Neutron Reflection and Reflectometers

Robert Dalgliesh

Summary

- A Brief History
- What can reflectometery tell us?
- Science Themes
- Current Instrumentation
  - Reactors
  - Spallation Sources
- Primary Instrument Design Criteria
- Major Components
- Sources of Background
- Polarisation
- Ancillary Equipment

A Brief History

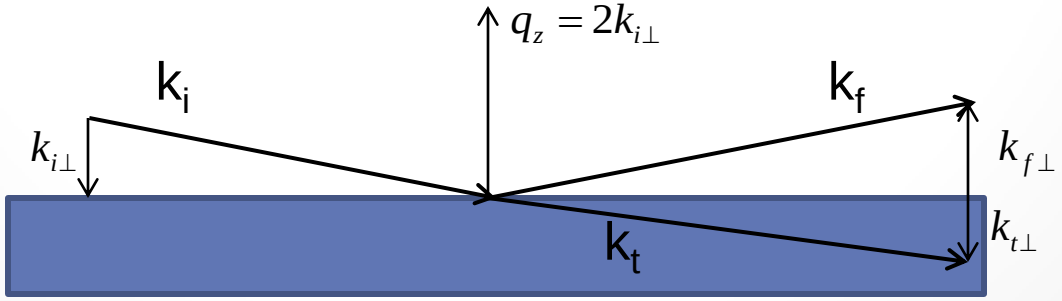
- 1946 - First observation of total external reflection of neutrons by Fermi and Zimm.
- 1947 - Fermi and Marshall performed detailed studies using monochromatic neutrons to measure scattering lengths of materials.
- Neutron Reflection then continued to be used to study bulk scattering lengths.
- 1976 - Hayter, Williams and Penfold used a modified IN11 at the ILL to measure interference fringes from a CoFe film on glass. (Nature, **262**, 569).
- 1981 – Hayter et al. Produced detailed simulations of thin film systems of interest to surface chemists. (J. Chem Soc. Faraday Trans. **77**, 1437-1448)
- 1981 – G.P. Felcher start works on magnetic thin films and begins building prototype TOF instruments at IPNS with polarisation options.
- Reflection experiments continue at reactors using monochromatic beams and θ-2θ geometry instruments.
- 1986 – CRISP is constructed at ISIS as a purpose built TOF reflectometer able to routinely study liquid surfaces.
- 2012 – 20+ instruments in operation world wide optimised for a huge variety of science

What can reflectometry tell us?

- Specular Reflectivity
  - A finely collimated beam of neutrons incident on an optically flat interface at grazing incidence will reflect from the surface in an analogous way to light.
  - Provided the scattering vector is small enough that it is a long way from satisfying a crystalline Bragg condition the material may be described using a bulk refractive index which is related to the Scattering Length Density (SLD).

