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Neutron reflectometry: uniquely characterising interfacial phenomena

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ISIS, Rutherford Appleton Laboratory

Outline

- Motivation
  - *Unique* and *quantitative* description of complex materials on the microscopic lengthscale
  - Relating the functional properties of materials to their atomic and nanoscale structure
- Introduction to ISIS
- Introduction to reflectometry
  - Technique Fundamentals
  - Science Drivers
- Design Considerations
- Rainbows
- Pinhole Camera
- Off-specular
- Summary

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Acknowledgements

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EUROPEAN SPALLATION SOURCE

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- R. Cubitt

NEUTRONS FOR SCIENCE

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Rutherford Appleton Laboratory

Attracts 1200 users per year
Delivers over 700 different experiments every year.
~500 publications per year.

Interdisciplinary Research Centre

- Physics
- Chemistry
- Materials science
- Earth sciences
- Engineering
- Pharmaceutical science
- Biomolecular science
- Cultural heritage

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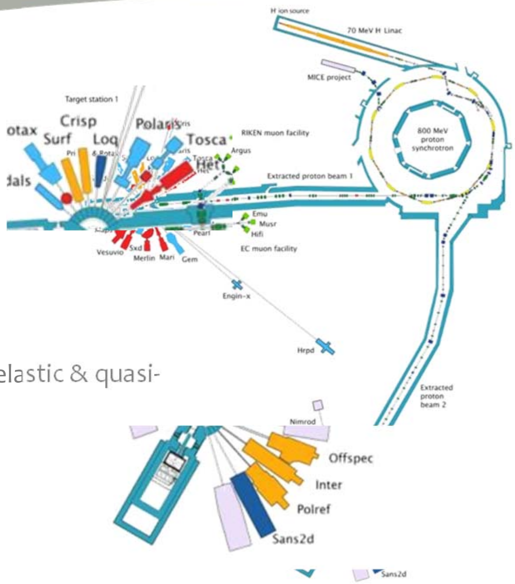
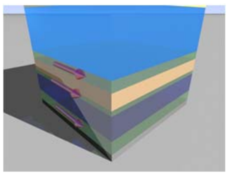
Structure (& morphology):
- powder diffractometers
- liquid diffractometers
- small angle scattering
- reflectometers
- imaging/tomography

Dynamics:
- neutron spectrometers (inelastic & quasi-elastic)
- muon spectrometers

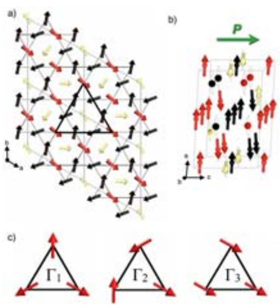
Others:
- support laboratories
- irradiation facility
- test facilities

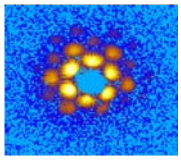


Neutrons provide…



- Absolute, quantitative information *both* on structure and magnetism
- From atomic to macroscopic lengthscales
- Relevant timescales
- Powders, single crystals, heterostructures, patterned structures & nanoparticles





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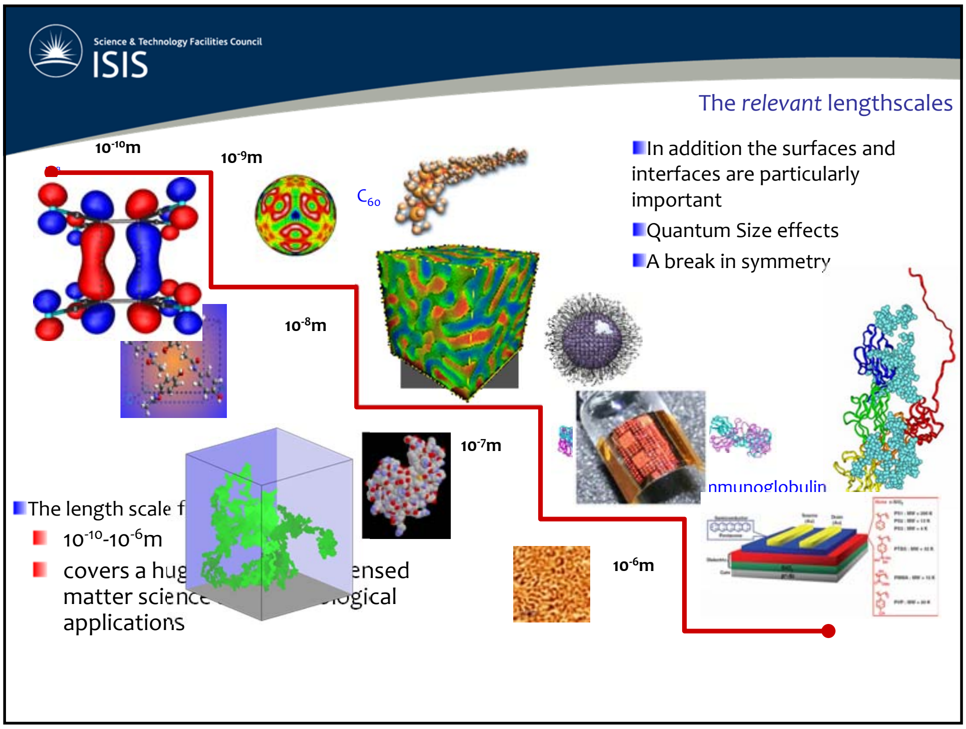
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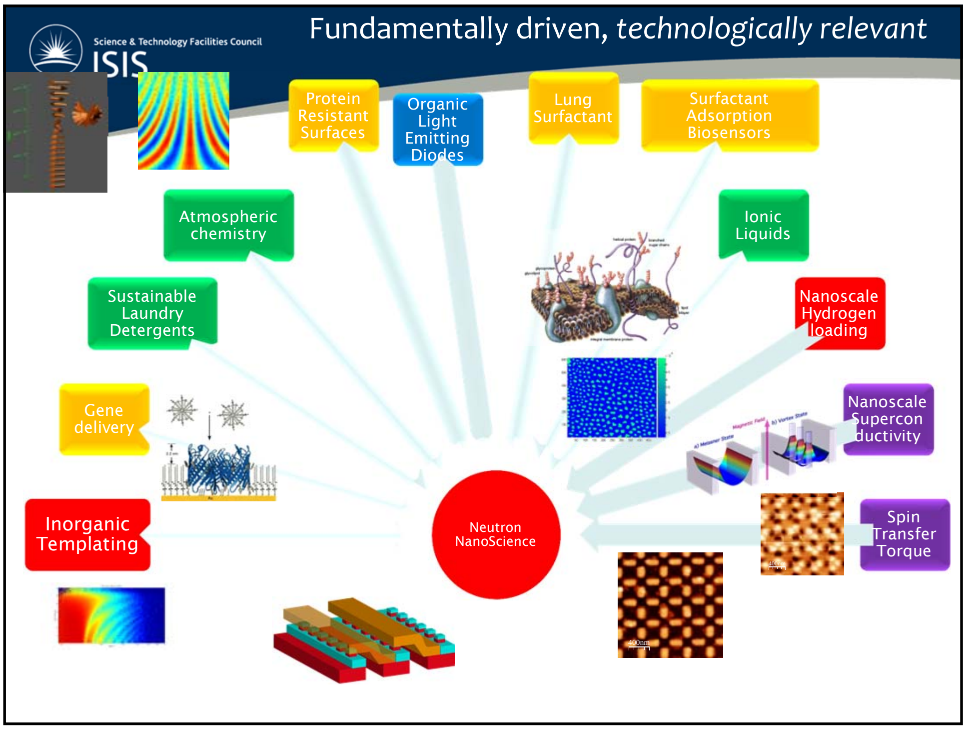
The *relevant* lengthscales
10^{-10}m
10^{-9}m
C_{60}
10^{-8}m
10^{-7}m
10^{-6}m
In addition the surfaces and interfaces are particularly important
Quantum Size effects
A break in symmetry
The length scale f
10^{-10}-10^{-6}m
covers a hug ensed matter science ogical applications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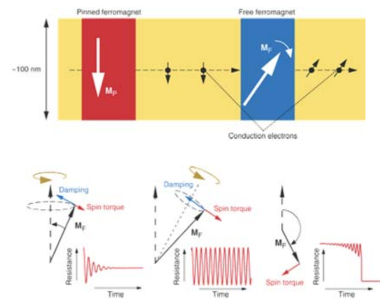
Fundamentally driven, *technologically relevant*
Protein Resistant Surfaces
Organic Light Emitting Diodes
Lung Surfactant
Surfactant Adsorption Biosensors
Atmospheric chemistry
Ionic Liquids
Sustainable Laundry Detergents
Nanoscale Hydrogen loading
Gene delivery
Nanoscale Supercon ductivity
Inorganic Templating
Neutron NanoScience
Spin Transfer Torque

The Importance of interfaces



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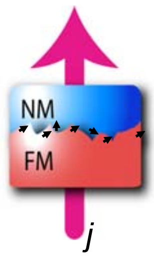

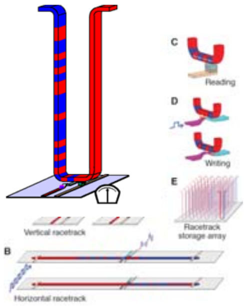

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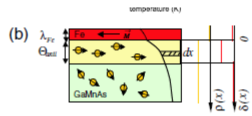
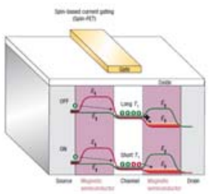
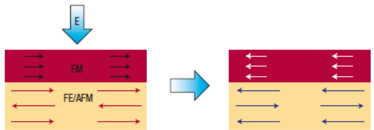


Maccherozzi et al. Phys. Rev. Lett. **101**, 267201 (2008)



Awschalom and Flatté Nat. Phys. **3** 153 (2007)



Ramesh and Spaldin Nat. Mat. **6**, 21 (2007)

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Scientific Drivers

- 3D structure of spatially confined materials
  - Specular, off-specular, g-sans, diffraction
  - Sub-nm to Micron in all dimensions
  - 10^{-8} from cm^2 sample
  - Kinetic information
- Non-destructive measurement
- Optimised sample environment
- Easy transition from data to understanding

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Scientific Drivers

- Weaker Effects
  - Magnetisation
    - 1nm (2u.c.) interface sensitivity
    - 2DEG, exchange bias, injected spin
    - 1emu cm^{-3}
  - Changes in scattering length density
  - Parametric Studies
- Smaller samples
  - Arrays
    - Spin injection
    - Spin-ices
  - Homogeneity
  - SE requirements
  - Compatible with TEM, x-ray, SQUID…

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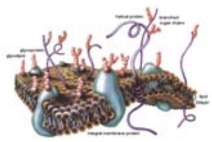
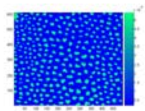
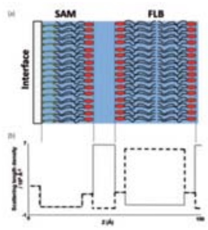
Scientific Drivers

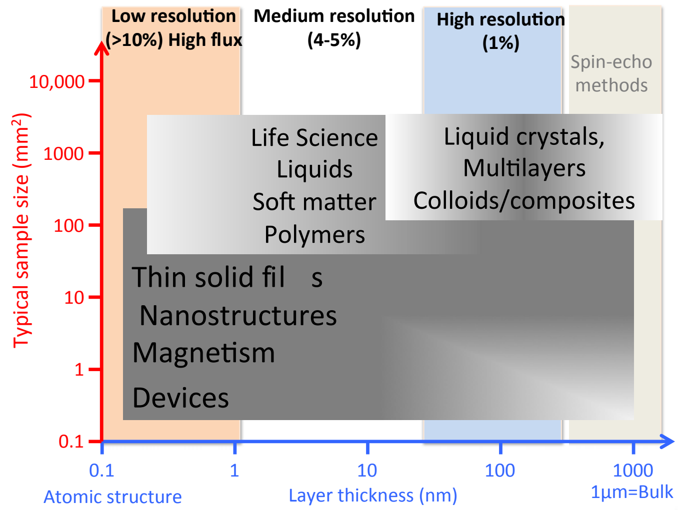




- Smaller lateral lengthscales
  - Membrane rafts
    - Micron to 10 nms
  - Changes in scattering length density
- Kinetics
  - What is the new kinetics on sub-second, millisecond timescale can we access
  - Wider community access
- Excitations
  - Interfacial and surface magnon-phonons
  - Capillary waves…
  - Offspec experience…



Requirements



HW ESS (2015)

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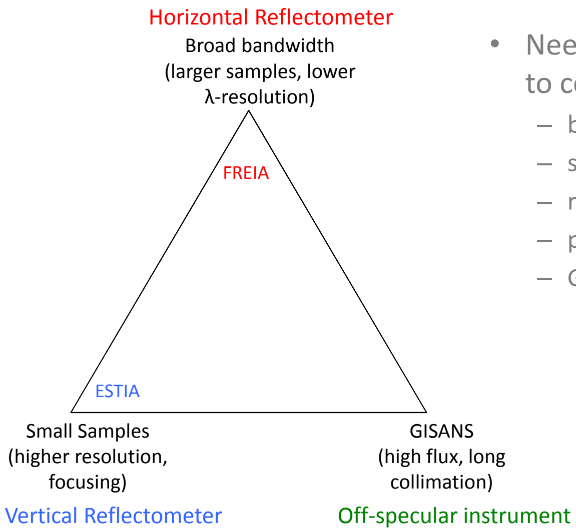
Reflectometer suite

