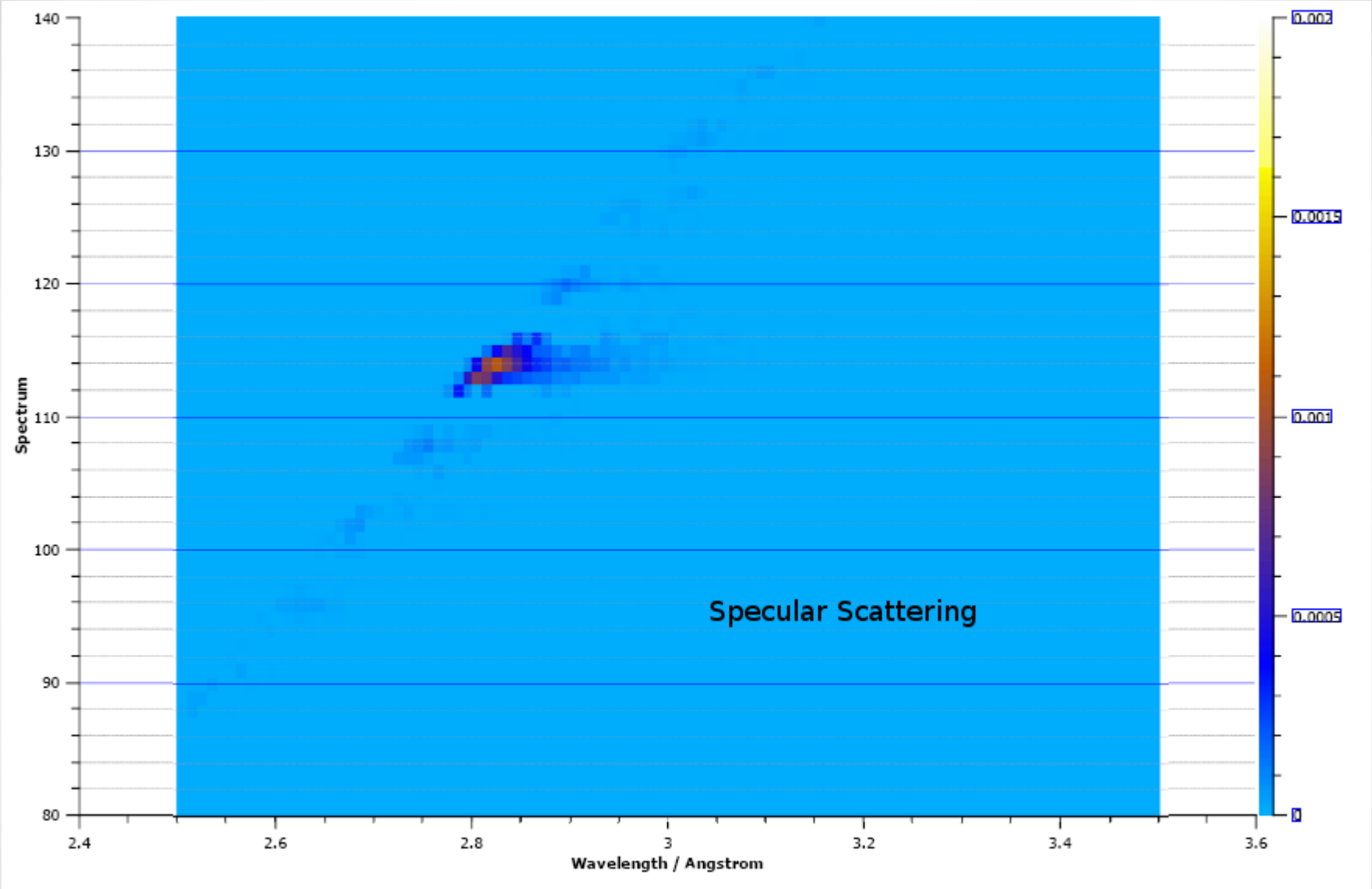

40nm Grating 1MHz 55° 1mm beam
Reflected Angle(°)
1.0
0.5
0.0
−0.5
−1.0
0
1000
2000
3000
4000
5000
Spin Echo Length (nm)