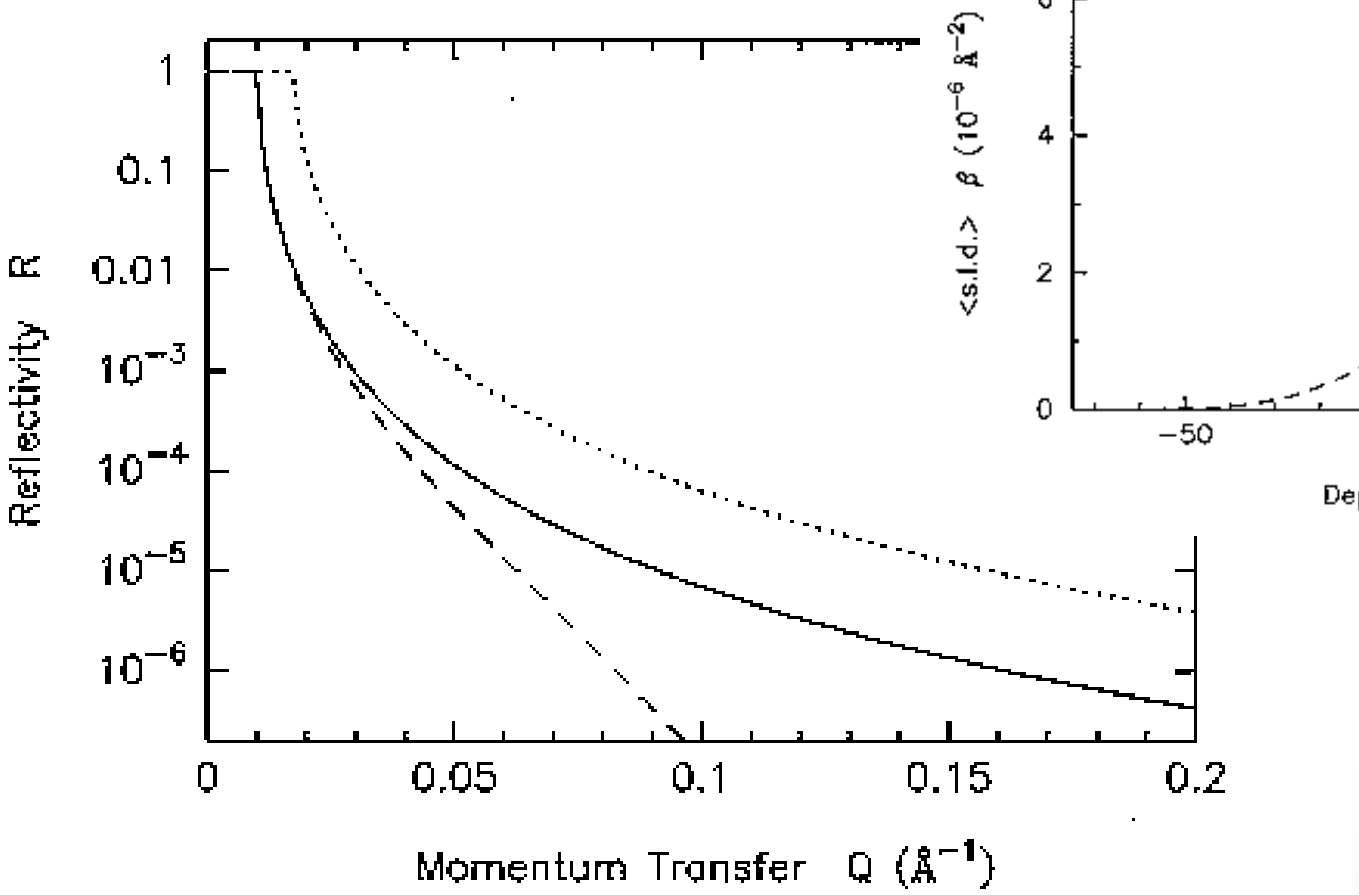
$$n \approx 1 - \frac{\lambda^2}{2\pi}\sum_i b_i N_i \qquad SLD = \sum_i b_i N_i$$

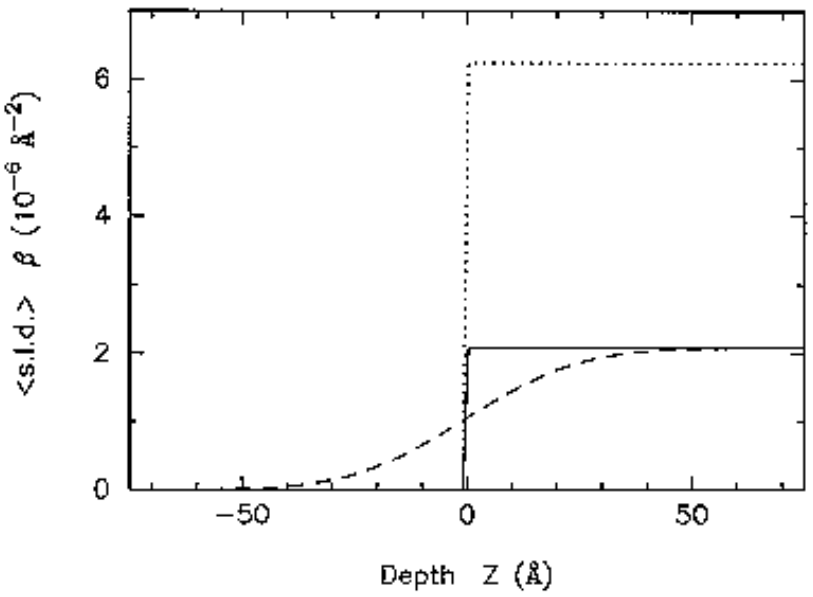
  - b is the scattering length and N the number density of a given atomic component and λ is the incident neutron wavelength.
  - The reflectivity, measured as a function of wavevector transfer perpendicular to the interface (q_z) be calculated using multilayer slab models of the SLD. Depending on the experimental design and instrument information can be obtained about length scales from 5-3000Å.