EUROPEAN SPALLATION SOURCE



- Need a suite of instruments to cover all requirements:
  - bandwidth
  - sample size
  - resolution
  - polarization
  - GISANS/Offspec

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Science Case and Instrument Requirements Summary

Kinetics
- Real time
- Stroboscopic

Exotic Interfaces
- Liquid-liquid
- Ceramic

Dilute Systems
- Phase separation

Complex Systems
- Competing interactions
- Multi component

Parametric Studies
- Temperature
- Pressure
- Magnetic Field

→

High brightness
- Optimised moderators
- Focussing optics

Small Sample Size
- <1cm²

Large simultaneous Q-range
- Large wavelength band
- RRM
- Detector coverage

Tuneable resolution
- Advanced optics
- Chopper systems

Variable geometries
- High load/precision benches
- Tuneable optics

Specular Scattering

Neutron reflectometry

How do we connect the scattering length profile with the reflectivity

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Isotope Substitution

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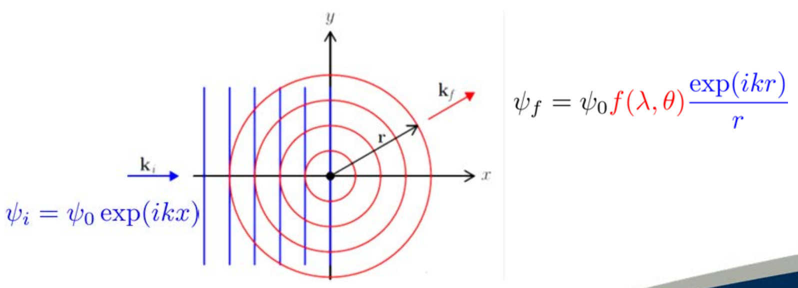
Scattering from a single (fixed) atom

- Atomic nuclei via the short-range (fm) strong force;
- Unpaired orbital electrons via a magnetic dipole interaction



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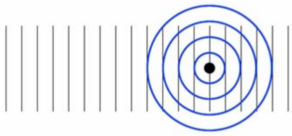
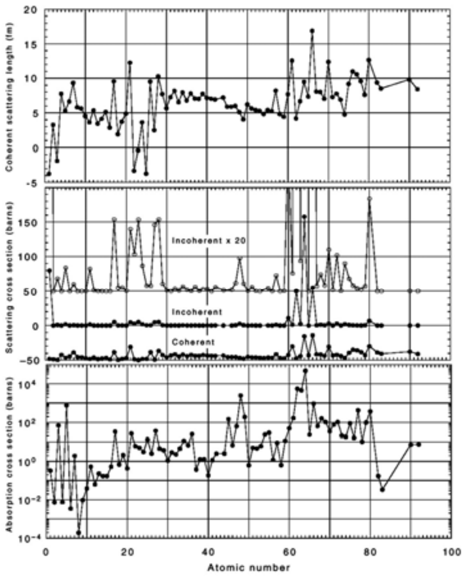
Scattering length

$$\sigma_{tot} = 4\pi b^2$$

Where *b* is the scattering length

- The sign of *b* is arbitrary
- A negative sign implies a change in the phase of the scattered wave
- *b* is sometimes complex and wavelength dependent due to resonant absorption
- *b* depends on the isotope
- *b* depends on the spin states of the neutron and nucleus





- Mass 1.67×10^{-27}kg 1.008665 atomic units
- Charge 0 (1.5 ±2.2) x 10^{-22}proton charge
- Spin ½
- Magnetic moment $-1.913\mu_N$
- Electric dipole moment

< 6 x 10^{-25}e-cm

$$\lambda = \frac{h}{mv}$$

$$E = \frac{h^2}{2m\lambda^2}$$

Isotope Dependence

Nickel Isotope	Scattering length b (fm)
^{58}Ni	15.0(5)
^{60}Ni	2.8(1)
^{61}Ni	7.60(6)
^{62}Ni	−8.7(2)
^{64}Ni	−0.38(7)

Hydrogen Isotope	Scattering length b(fm)
1H	−3.7409(11)
2D	6.674(6)
3T	4.792(27)
O	5.803

$$|11\rangle = \uparrow\uparrow$$
$$|10\rangle = (\uparrow\downarrow + \downarrow\uparrow)/\sqrt{2}$$
$$|1\ -1\rangle = \downarrow\downarrow$$
$$|00\rangle = (\uparrow\downarrow - \downarrow\uparrow)/\sqrt{2}$$

- Isotopic substitution for contrast
- Isotopic substitution to move peak positions in spectroscopy
- Incoherent scattering

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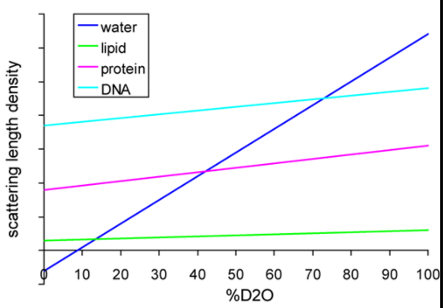
Scattering length density (SLD)

$$SLD = \sum_i b_i \frac{DN_a}{M_w}$$

Solvent	d (h form) (´ 10^{10} cm^{-2})	d (d form) (´ 10^{10} cm^{-2})	Polymer	d (h form) (´ 10^{10} cm^{-2})	d (d form) (´ 10^{10} cm^{-2})
Water	- 0.56	+ 6.38	PB	- 0.47	+ 6.82
Octane	- 0.53	+ 6.43	PE	- 0.33	+ 8.24
Cyclohexane	- 0.28	+ 6.70	PS	+ 1.42	+ 6.42
Toluene	+ 0.94	+ 5.66	PEO	+ 0.64	+ 6.46
Chloroform	+ 2.39	+ 3.16	PDMS	+ 0.06	+ 4.66
Carbon Tet.	+ 2.81		PMMA	+ 1.10	+ 7.22



Intro to SANS SM King

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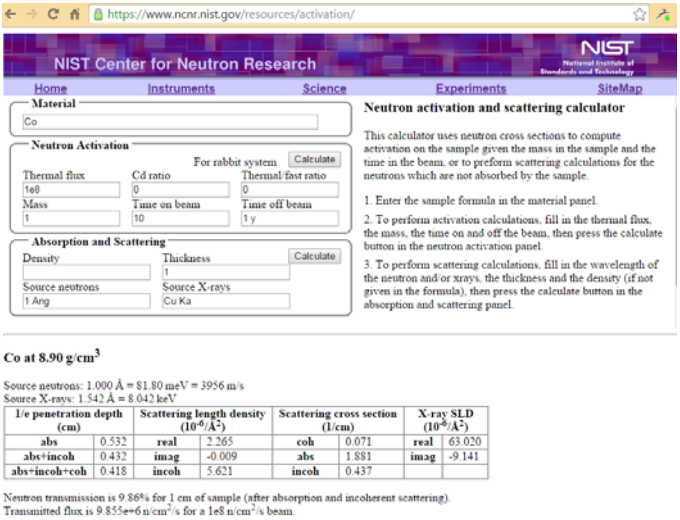
SLD

http://www.ncnr.nist.gov/resources/sldcalc.html


https://www.ncnr.nist.gov/resources/activation/

NIST Center for Neutron Research

Home Instruments Science Experiments SiteMap

Material

Co

Neutron Activation

For rabbit system — Calculate

Thermal flux: 1e8 | Cd ratio: 0 | Thermal/fast ratio: 0

Mass: 1 | Time on beam: 10 | Time off beam: 1 y

Absorption and Scattering

Density: | Thickness: 1 — Calculate

Source neutrons: 1 Ang | Source X-rays: Cu Ka

Neutron activation and scattering calculator

This calculator uses neutron cross sections to compute activation on the sample given the mass in the sample and the time in the beam, or to preform scattering calculations for the neutrons which are not absorbed by the sample.

1. Enter the sample formula in the material panel.

2. To perform activation calculations, fill in the thermal flux, the mass, the time on and off the beam, then press the calculate button in the neutron activation panel.

3. To perform scattering calculations, fill in the wavelength of the neutron and/or xrays, the thickness and the density (if not given in the formula), then press the calculate button in the absorption and scattering panel.

Co at 8.90 g/cm^3

Source neutrons: 1.000 Å = 81.80 meV = 3956 m/s
Source X-rays: 1.542 Å = 8.042 keV

1/e penetration depth (cm)		Scattering length density ($10^{-6}/Å^2$)		Scattering cross section (1/cm)		X-ray SLD ($10^{-6}/Å^2$)	
abs	0.532	**real**	2.265	**coh**	0.071	**real**	63.020
abs+incoh	0.432	**imag**	-0.009	**abs**	1.881	**imag**	-9.141
abs+incoh+coh	0.418	**incoh**	5.621	**incoh**	0.437		

Neutron transmission is 9.86% for 1 cm of sample (after absorption and incoherent scattering).
Transmitted flux is 9.855e+6 n/cm^2/s for a 1e8 n/cm^2/s beam.

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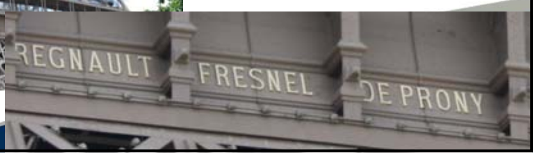
Interference effects



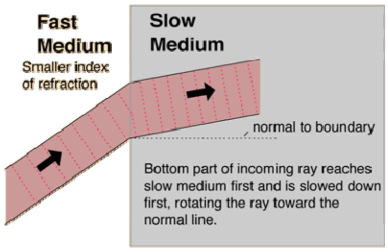
- Fresnel reflection 1815

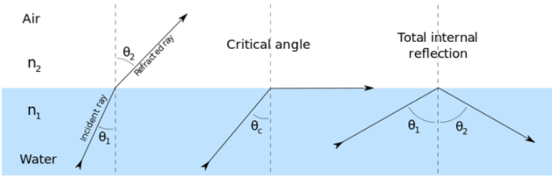
Refractive Index

$$n = \frac{c}{v}$$

- *n* varies with wavelength: dispersion

$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$





http://hyperphysics.phy-astr.gsu.edu
en.wikipedia.org

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$$n = 1 - \lambda^2 A - i\lambda B \quad (1)$$
$$A = \frac{Nb}{2\pi} \quad (2)$$
$$B = \frac{N(\sigma_a + \sigma_i)}{4\pi} \quad (3)$$

$$n = 1 - \alpha - i\beta \quad (1)$$
$$\alpha = \frac{N\lambda^2|r_e}{2\pi} \quad (2)$$
$$\beta = \frac{\lambda\mu}{4\pi} \quad (3)$$

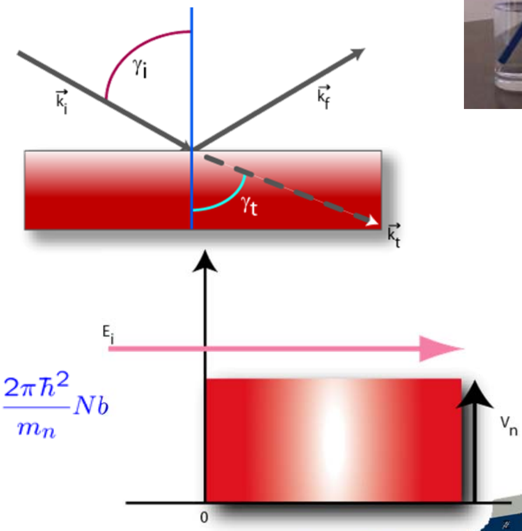
n <1 Total External reflection

Index of Refraction: Neutrons

$$
\begin{aligned}
n &= \frac{\sin \gamma_i}{\sin \gamma_t} \\
&= \frac{|\vec{k}_t|}{|\vec{k}_i|}
\end{aligned}
$$

$$
\begin{aligned}
n^2 &= \frac{|\vec{k}_t|^2}{|\vec{k}_i|^2} \\
&= \frac{E_t}{E_i} \\
&= \frac{E_i - V_n}{E_i} \\
&= 1 - \frac{4\pi}{k_i^2} Nb
\end{aligned}
$$

$$V_n = \frac{2\pi\hbar^2}{m_n} Nb$$

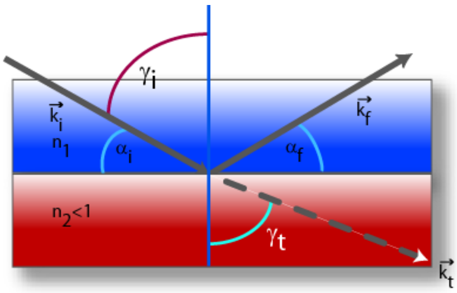


Critical Reflection

- $\frac{\cos \alpha_i}{\cos \alpha_f} = \frac{n_2}{n_1}$
- At the critical angle

  $\frac{\cos \alpha_i}{\cos 0} = n$

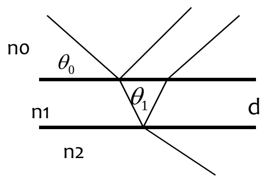
$$
\begin{aligned}
Q_c &= \frac{4\pi}{\lambda} 2k \sin \alpha_c \\
&= 2k\sqrt{1 - \cos^2 \alpha_c} \\
&= \sqrt{4k^2\left(1 - n^2\right)} \\
&\cong \sqrt{4k^2 \cdot 2\delta} \\
&= \sqrt{16\pi Nb}
\end{aligned}
$$



Material	θ_c / Å
Ni	0.1
Cu	0.083
Al	0.047
Si	0.047
D_2O	0.082

- Q_c only depends on the material!