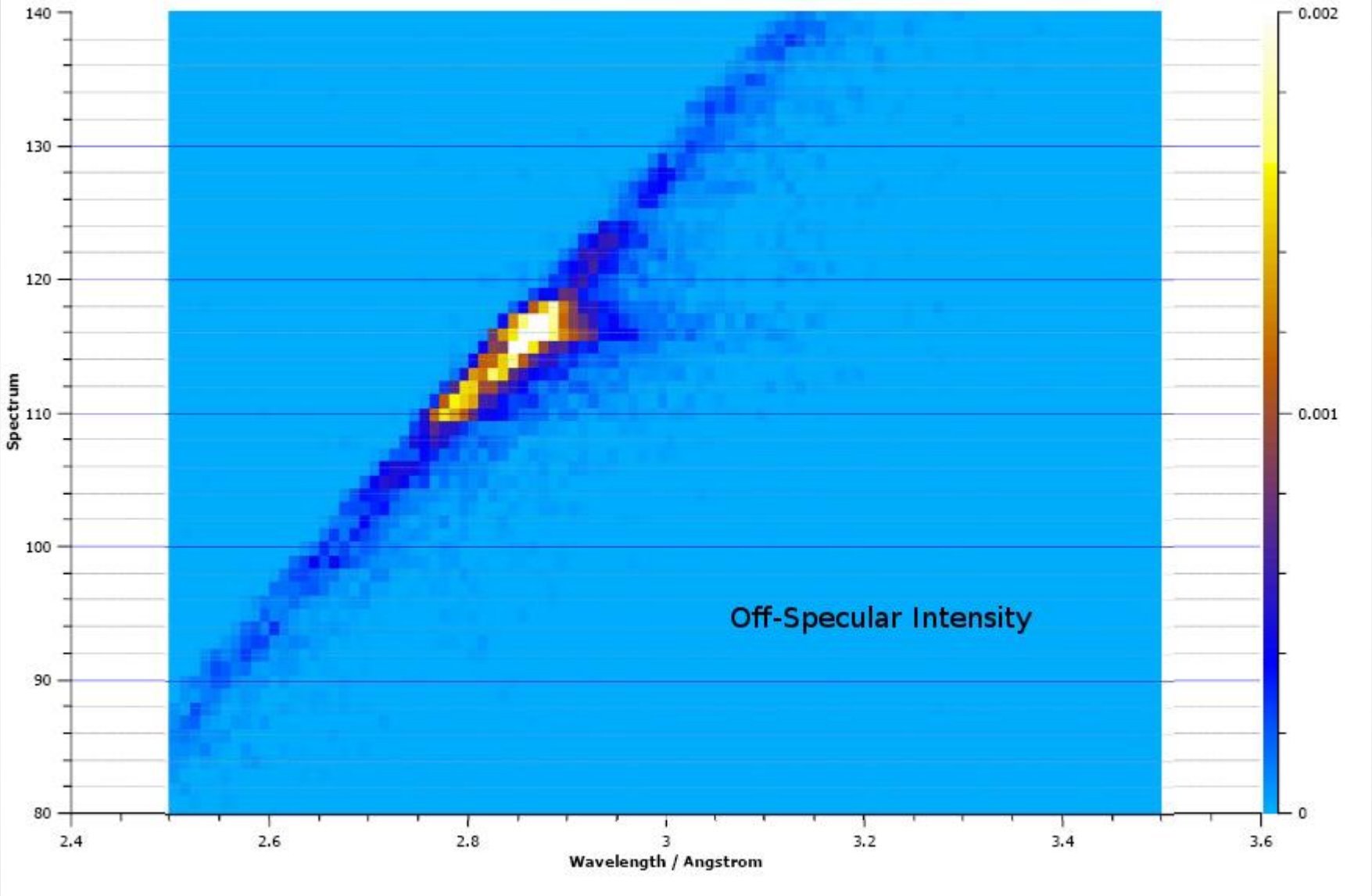


- Specular/Off-Specular
- Separation

Reflection from a liquid crystal close to a phase boundary

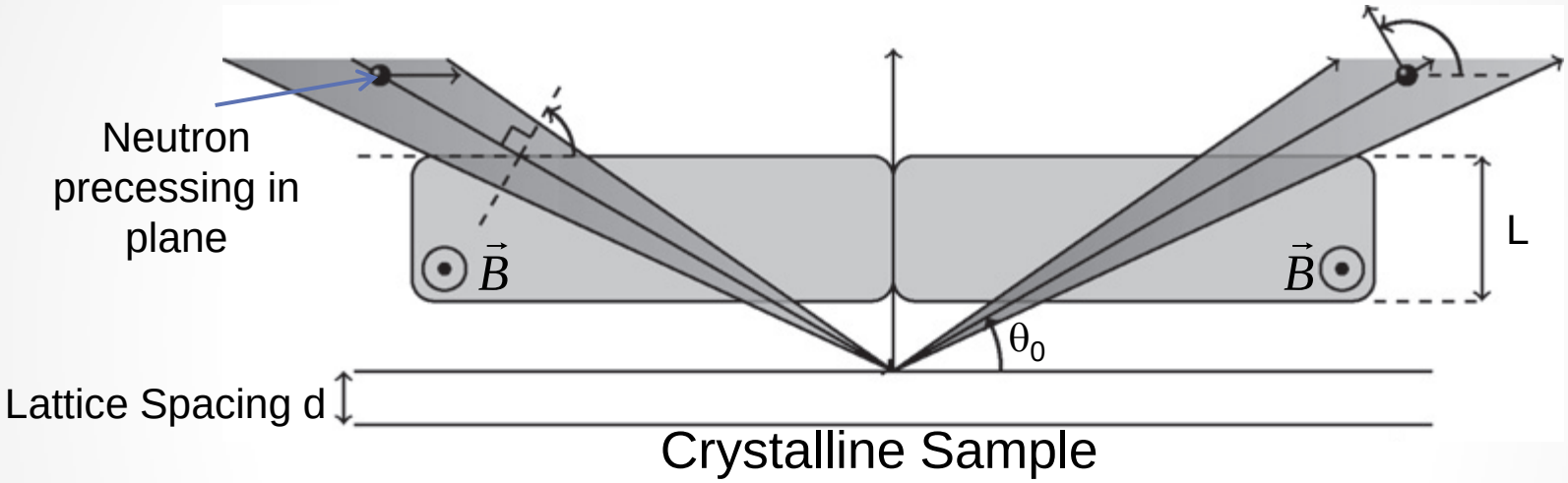



Specular Scattering
Spectrum
Wavelength / Angstrom
140
130
120
110
100
90
80
2.4
2.6
2.8
3
3.2
3.4
3.6
0.002
0.0015
0.001
0.0005
0


Off-Specular Intensity
Spectrum
Wavelength / Angstrom
140
130
120
110
100
90
80
2.4
2.6
2.8
3
3.2
3.4
3.6
0.002
0.001
0