Specular Reflectivity

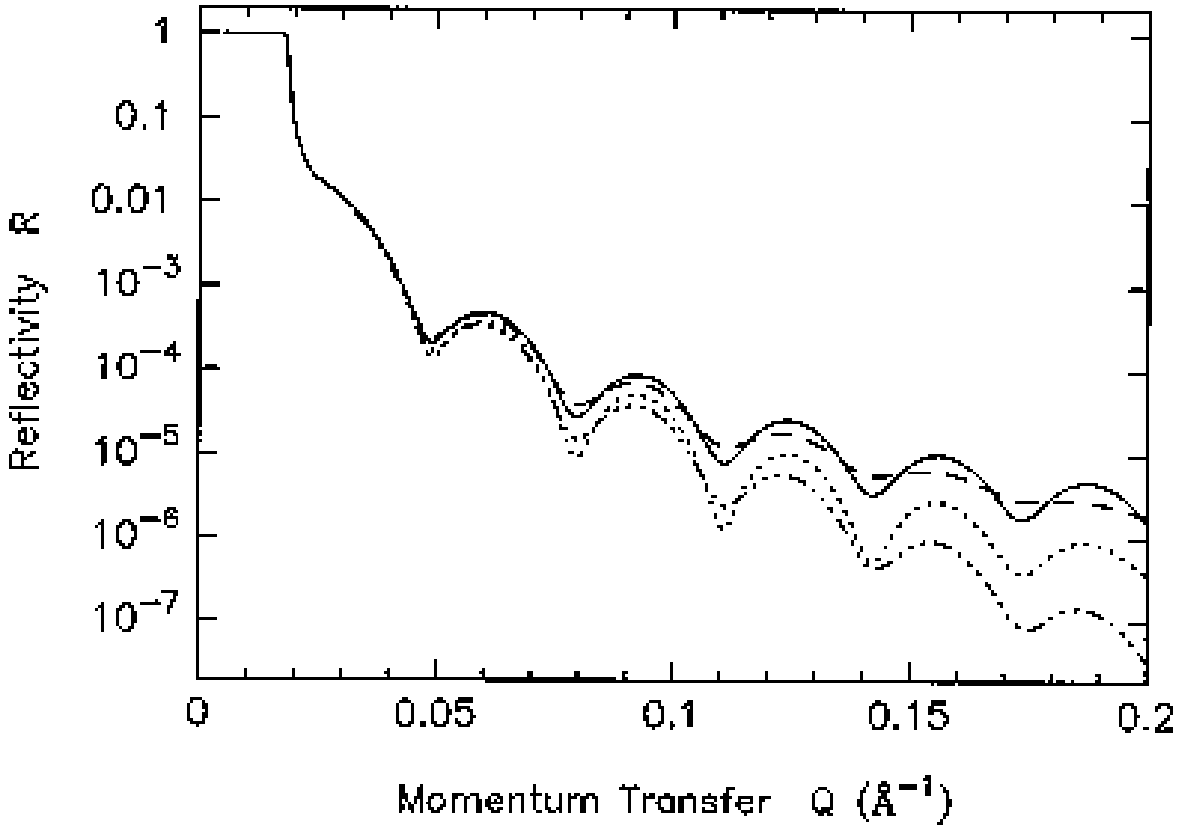
- Surface roughness and scattering length density changes are readily observed.