Fresnel's law for a surface and thin film



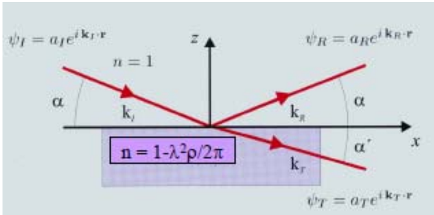
$$k_i = n_i \sin\theta$$

$$\beta_i = \frac{2\pi}{\lambda} n_i d_i \sin\theta_i$$

$$R(Q) = \left| \frac{r_{01} + r_{12}\exp(-2i\beta_i)}{1 + r_{01}r_{12}\exp(-2i\beta_i)} \right|^2$$

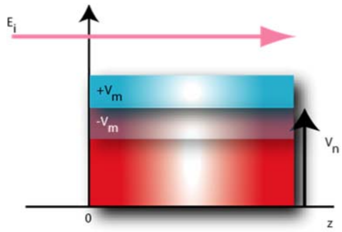
$$r_{ij} = \frac{a_r}{a_i} = \frac{k_i - k_j}{k_i + k_j}$$

$$t_{ij} = \frac{a_j}{a_i} = \frac{2k_i}{k_i + k_j}$$



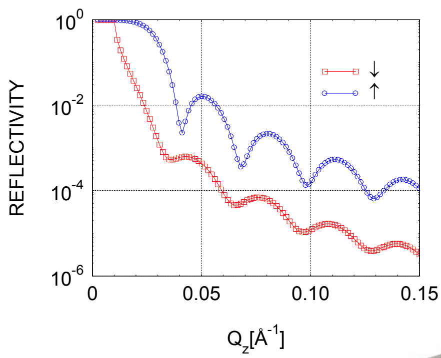
http://www.che.udel.edu/cns/pdf/Reflectometry.pdf

PNR from a single layer

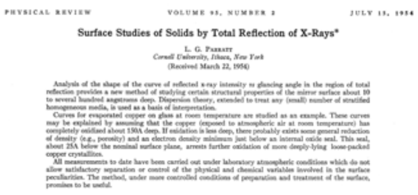


$$V = V_n + V_m \quad (1)$$

$$V = \frac{\hbar}{2\pi m} N(b \pm p) \quad (2)$$



Parratt Iteration



PHYSICAL REVIEW VOLUME 93, NUMBER 2 JULY 15, 1954

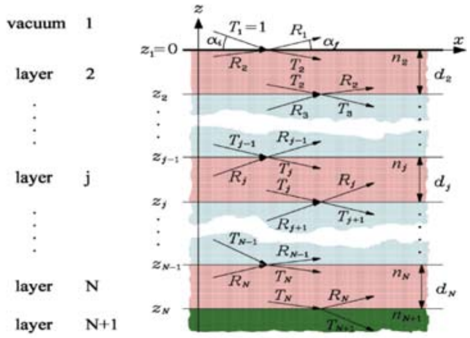
Surface Studies of Solids by Total Reflection of X-Rays*

L. G. PARRATT
Cornell University, Ithaca, New York
(Received March 22, 1954)

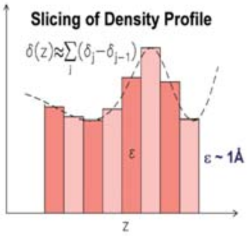
Analysis of the shape of the curve of reflected x-ray intensity vs glancing angle in the region of total reflection provides a new method of studying certain structural properties of the mirror surface about 10 to several hundred angstroms deep. Dispersion theory, extended to treat any (small) number of stratified homogeneous media, is used as a basis of interpretation.

Curves for evaporated copper on glass at room temperature are studied as an example. These curves may be explained by assuming that the copper (exposed to atmospheric air at room temperature) has completely oxidized about 150A deep. If oxidation is less deep, there probably exists some general reduction of density (e.g., porosity) and an electron density minimum just below an internal oxide seal. This seal, about 25A below the nominal surface plane, arrests further oxidation of more deeply-lying loose-packed copper crystallites.

All measurements to date have been carried out under laboratory atmospheric conditions which do not allow satisfactory separation or control of the physical and chemical variables involved in the surface peculiarities. The method, under more controlled conditions of preparation and treatment of the surface, promises to be useful.

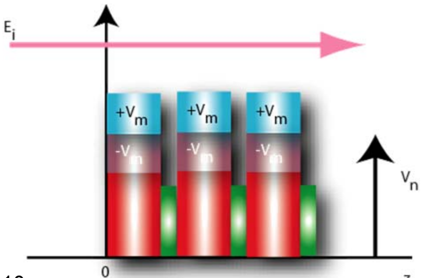


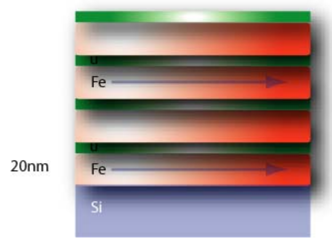
$$X_j = \frac{R_j}{T_j} = \exp(-2ik_{z,j}z_j)\frac{r_{j,j+1} + X_{j+1}\exp(2ik_{z,j+1}z_j)}{1 + r_{j,j+1}X_{j+1}\exp(2ik_{z,j+1}z_j)}$$

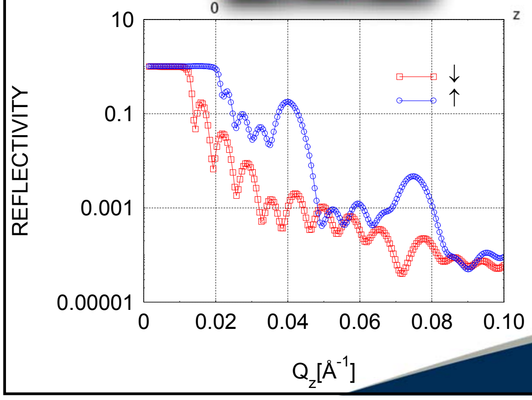


Can now simulate profile with a "*slice and dice*" approach

PNR from a multiple layers

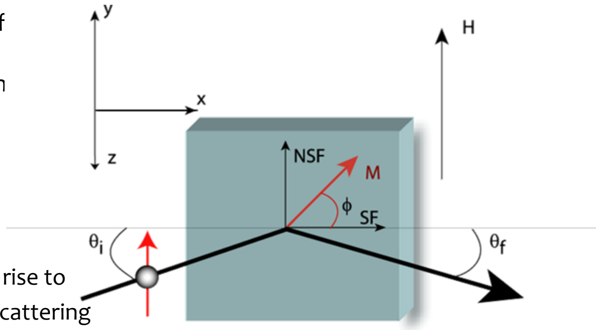






Spin dependent cross-section

- In-plane orientation of magnetisation obtainable from 4 spin dependent cross-sections



- Components of the magnetisation, m give rise to
- m||H: Non Spin Flip Scattering (NSF)
- m ⊥H: Spin Flip Scattering (SF)
- Dynamical analysis gives absolute depth dependence profile

$$b = b + p \sin\phi$$

$$p_m \cos\phi = p_x$$

$$\left[\frac{-\hbar^2}{2m_n}\nabla^2 + V(r)\right]\Psi^{\uparrow,\downarrow} = E\Psi^{\uparrow,\downarrow}$$

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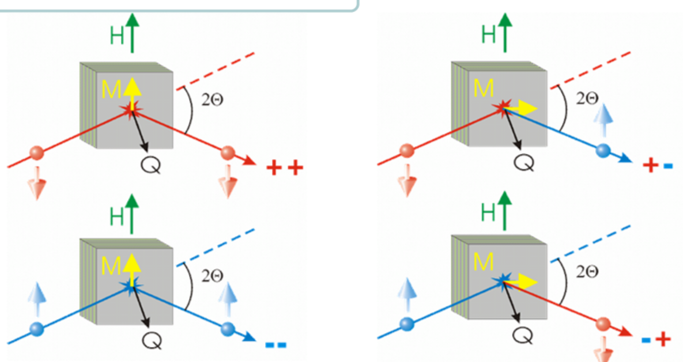
Polarisation

Polarised neutron reflection



Non spin flip
++ measures b + M_z
– – measures b – M_z

Spin flip
+ – measures M_x + iM_y
– + measures M_x – iM_y

By fitting all components the direction and strength of the magnetic moment can be measured as a function of depth

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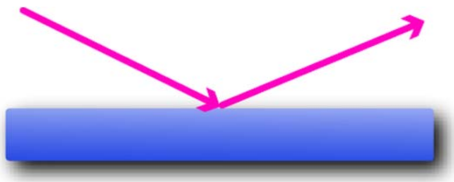
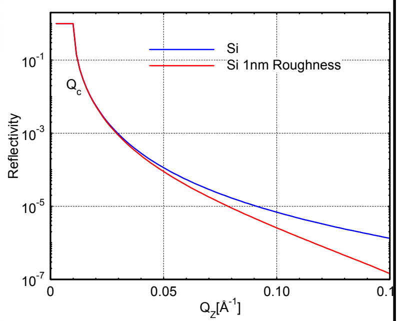
Roughness

Structural and magnetic interfacial phenomena

Structural Roughness



$$R(Q) = R_F \exp\left(-Q^2\sigma^2\right)$$



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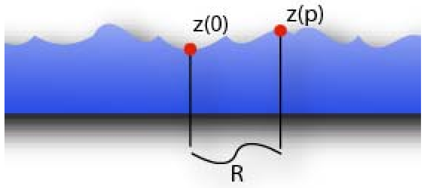
PHYSICAL REVIEW B VOLUME 38, NUMBER 4 1 AUGUST 1988

X-ray and neutron scattering from rough surfaces

S. K. Sinha, E. B. Sirota, and S. Garoff*
Corporate Research Science Laboratory, Exxon Research and Engineering Company, Clinton Township, Route 22 East, Annandale, New Jersey 08801

H. B. Stanley†
University of Maryland, College Park, Maryland 20742
(Received 30 November 1987)

$$C(R) = \left\langle \left[z(x,y) - z(x',y') \right]^2 \right\rangle$$
$$= \sigma^2 \exp(-r/\xi)^{2h}$$



- σ= roughness
- ξ= cut-off length:
  - for R > ξ, interface appears smooth,
  - for R < ξ, interface appears rough, fractal behaviour
- h=3-D Hurst parameter for jaggedness(0<h<1)
  - smooth: D=2, h=1
  - very rough D=3, h=0

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Off-specular scattering

Probing in-plane lengthscales

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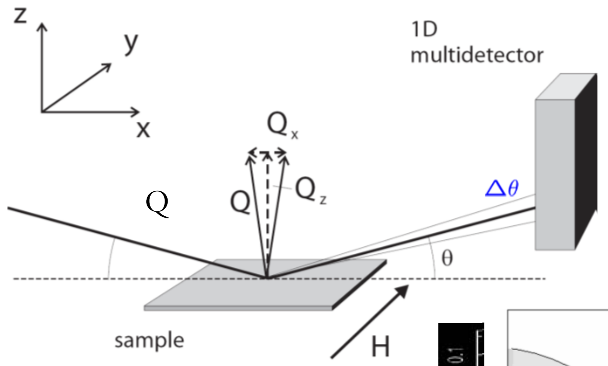
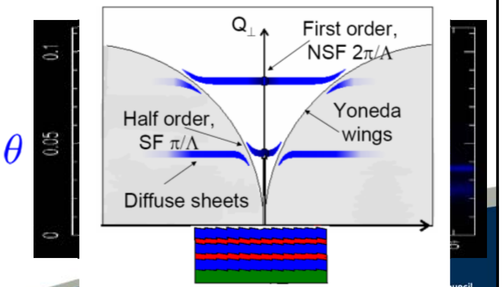
Experimental Geometry

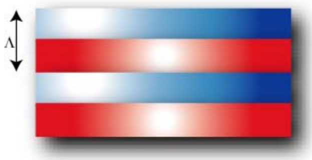


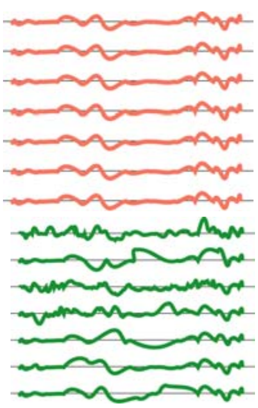
$$Q_x = |Q| \sin\theta \times \Delta\theta$$
$$Q_y = |Q| \times \Delta\phi$$

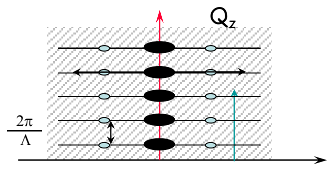


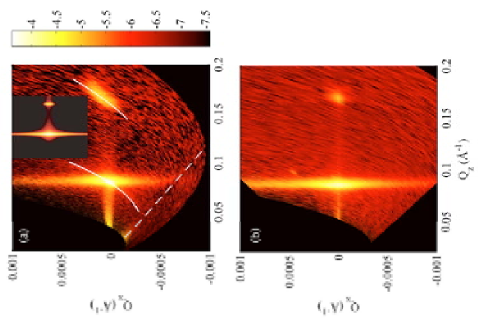
- λ range 0.5-6.5Å
- 1-d Detector
- Typical acquisition~2hours/field
- Measured coherence length ~30μm

Diffuse scattering









Holý and Baumbach, prb, 49, 10668, (1994)