Larmor Diffraction

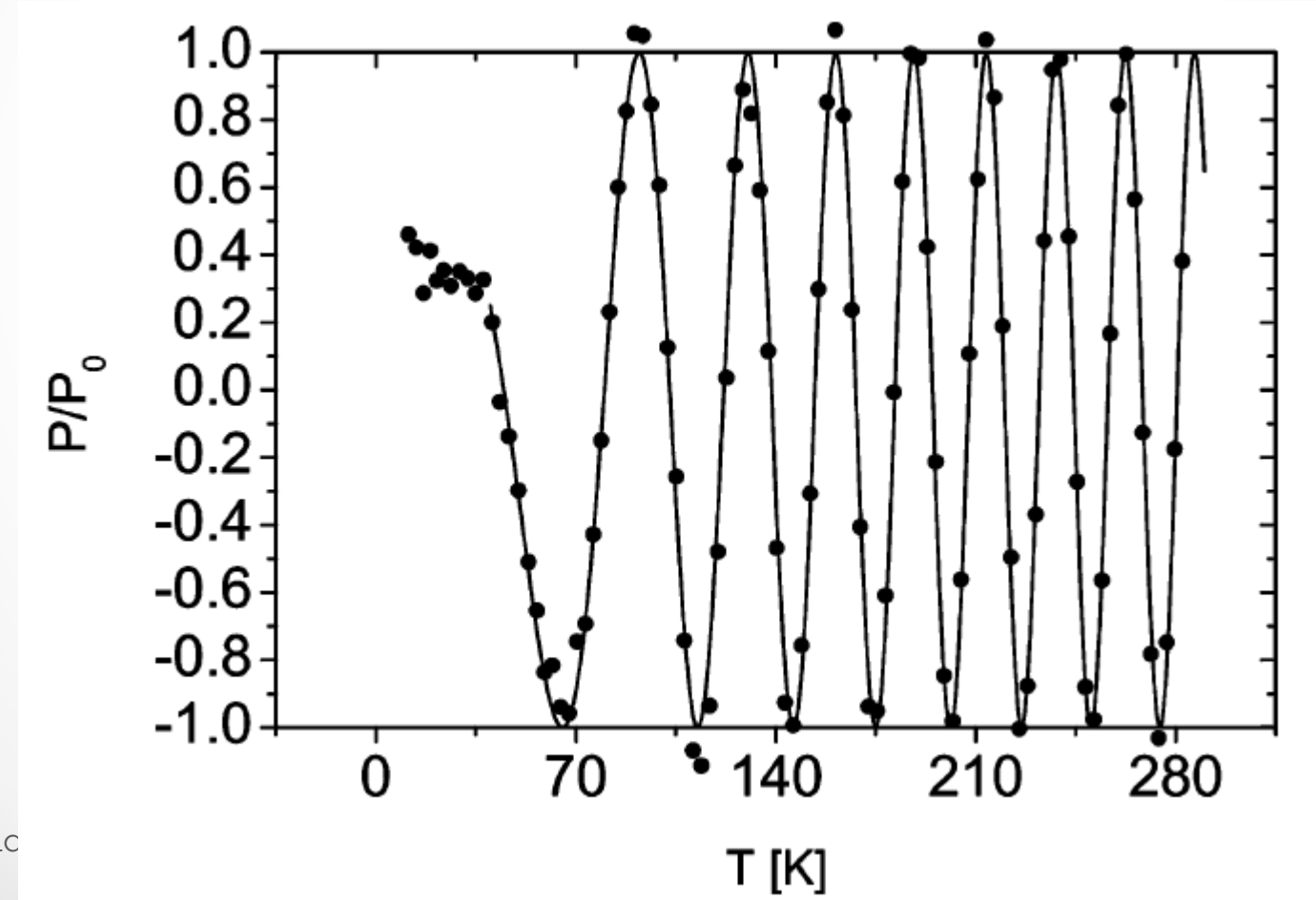
- Use angle encoding to perform very high resolution diffraction measurements.
  - E.g. Martin et al., Physica B406(2011)2333–2336