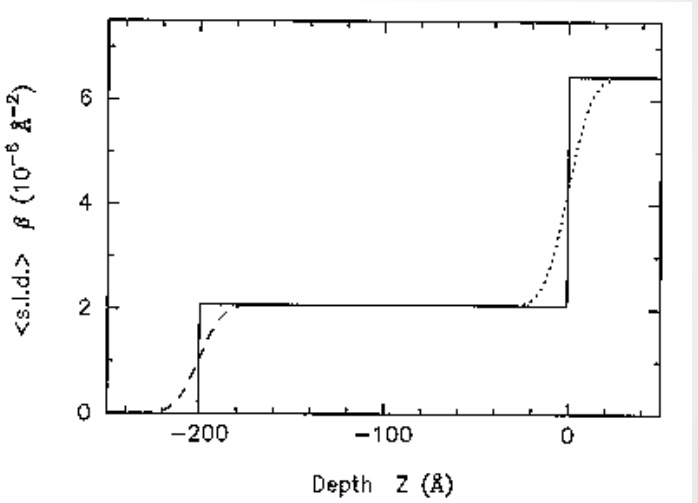




Speular Reflectivity

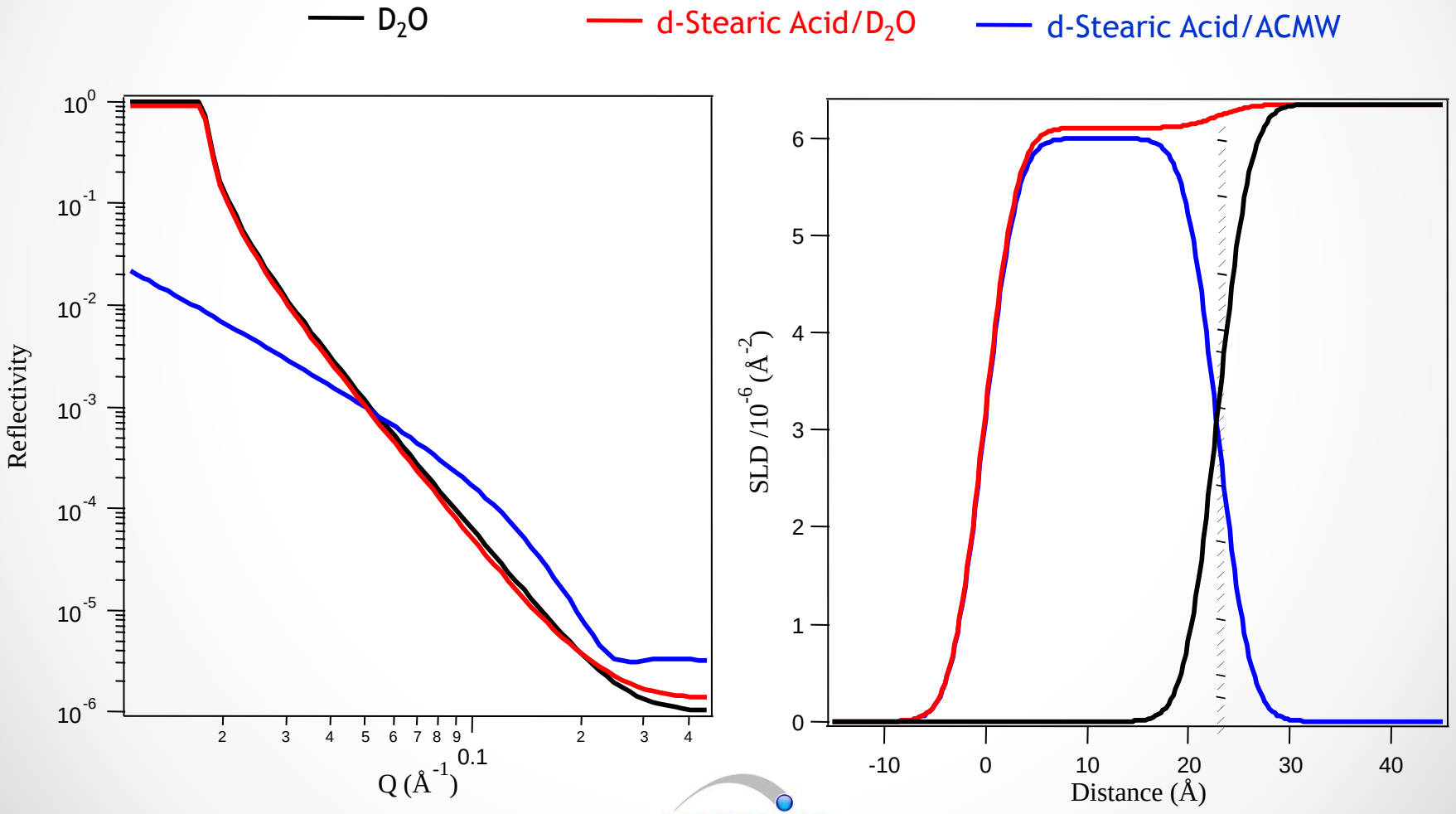
- Thin films at the interface lead to interference fringes.
- Interfacial roughness makes lie more complicated