Simulation Packages

Data Analysis

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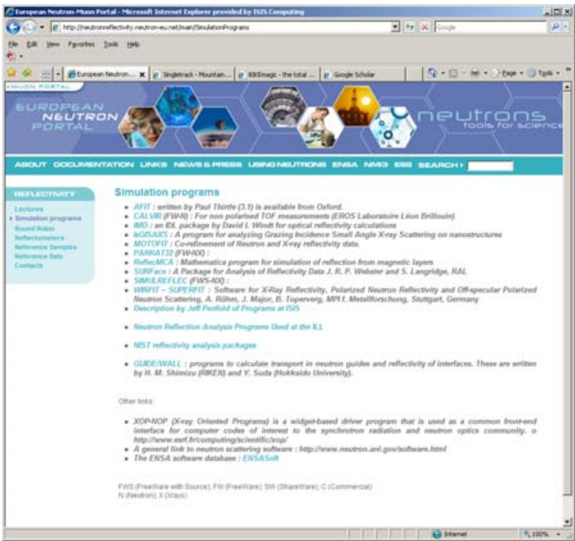
Simulation Packages: neutron

http://neutronreflectivity.neutron-eu.net/main/SimulationPrograms



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Software

- Strengths
  - Absolute cross-section
  - Small hardware footprint (?)
  - Diversity
- Opportunities
  - New algos
  - Off-specular/GISANS
  - HPC (DFT, MD etc.)
  - Inverse problem

INSTRUMENT OUTLINE

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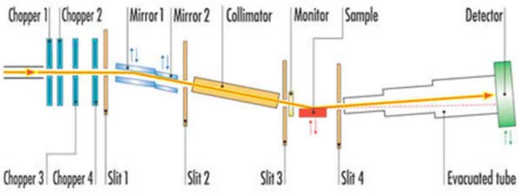
FIGARO – horizontal sample reflectometer at ILL



Q range=0.005 Å^{-1} < Q < 0.4 Å^{-1}
Horizontal samples
Free liquids
Liquid/liquid interfaces
TOF

http://www.ill.eu/instruments-support/instruments-groups/instruments/figaro/

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Simulation

- Detailed simulation of instrument to investigate accessible parameter space
  - Pulse shaping
  - Resolution
  - Guide options
  - Be cautious

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Challenges

- Focussing (Exploit optics)
  - x25 gain for purely specular
  - Homogeneous profile
  - Directional moderators
- Backgrounds
- Spin separation/recombination
  - Larmor labelling
- Detectors
  - Dynamic range (background)
  - Spatial resolution
- Out of plane resolution
- Lengthscales
- Quality control (1=1?)
  - Statistical & *systematic* errors
- Analysis
  - Reduction/visualisation
  - Theory
  - HPC

- Deuteration
  - Do the best experiment
- Software
  - Reduction/Analysis
  - Pipeline development
  - Data mining (Big data)
- Sample Environment
  - H,T,P
  - Shear
  - Realistic surfaces
    - Beyond Si
- Dual colour measurements
- Pipelines

Polref schematic

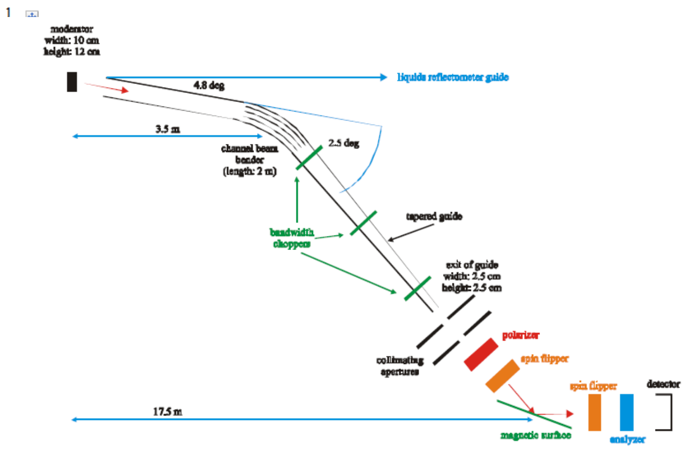

Figure 1 An outline side schematic of polREF.

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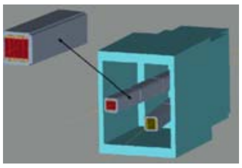
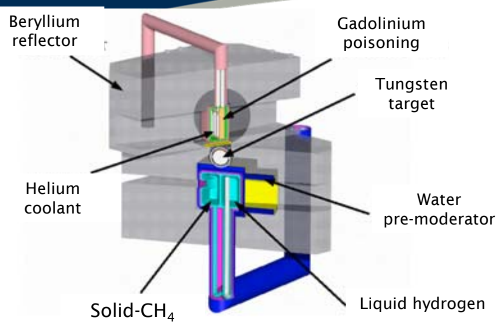
Internal Dimensions	Guide Type	Coating	Starting Position	End Position	Internal
10 cm (H) x 12 cm (V)	flight tube, absorbing walls	no coating	1.00 m	2.50 m	^{4}He
10 cm (H) x 12 cm (V)	horiz.beam bender 6 channels	3.6 x θ_c Ni (H) 3.6 x θ_c Ni (V)	2.50 m	4.50 m	^{4}He
10 cm (H) to 3 cm (H) x 12 cm (V) to 3 cm (V)	tapered neutron guide	3.6 x θ_c Ni (H) 3.6 x θ_c Ni (V)	4.50 m	16.50 m	Vacuum

- Moderator-Guide coupling
- Guide System
- Bender
- Choppers
- Collimation
- Sample position
- Polarisation
- Detection

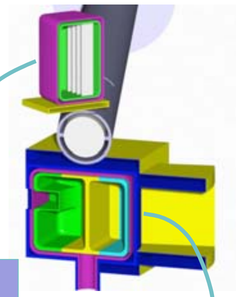


- Moderator
  - Cryogenic moderator (relaxed time resolution)
- Guide System
  - Couple efficiently to the moderator
  - Efficiently transport neutrons to the sample/detector
  - Bender
    - Move from line of sight
    - Multi-channel (~6)
    - Vertical and horizontal guide coating
    - Alignment

Target/Moderator Assembly



Highly compact and efficient target-moderator-reflector assembly

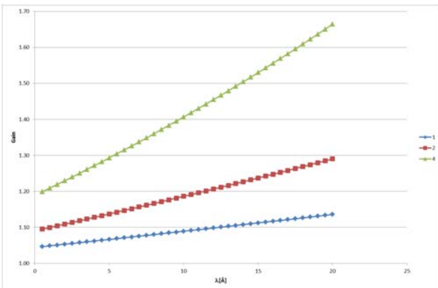


Target	
Proton energy	800MeV
Proton current	60μAmps
Average power	48 kW
Pulse rate	10Hz
Proton beam Size	1.5cm FWHM
Target	W Clad in Ta
Target coolant	D_2O
Reflector	Be
Reflector coolant	D_2O

Decoupled moderator
- solid methane (26K)
- asymetrically poisoned

Coupled moderator
- solid-methane / liquid-hydrogen composite
- pre-moderated with H_2O
- Grooved face
- Hydrogen face

- Vacuum
- Chopper system
  - To
    - Only necessary for line of sight
    - Bandwidth chopper (control $\Delta\lambda$)
- Data Acquisition
- Polarisation
  - High degree of polarisation required
- Detector
  - High count rate 10^6 per pixel
  - Low gamma sensitivity
  - Magnetically insensitive
  - High efficiency
  - 2D spatial resolution <1mm
  - Large dynamic range



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REFLECTOMETRY@ISIS

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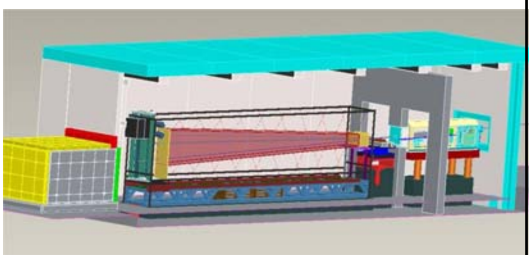
Inter

- Designed for the study of chemical interfaces, with a particular emphasis on the air-water interface
- >10 times the flux of SURF
- Much wider dynamic range
- Tuneable resolution



Scientific Opportunities

- Biology

D-spacing range	1 – 16 (22) Å
Moderator	Coupled s-CH_4 grooved – 26K
Primary flight path	17m (m=3 supermirror guides)
Secondary flight path	3-7 m
Beam size	60(h) x 30(v) mm
Flux at sample	~10^7 n/s/cm^2

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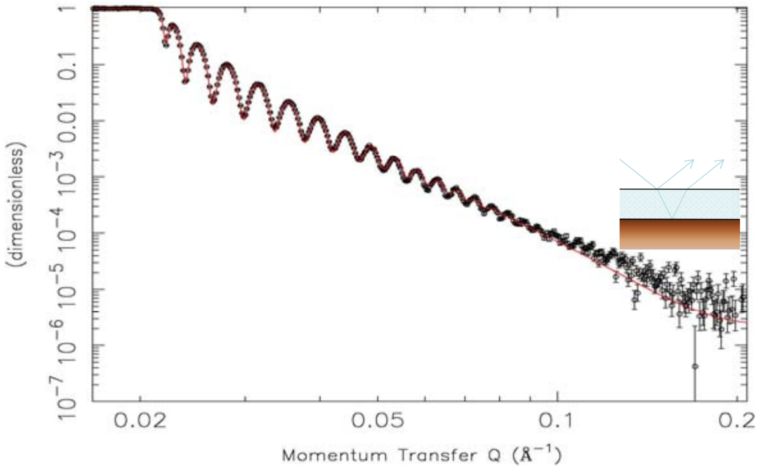
First Results 29 sep 08



First reflectivity measured on INTER. Nickel/Carbon film (1216 Å) on glass

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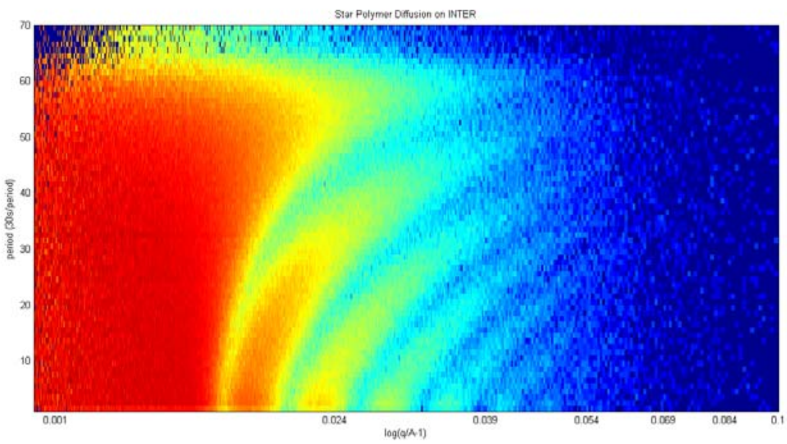
Kinetics



- Star Polymer
- D.G. Bucknall *(2010)*

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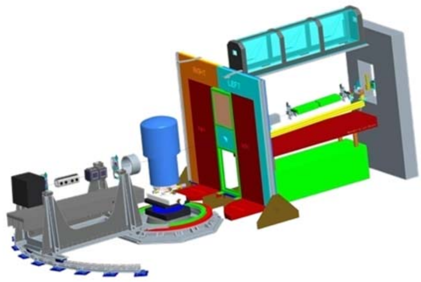
PolRef

- Uses polarised neutrons to study the inter an intra-layer magnetic ordering in thin films and surfaces
- >10 times the flux of CRISP
- Much wider dynamic range
- Flexible polarisation
- Dual Geometry
- High precision sample stage



Scientific Opportunities

- Spin Electronics
  - Spin-Injection
  - Spin Torque
  - Dilute magnetic semiconductors
  - Giant/Tunnelling magneto-resistance
- Model Magnetic Systems
  - Ultrathin films (finite size effects)
  - Exchange springs (domain walls, surface magnetic phase transitions)
  - Stabilise new single-crystal phases (Ce, Mn,..)