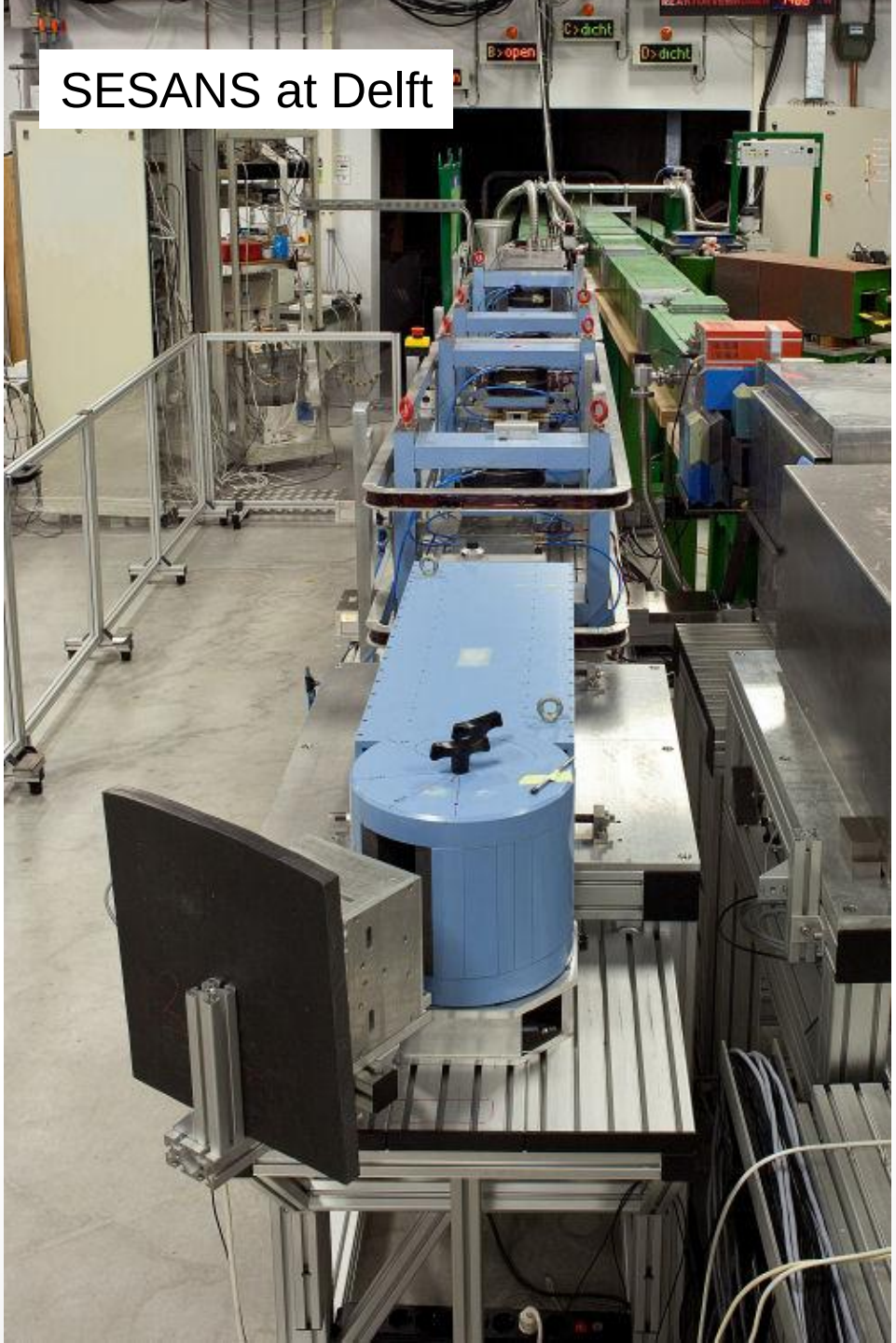
- The Larmor phase can be measured very accurately.
- Technique used to measure very small shifts in lattice spacing $\Delta d/d<10^{-4}$.