Specular Reflectivity

- Isotopic substitution (often using H and D) allow simultaneous fitting, may mitigate some phase problems, and allows detailed studies of soft matter.

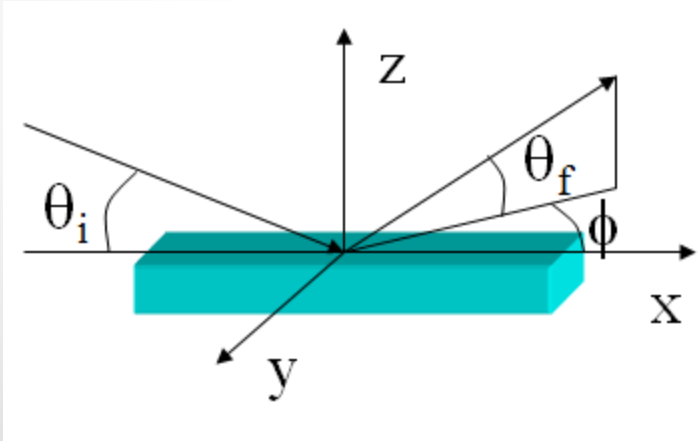


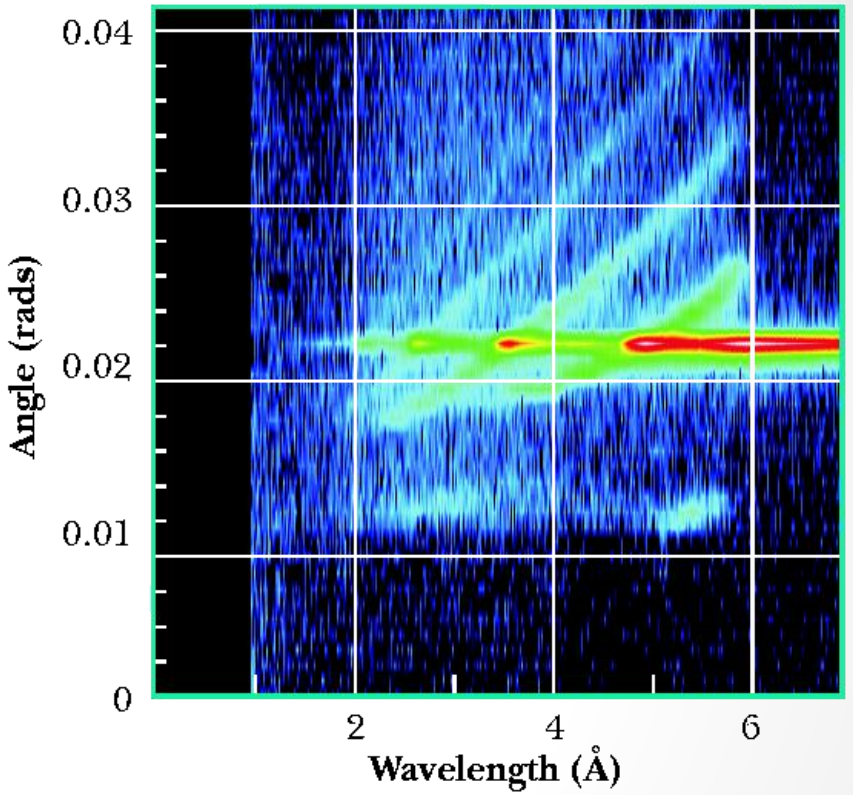
Off Specular Reflectivity

- Any Scattering that does not satisfy:

  $\theta_i=\theta_f$ and $\phi=0$

- Reveals information about in-plane structure.