D-spacing range	0.9 – 15 Å
Moderator	Coupled s-CH_4 grooved – 26K
Primary flight path	23m
Secondary flight path	3 m
Beam size	60(h) x 30(v) mm
Flux at sample	~10^7 n/s/cm^2

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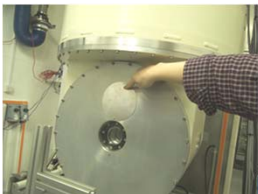
polref

- Moderator coupled grooved sCH_4
- Wavelength Range 1Å - 16Å
- Beam size 60 mm x 30 mm H x V
- Straight optics: no direct line of sight
- Flexible neutron polarisation
- 3D magnetic field
- Bham 17T magnet
- Dilution option
- Modes of Operation
  - Polarised and un-polarised
  - Vertical and Horizontal scattering geometries
  - 1(2)D detector



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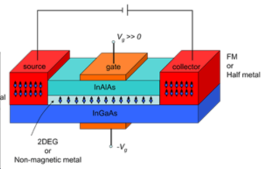
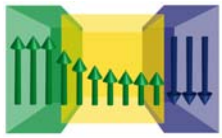
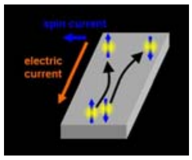
PNR Roadmap

System	Motivation	M (emu/cm³)	Degree of difficulty	Challenge
Transition, Fe, Co etc., and rare earth elements.	Exchange bias (spin valves) Exchange coupling (magnets)	10^3	"Easy"	To discern a weakly magnetic material in proximity to a strongly magnetic material. Highly polarized neutron beams are very desirable.
Complex oxides, LCMO etc.	Faster switching speed 10-100THz (orbital) vs. 1-100 GHz (spin)	10^2	Moderate	Dots, domains and patterned media are still possible to study.
(Ga,Mn)As	Spin FET⇒ ten-fold reduction in number of devices	10^1	Challenging	
Doped "non-magnetic" oxides, e.g. Cr-ZnO, nuclear polarization etc.	ditto	10^0	Tour de force	Instrument bias is a serious problem. Bias ~1 part in 1000 has been achieved under non-routine conditions.
Spin currents in nanostructures	ditto	10^{-2}	Presently unfeasible	Bias ~ 1 part in 10^8 has been achieved for nuclear physics experiments. Intense neutron beams an advantage.
Spin currents in Fe/GaAs FET's	ditto	10^{-4}	Presently unfeasible	

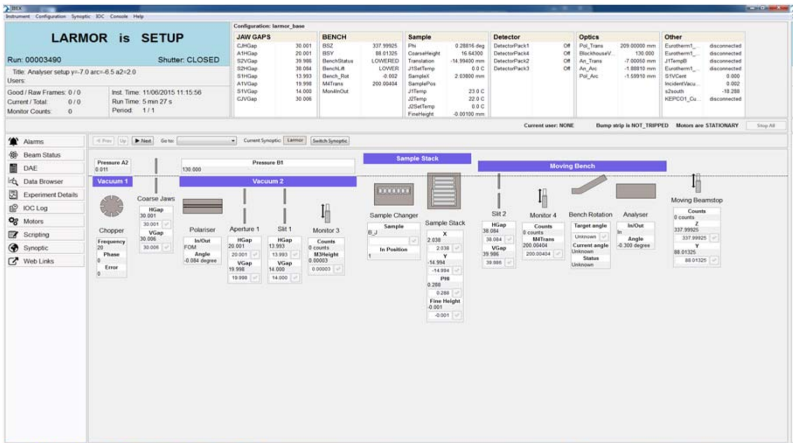






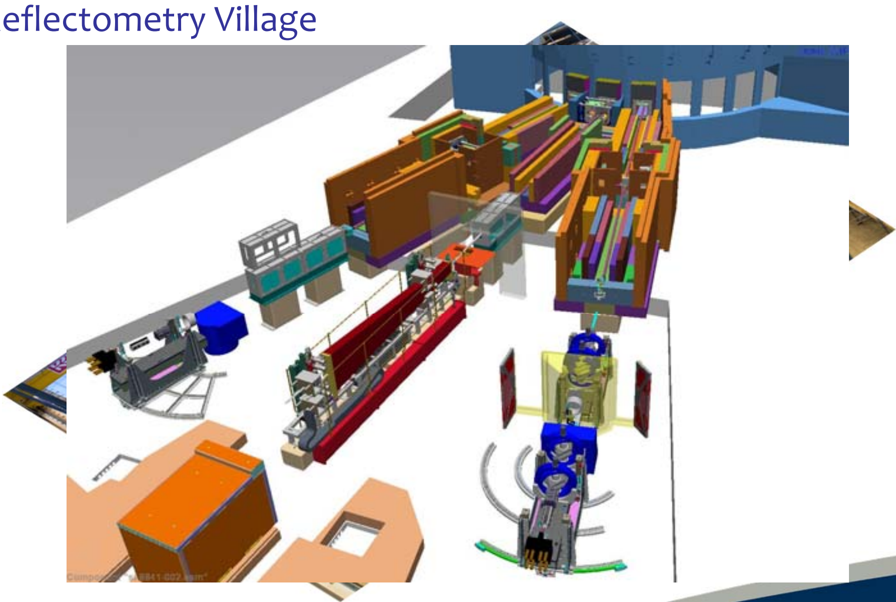
Adapted with permission, Fitzsimmons (ORNL)

IBEX Experiment Control



IBEX, the new EPICS based experiment control program, is being used on LARMOR, providing a powerful and flexible client-server based system

Reflectometry Village



blockhouse would be of the order of 5-7m wide at the sample

supermirror would be inserted into the beam in order to
the sample. It is envisaged that no more than 2 angles would
than that available on SURF and CRISP in three separate

nts beam
ısing a
permirror
rst set of
n would
elds with
larisation
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ıding on
ent. ^{3}He
ıce range
e x and y
d greater
stack of
ers. This
CRISP but does not allow analysis of the beam polarisation in

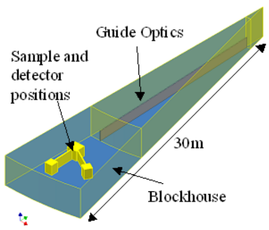


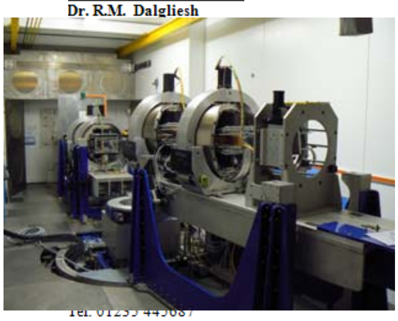
Figure 4: An illustration of the possible layout of the Offspec beamline (to scale)

reflected beam would be achieved using a method based on
l [7] illustrated in figure 6. The simplicity of the apparatus
performed with a monochromatic beam. For a polychromatic
would be replaced with either non-adiabatic coils or Dabbs
laced with appropriate, phased, pulsed magnetic fields. It is

Page 5 of 6

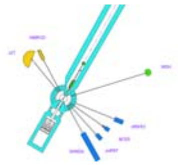
Be

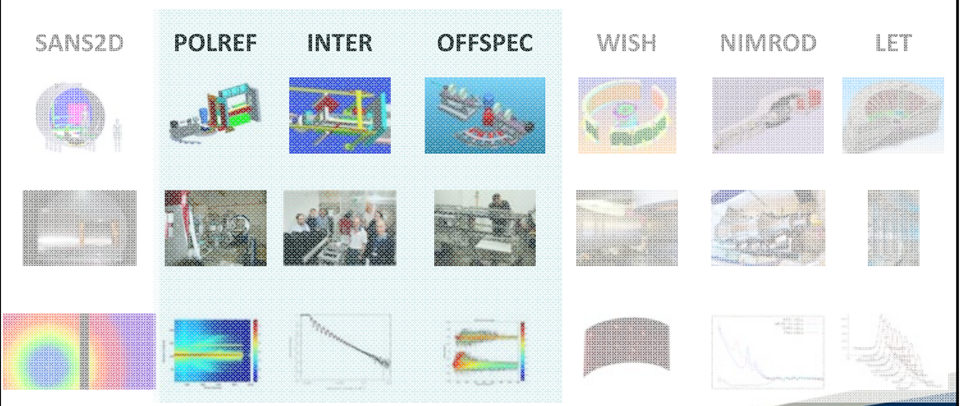
ISIS Contacts:
Dr. R.M. Dalgliesh



Fax: 01235 445720
Email: s.langridge@rl.ac.

TS2 Phase I instruments operational





TS2 Phase I: 2004-2009

Design Philosophy

- Optimised science driven design
  - Strong community involvement
- International Partnerships
- Complete integration
  - Target-Reduction
  - design-implementation-operation
- Need for state of the art computing
  - Simulation
  - Instrument control
  - Reduction software
- Integrated Instrument Spaces/Villages

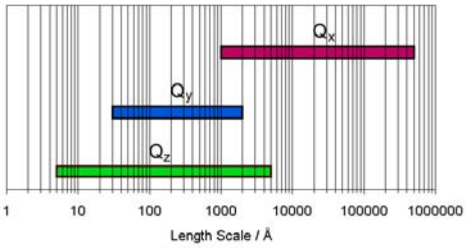
Instrument Concepts

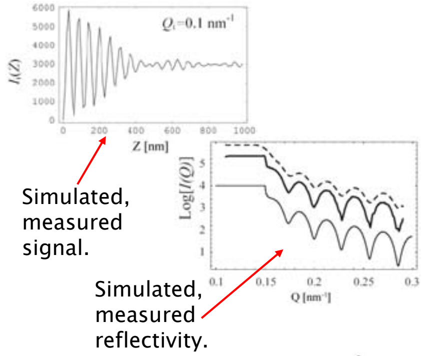
- Conventional
- Larmor Labelling
- Prism Encoding
- Pinhole geometry
- Divergence transport

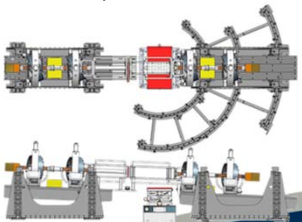
LARMOR LABELLING

Neutron Spin-Echo

In-plane dynamic range of 50Å–42μm







Scientific Opportunities

- **In-plane Structures**
  - Patterned Storage Media
  - Mesoporous films
  - Polymers
  - Biological membranes
  - Surfactants
- **Grazing Incidence Diffraction**
  - Surface crystalline structure
  - Surface phase transitions
  - Magnetic surface structure

TUDelft Delft University of Technology

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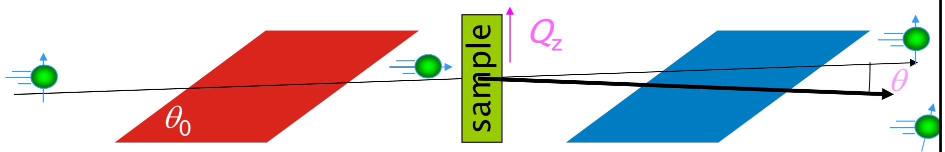
Larmor precession codes scattering angle



Unscattered beam gives spin echo (net precession) Independent of height and angle	$\phi = 0$
Scattering by sample over angle *q* results in a net precession	$\phi = c\lambda BL\cot(\theta_0)\,\theta \approx zQ_z$
Proportional to the **spin echo length** *z*	$z = c\lambda^2 BL\cot\theta_0/2\pi$
Measure polarisation	

$$\frac{P}{P_0} = \int d\Omega \cos\phi f(\phi) \approx \int dQ_x dQ_y dQ_z \cos(zQ_z) S(Q)$$

Keller *et al.* Neutron News **6**, (1995) 16
Rekveldt, NIMB **114**, 366 (1996)

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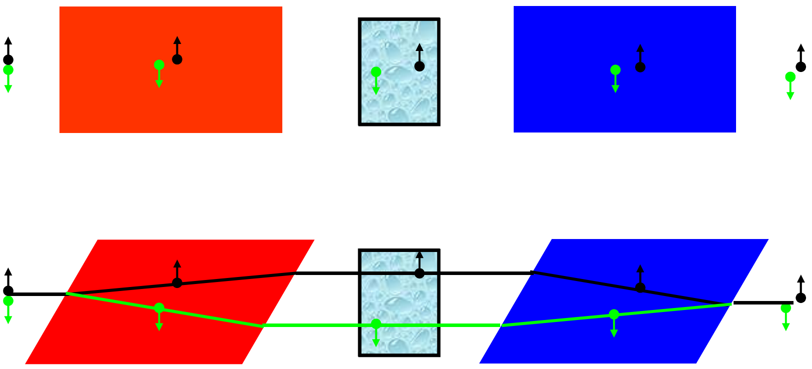
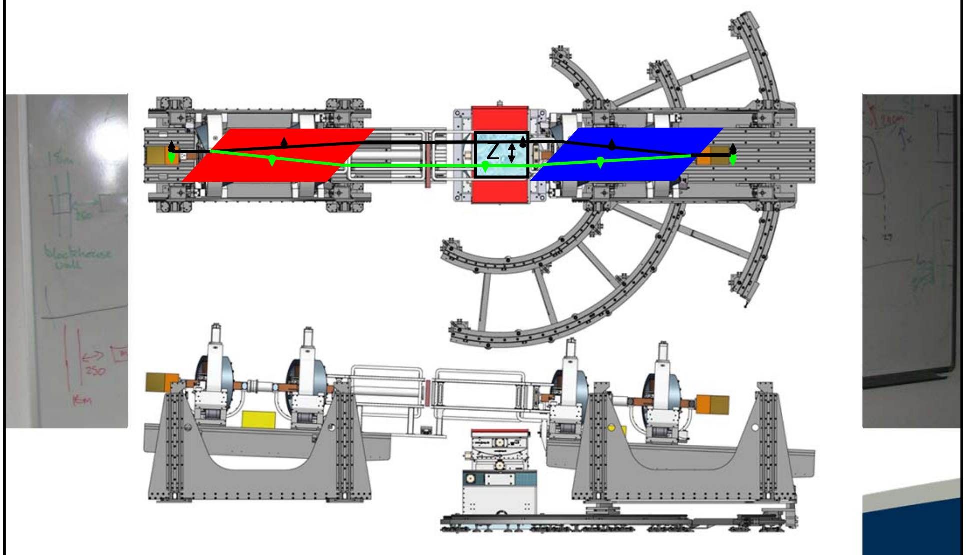
Spin-echo



Realisation/Design



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5 modes of operation

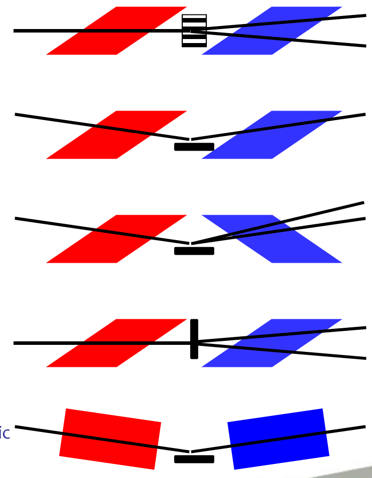
SE reflection measurements to probe in plane structure (SERGIS)

SE reflectivity with "high resolution" at low q and "wavy surface"

Spin-echo reflection "separation" of specular and off-specular reflection

Spin echo small angle scattering in transmission (SESANS)

Classical Spin echo in transmission or reflection of inelastic samples

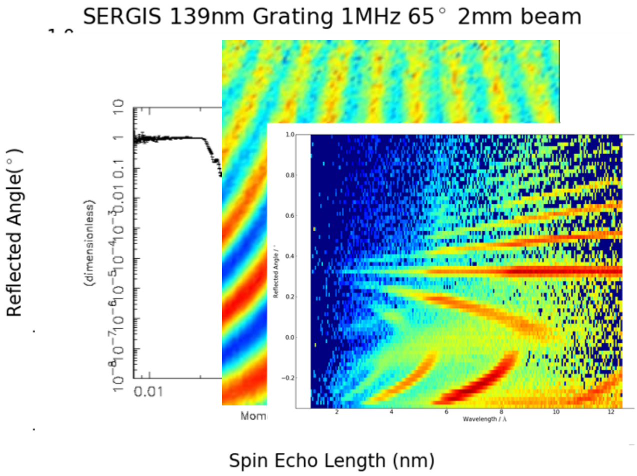


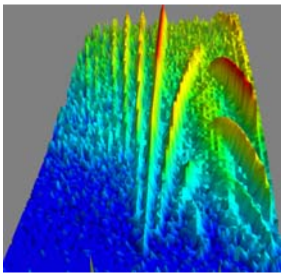
Bouwman, W. G. *et al.* Real-space neutron scattering methods. *Nucl. Inst. Methods Phys. Res. A* **586,** 9–14 (2008).