Larmor Diffraction

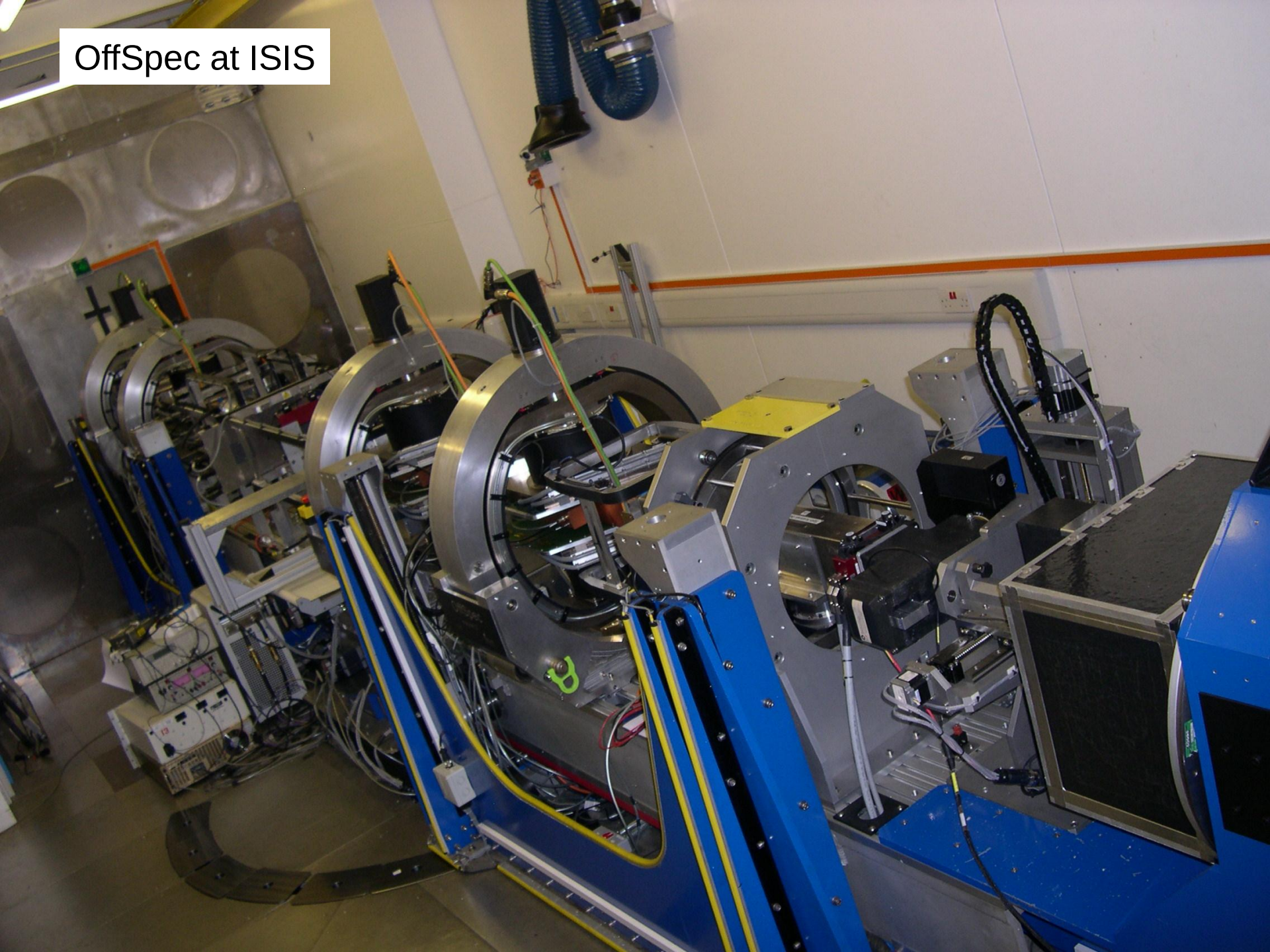
- Change in measured polarisation as a function of temperature for a polycrystalline Al sample.
- The change corresponds to a lattice constant change of 1 part in 10^4.
- Rekveldt et al. J. Appl. Cryst. **35**, 28 (2002)