4th March 2009

Offspec

- High performance reflectometer
- First Spin Echo @ ISIS
- SESANS
- SERGIS
- Low background





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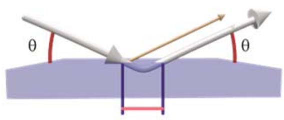
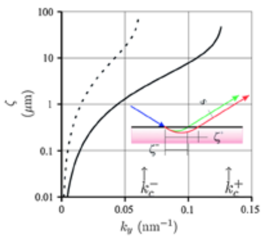
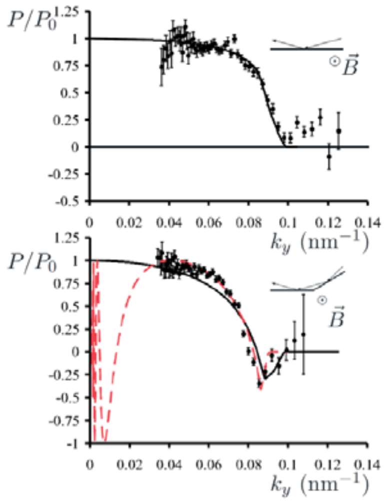
- Observation of G–H shift with neutrons







- de Haan, SL *et al.* prl **104**, 010401 (2010)
- de Haan, SL et al. **81**, 094112 (2010)

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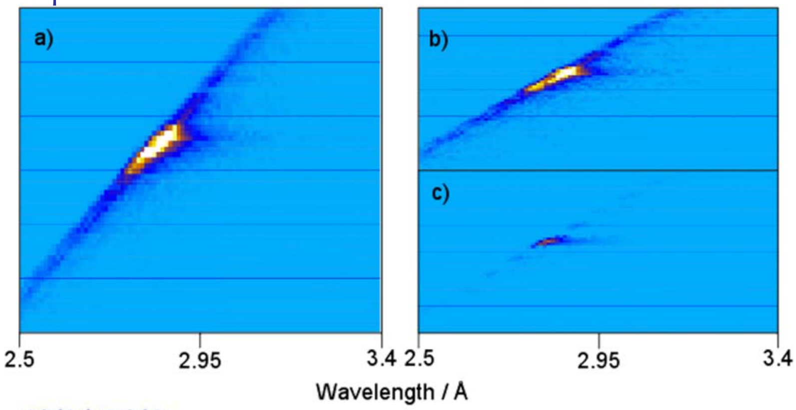
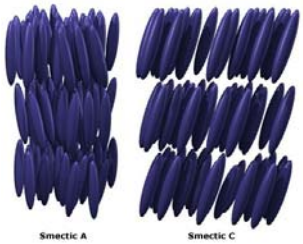
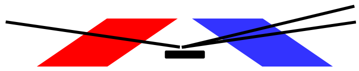
Off specular separation





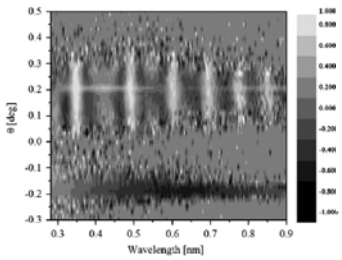


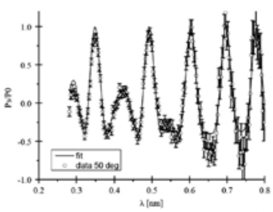
Courtesy of Richardson, Dalgliesh et a

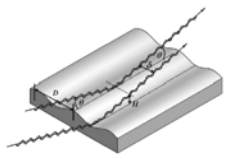
http://barrett-group.mcgill.ca/teaching/liquid_crystal/LC02.htm

SERGIS from buried gratings

- Take a model system and increase its complexity gradually.
  - 820nm commercial Aluminium grating deposited on glass. (Well understood and modelled)
  - d-PMMA polymer has a high scattering length density and should obscure the grating
    - Annealing the film should recover the grating pattern with modification.
  - By using a grating it is hope to amplify interface scattering signals from more complex systems which are of scientific interest.







J. Plomp SL, et al. Physica B 406 (2011) 2354-2356

Refractive Analysis of Interfaces with Neutron Beams Optimised for a White Spectrum
RAINBOWS

R. Cubitt & J. Stahn

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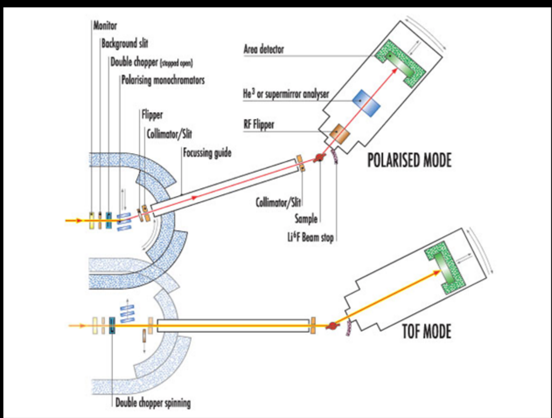
D17 – vertical sample reflectometer at ILL



Q range=0.005 Å^{-1} < Q < 4 Å^{-1}
Polarized neutrons+He3
Vertical samples only
TOF and Mono

http://www.ill.eu/instruments-support/instruments-groups/instruments/d17/

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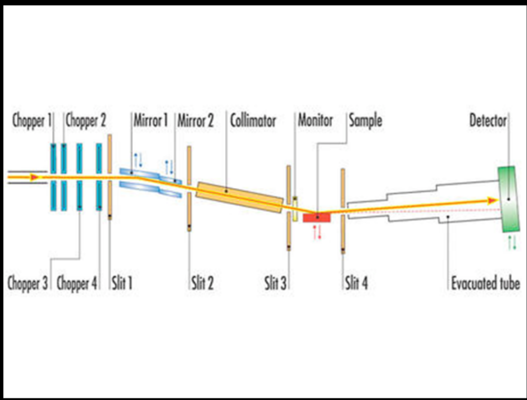
FIGARO – horizontal sample reflectometer at ILL



Q range=0.005 Å^{-1} < Q < 0.4 Å^{-1}
Horizontal samples
Free liquids
Liquid/liquid interfaces
TOF

http://www.ill.eu/instruments-support/instruments-groups/instruments/figaro/

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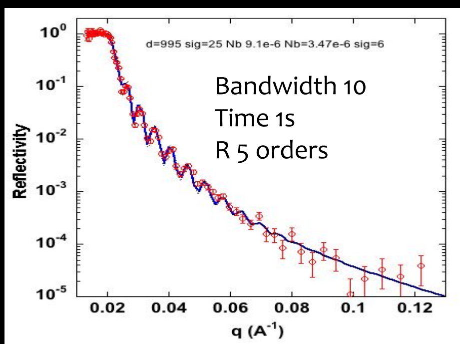
D17 ILL



The challenge - how to measure reflectivity even faster to enable new science

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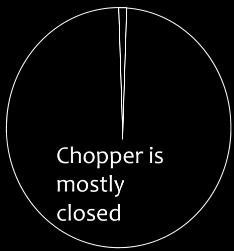
Choppers vs. Prism



Beam is only truly open on D17 for an average of ~8 minutes a day!!

Transmission 2-20 Å: 0.3-2 %
5% resolution at 2 Å

Cost~€200,000

MgF_2 Transmission 2-20 Å: 80-20 %
5% resolution at 2 Å

Cost~€2000

Theoretical gain with prism @2 Å: 240 X

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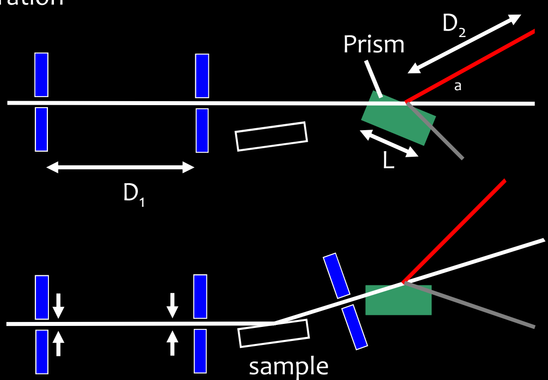
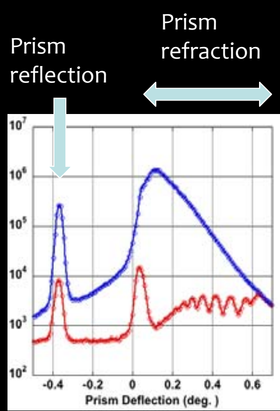
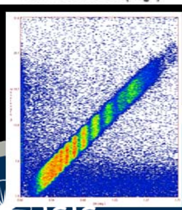
Reflectometry experiment using a MgF_2 prism

Only need to know Nb of prism for calibration







Price-no longer get off specular for free

R. Cubitt & J. Stahn Eur. Phys. J. Plus 126 111 (2011)

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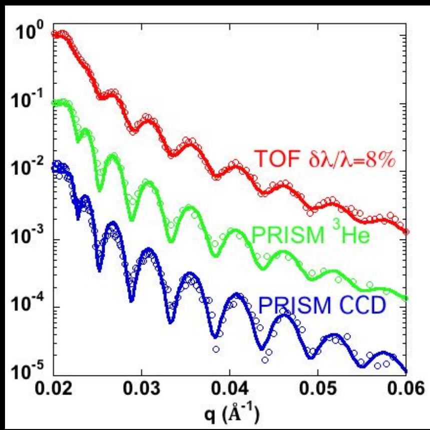
Prism with a high resolution CCD camera-AMOR PSI



Elliptical guide

R. Cubitt & J. Stahn Eur. Phys. J. Plus **126** 111 (2011)

Ni/Glass sample as before

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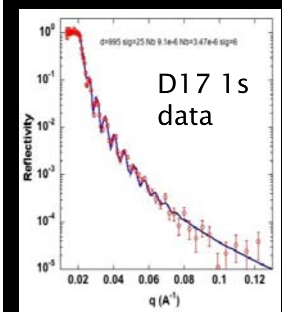
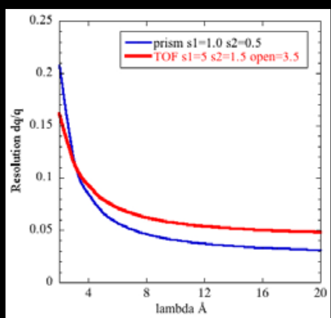
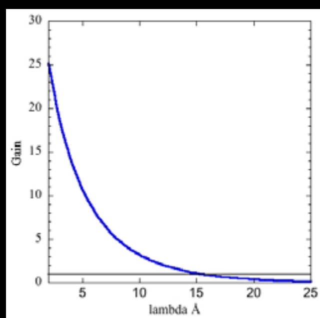
Gain over TOF at Similar Resolution







16 Å 5.4 Å 2.7 Å

Better resolution above 4Å

where we need it

Statistical limit at high q

Should be able to measure

same sample in 50ms

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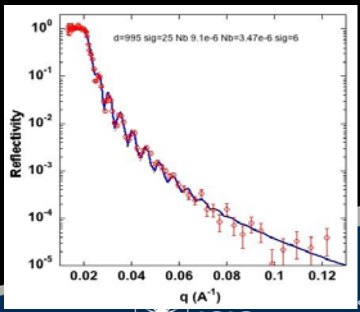
Conclusions

A refractive encoding machine promises great gains in performance for little financial investment enabling new science in kinetics or small samples.

Ideal for intermediate resolution

Technically simple and relatively cheap instrument.

Proven technique- should be able to measure 5 orders of reflectivity in ~50 ms!



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Coherently Summing Reflectometry Data

- R. Cubitt
- P. Gutfreund
- T. Saerbeck
- R. Barker
- G. Fragneto
- R. Campbell

Original idea with monochromatic beams and flat samples:
Hamilton W. A., Hayter J. B. & Smith G. S. (1994) *J. Neutron Research* **V2** N1 1-19

ESS Estia instrument project exploiting this idea: J. Stahn

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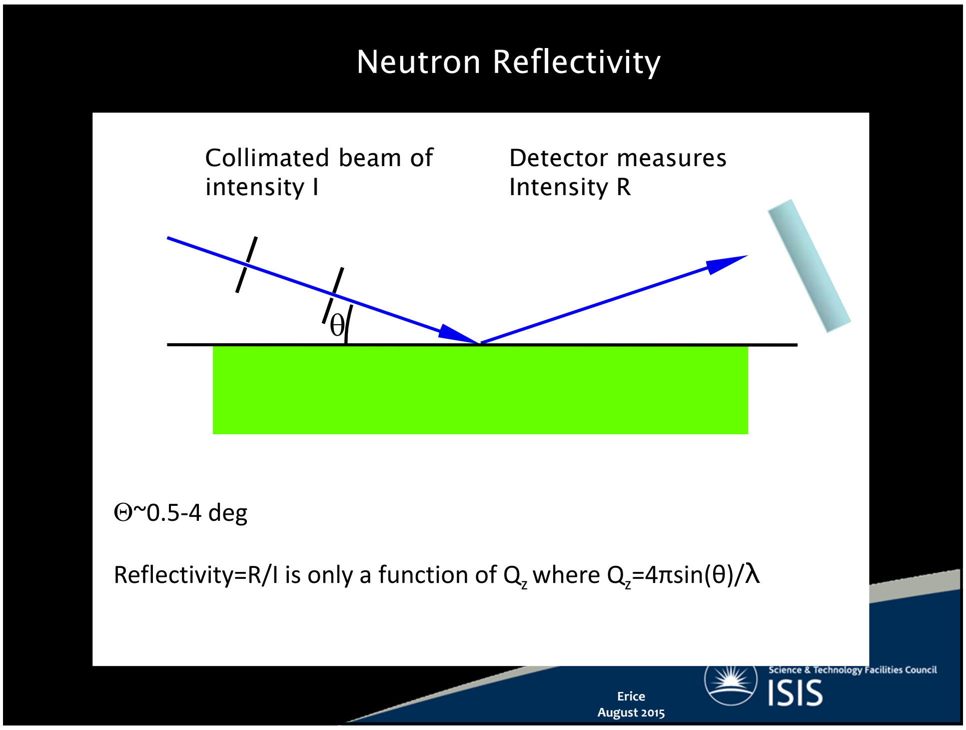
Neutron Reflectivity
Collimated beam of intensity I
Detector measures Intensity R
θ
Θ~0.5-4 deg
Reflectivity=R/I is only a function of Q_z where $Q_z=4\pi\sin(\theta)/\lambda$
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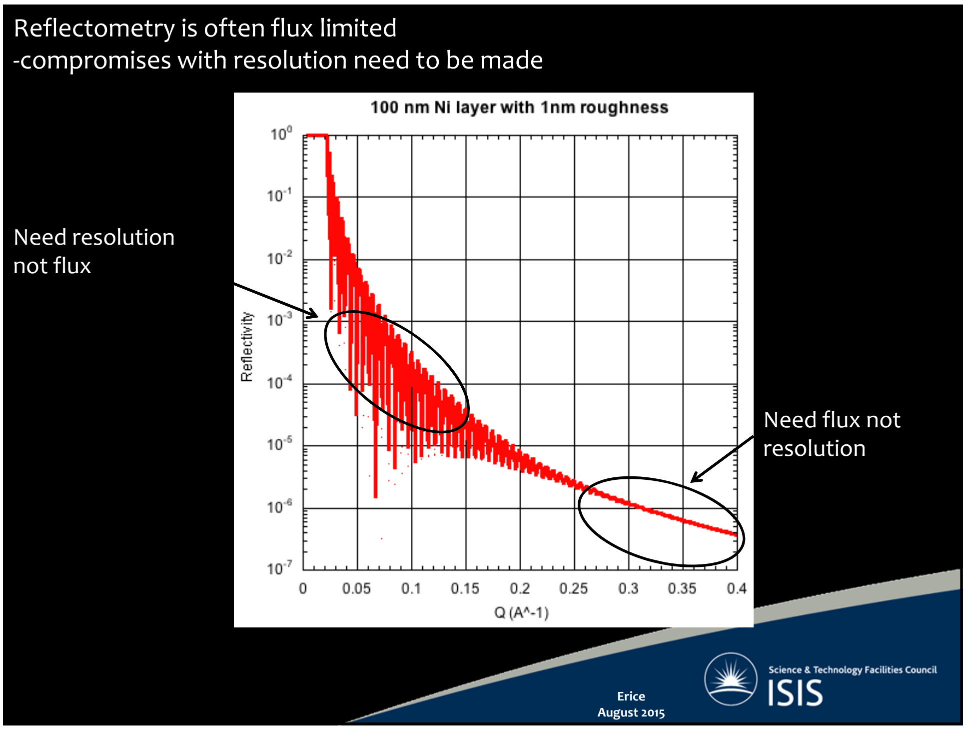
Reflectometry is often flux limited
-compromises with resolution need to be made
100 nm Ni layer with 1nm roughness
Need resolution not flux
Need flux not resolution
Reflectivity
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
0 0.05 0.1 0.15 0.2 0.25 0.3 0.35 0.4
Q (A^-1)
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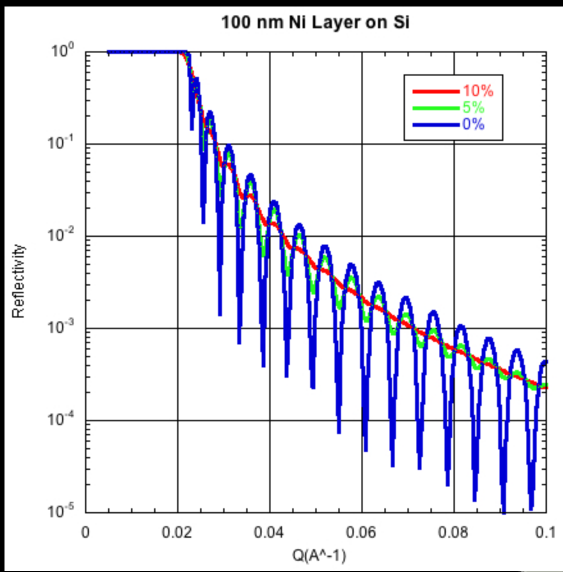
Resolution in Q_z



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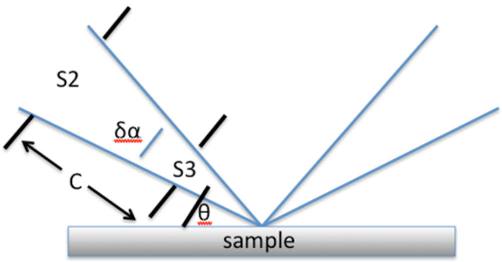
Uncertainty in Q_z

$Q_z = 4\pi \sin(\theta)/\lambda$

$(\delta Q_z/Q_z)^2 = (\delta\theta/\theta)^2 + (\delta\lambda/\lambda)^2$ (for small angles)

Range of incoming angles $(\delta\alpha)^2 = (0.68*S2/C)^2 + (0.68*S3/C)^2$

$\delta\theta = \delta\alpha$



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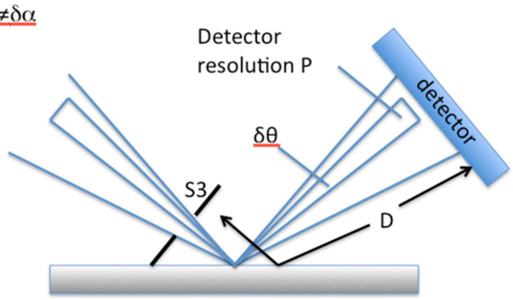
Uncertainty in Q_z

$Q_z = 4\pi \sin(\theta)/\lambda$

$(\delta Q_z/Q_z)^2 = (\delta\theta/\theta)^2 + (\delta\lambda/\lambda)^2$ (for small angles)

Uncertainty in θ $(\delta\theta)^2 = (0.68 * S3/D)^2 + (P/D)^2$

$\delta\theta \neq \delta\alpha$



Better provided $(\delta\alpha)^2 > (s2/D)^2 + (P/D)^2$

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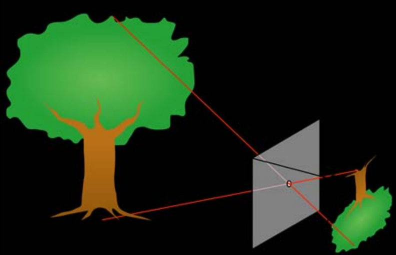
Pinhole camera



© Paul Debois "Pinhole Impressions" - Portfolio Series
International Garden Photographer of the Year Competition.

The resolution is not determined by the angular size of the source but the combination of the pinhole diameter and film (detector) resolution

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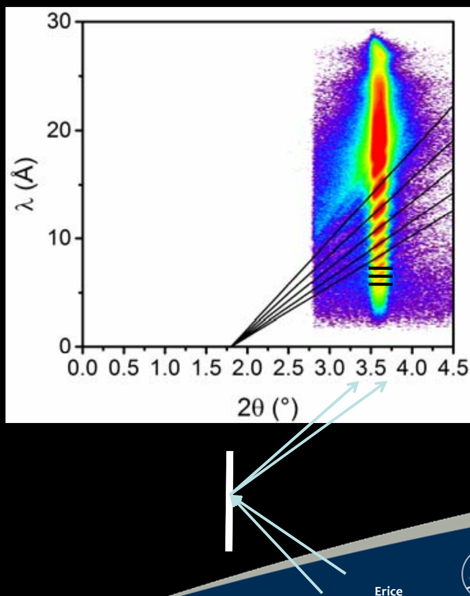
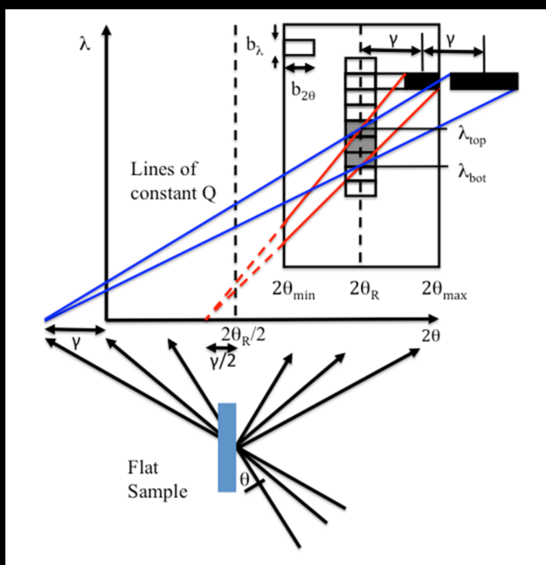
Divergent beam on a flat sample 1000Å Ni



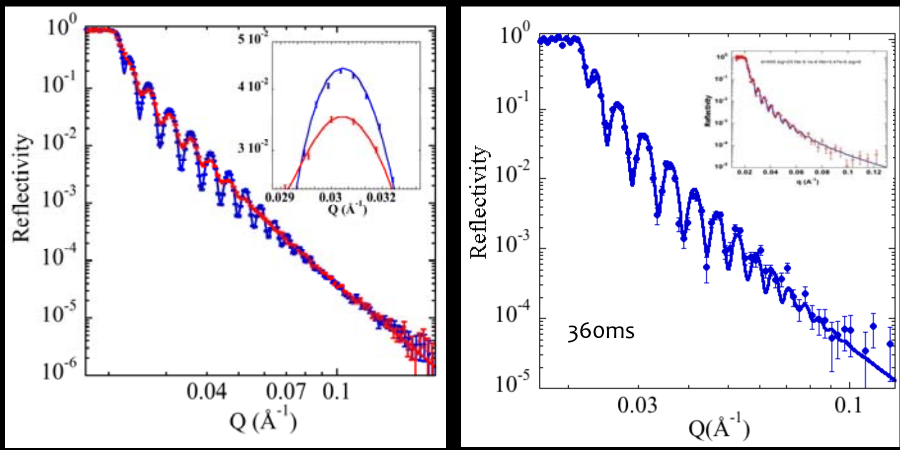
Conventional incoherent summing involves adding the reflectivities from the same wavelengths leading to a loss of resolution

Loop over all pixels and share the reflectivity between pixels of the same Qz



Loop over all pixels and share the reflectivity into all the pixels on a chosen 2θ line having a virtual wavelength of the same Q_z

Improved resolution limited only by the detector



Using only 10 mm of the 30 mm beam available on D17
-120 ms possible!