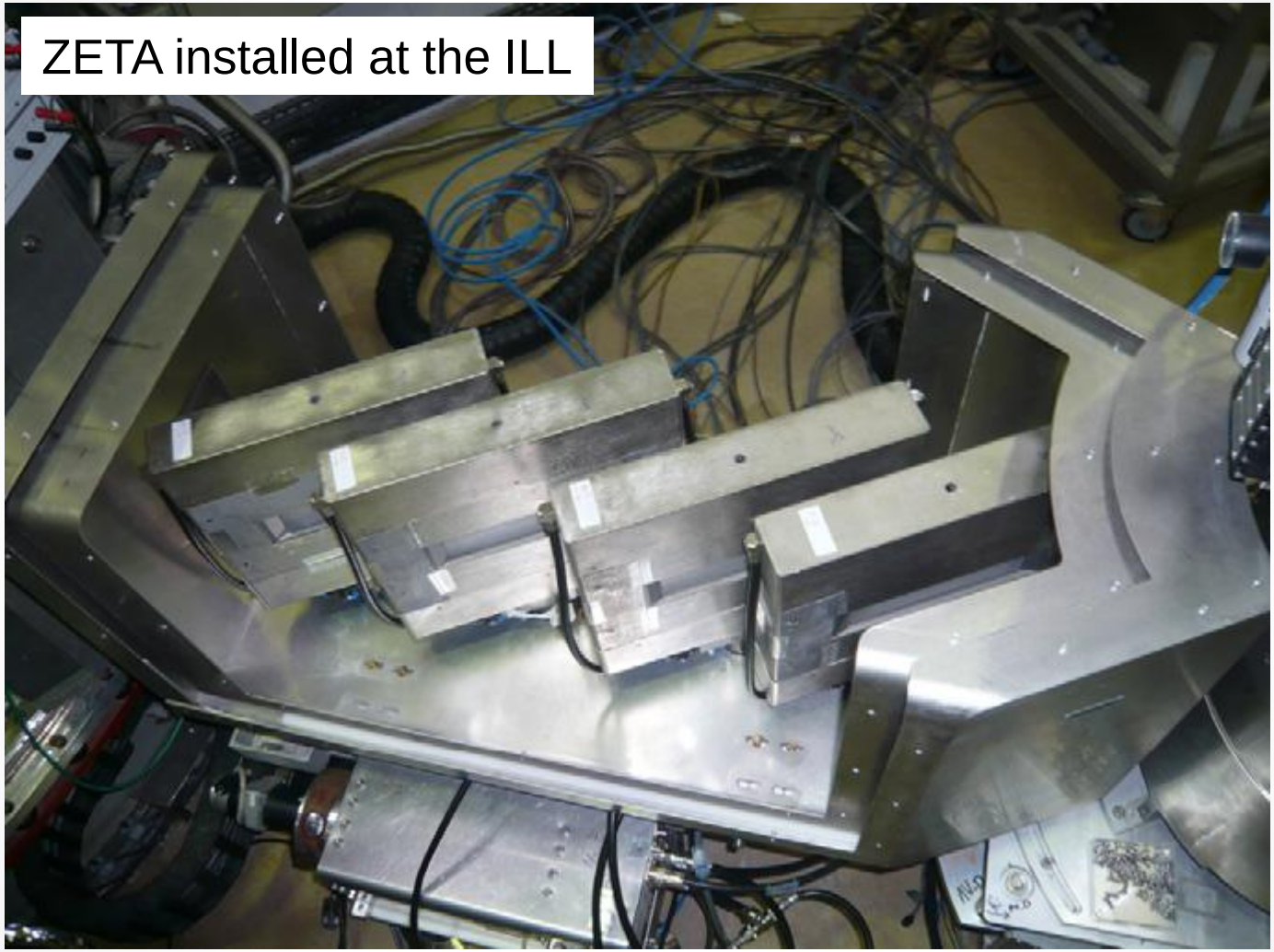
SESANS at Delft



OffSpec at ISIS



SESAME Indiana University


ZETA installed at the ILL

Summary of Part 2

- Current Instrumentation
- SESANS at TU Delft
- OffSpec at ISIS
- SESAME
- ZETA
- Practical Issues
- Precession region options
- Future Developments