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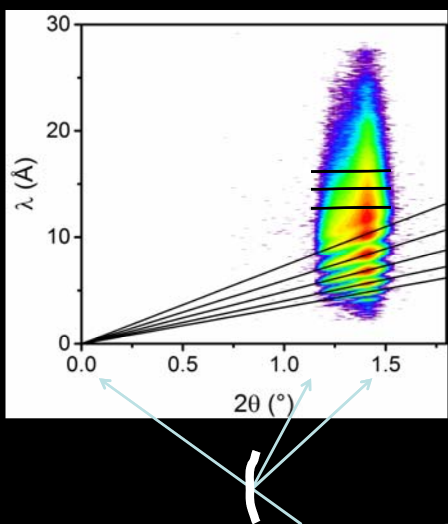
Bent samples can also have a large range of θ



SiO_2 and amorphous carbon both 1000 Å thick
separated by a SiOCN layer of 160 Å

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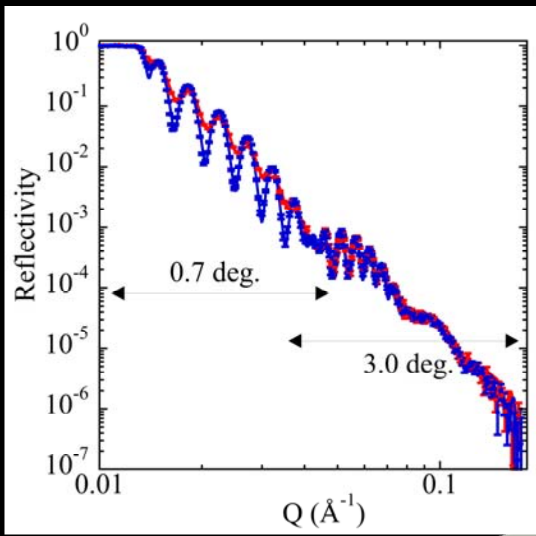
Significantly improved resolution



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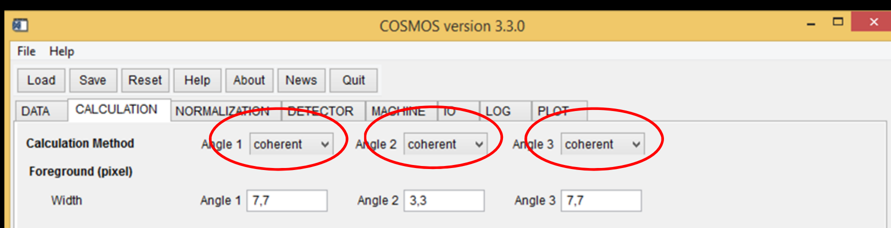
Conclusions

- Improved software reduction for divergent beams and bent samples enables kinetics ~120 ms time resolution.
- Equivalent to significant instrument flux increase >10x
- Strong need for higher resolution detectors
- Use it whenever the signal on the detector is wider than the resolution
- Incorporated into ILL data reduction software COSMOS



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LPSS gains

- High power → High flux
- Pulse width
- Longer instrument → Lower backgrounds
  - Increased space
    - Polarisation without loss
    - Pulse Shaping
    - Inelastic-elastic separation
- 10^{-8} from cm^2 sample

Outline Layout

- Comparison of possible options
  - 16.6Hz, 30-40m, 2ms
  - 6Å, $\Delta\lambda$=0.2Å
  - 5% resolution
- Must be out of line of sight
- Switchable polarised/non-polarised option
- Moderator
- Guide Size
  - Bender
  - Square guide
    - H/V scattering geometries
  - Background minimisation
- Performance Metric
  - IntFlux x λ^4
- 60m Instrument "tuneable"

Fast Reflectometer for Extended Interfacial Analysis

Hanna Wacklin (ESS/UCPH)
Anette Vickery (UCPH/DTU)

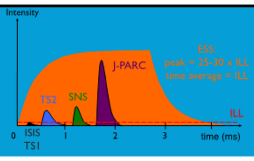
Freia, (Frejya, Freyia, Frøya, Frøjya, and Freja) in Old Norse the "Lady", one of the Vanir gods, rules over the heavenly afterlife field Fólkvangr and there receives half of those that die in battle.

www.europeanspallationsource.se

21st May 2014

The Challenge

Experiments will be x 10 faster than today!

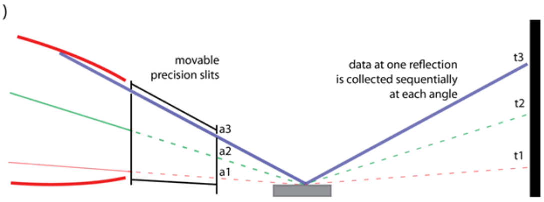


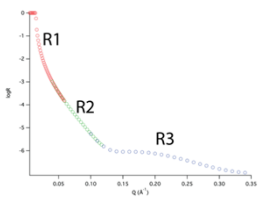
- Need to think of how experiments will be done
- Need to measure 2-3 angles of incidence to cover Q-range
- Moving the instrument between angular settings takes up to 1min.
- This becomes a significant issue at ESS as measurement times decrease
- Can experiments be done without moving the sample/instrument?

The FREIA Solution

- Deliver a broad angular divergence focused onto the sample surface
  - angle selection by moving slits only

1)

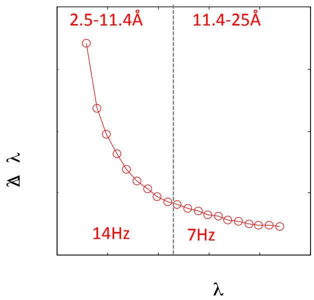


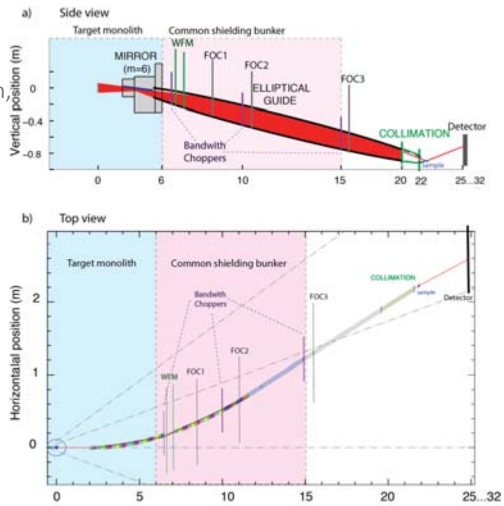


Instrument layout

- Inclined elliptical guide (-2°) to access horizontal surfaces

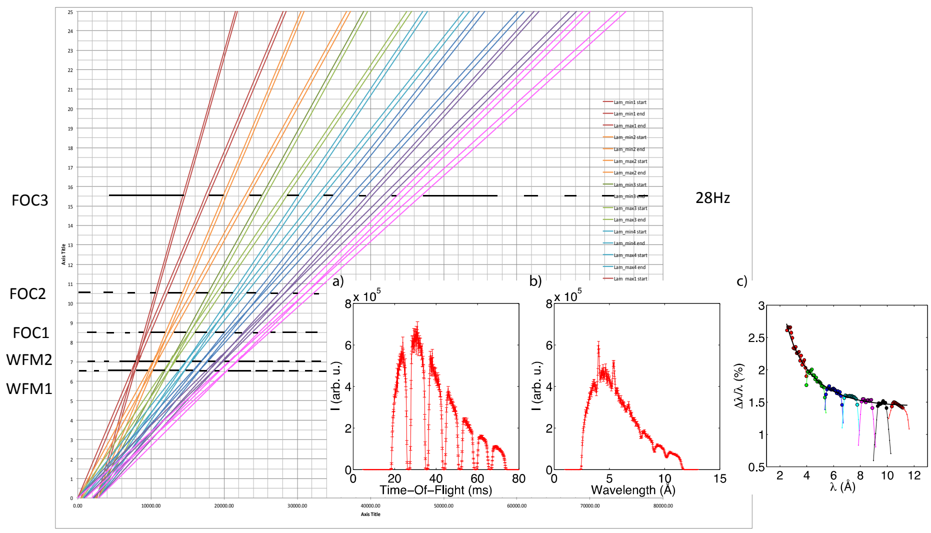
- 4° of vertical divergence transported
- 9.5m bender to eliminate fast neutrons
  High flux mode: dλ/λ = 3-17%
- choppers for wavelength band selection, pulse-skipping and high resolution.
- 25m instrument - Broad bandwidth
- Full science case covered in 1st frame





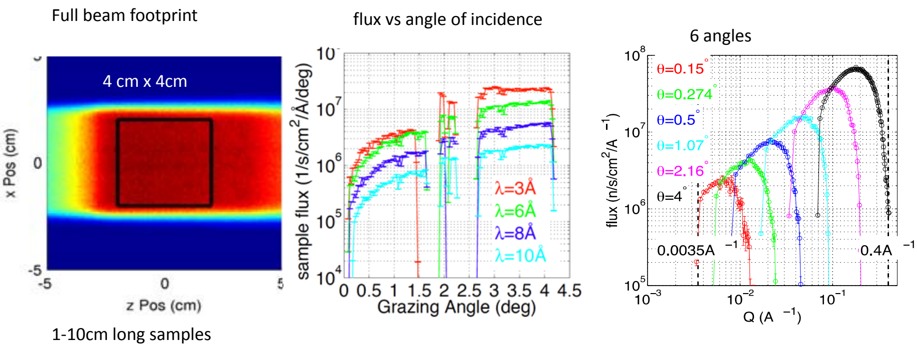
High-resolution option

- 7-fold Wavelength Frame Multiplication with dλ/λ = 1.5% (FWHM)



Beam characteristics

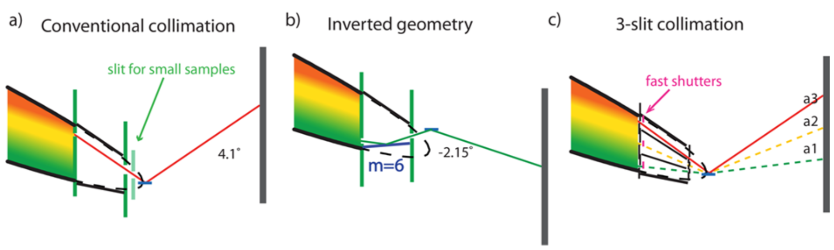
- Sample size: up to 5cm x 10cm for weakly scattering samples
- 4° of vertical divergence, 0.005Å^{-1} < Q < 0.45Å^{-1} on liquid surfaces
- flexible choice of angles mitigates
- 3 angles with good overlap
- more angles to minimize counting time in overlap regions for e.g. small samples



Collimation Options :

- Interchangeable guide sections (2m) for different experiments:

a) conventional slit collimation with extra slit for small samples
b) deflection mirror for inverted geometry (liquid-liquid interfaces, sample environments)
c) three-slit collimation + fast shutters for kinetics at the ms-s timescales

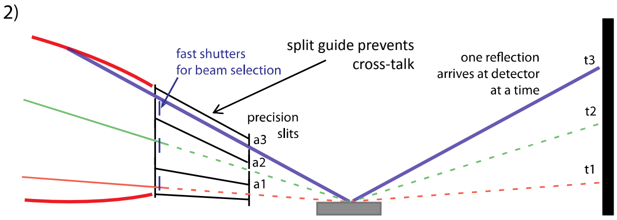


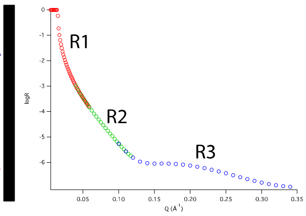
+ Optional horizontal focusing for small samples
+ GISANS collimation: 8m, 6.4m, 4m, 2m
+ Polarisation option: S-bender before first slit

Fast kinetics set up

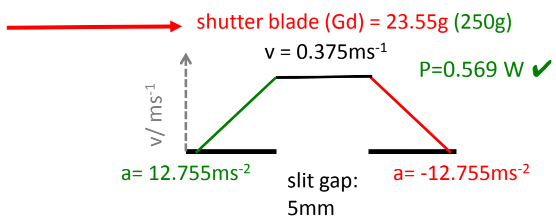
- Selection of angles as function of time
  – three pairs of precision slits + fast shutter mechanism to select open slit as function of time

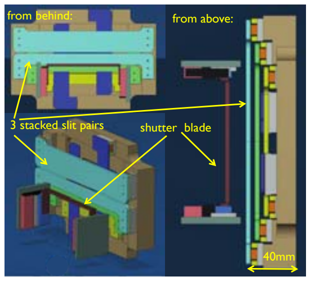
2)





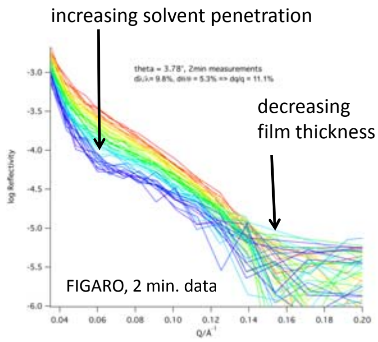
- selection 1 or more pulse per angle
- Full Q-range (0.0065 < Q < 0.4Å⁻¹) recorded "simultaneously"

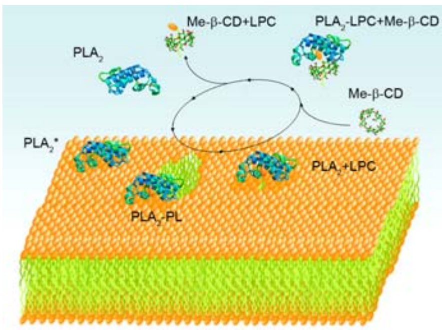




Summary

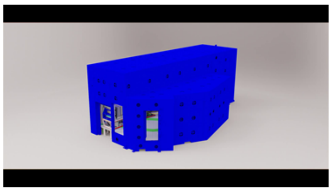
- FREIA will be a versatile reflectometer that will excel at 75% of the science
- measurement/resolution options for up to 90% of all experiments
- gains of at least 25-30 in counting time for most experiments
- operates without moving the sample
- fast kinetic studies with a very broad dynamic q-range
- ms-s time resolution for " one-shot" experiments over whole Q-range





Summary

- Science driven design
- Flexible design, but not too flexible
- Key performance indicators
  - Flux
  - Background
  - Dynamic range
  - Sample environment
  - Software



Dudley Moore: Do you feel you've learnt by your mistakes here?
Peter Cook: I think I have, yes, and I think I can probably repeat them almost perfectly.

Erice
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