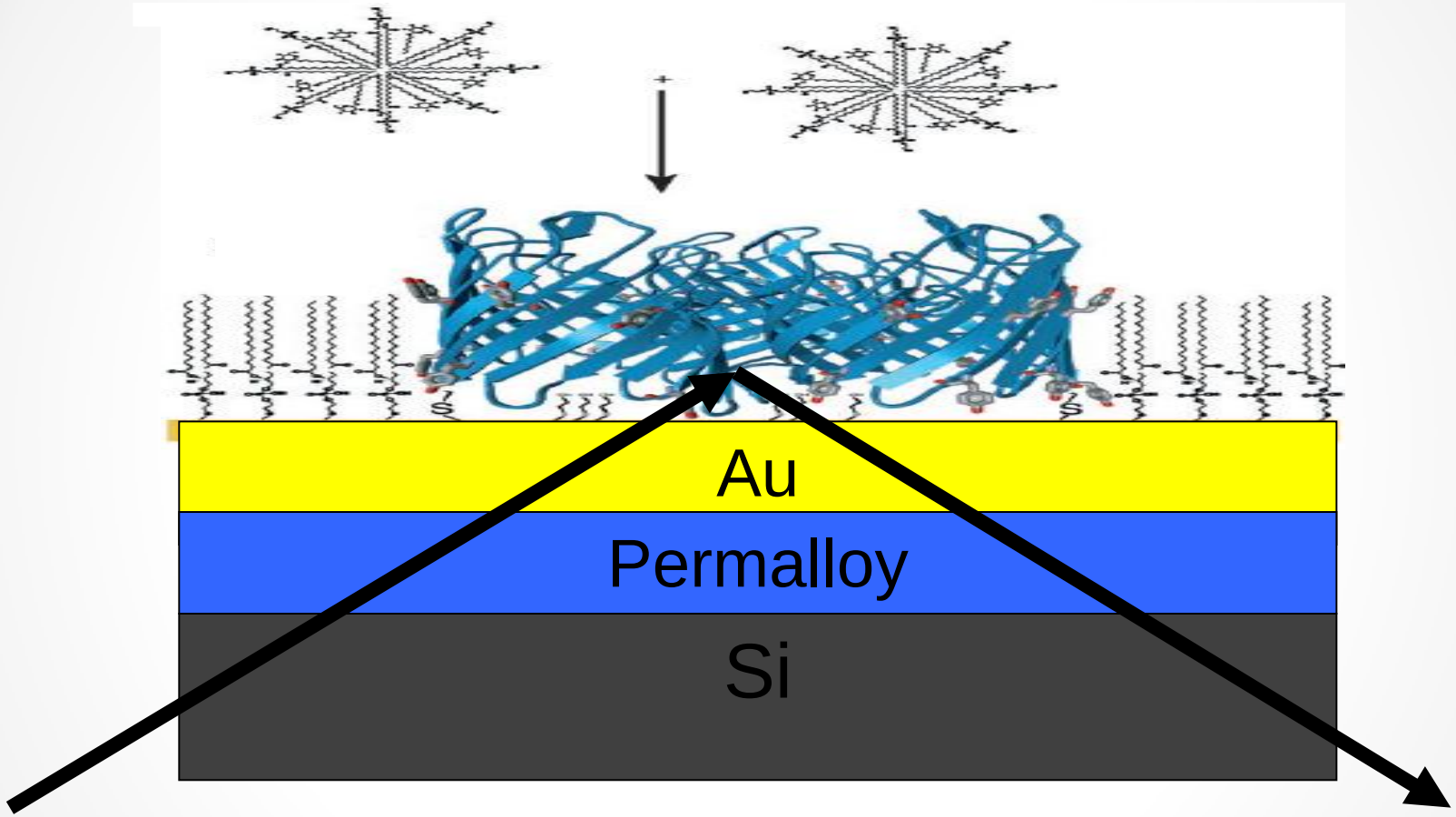




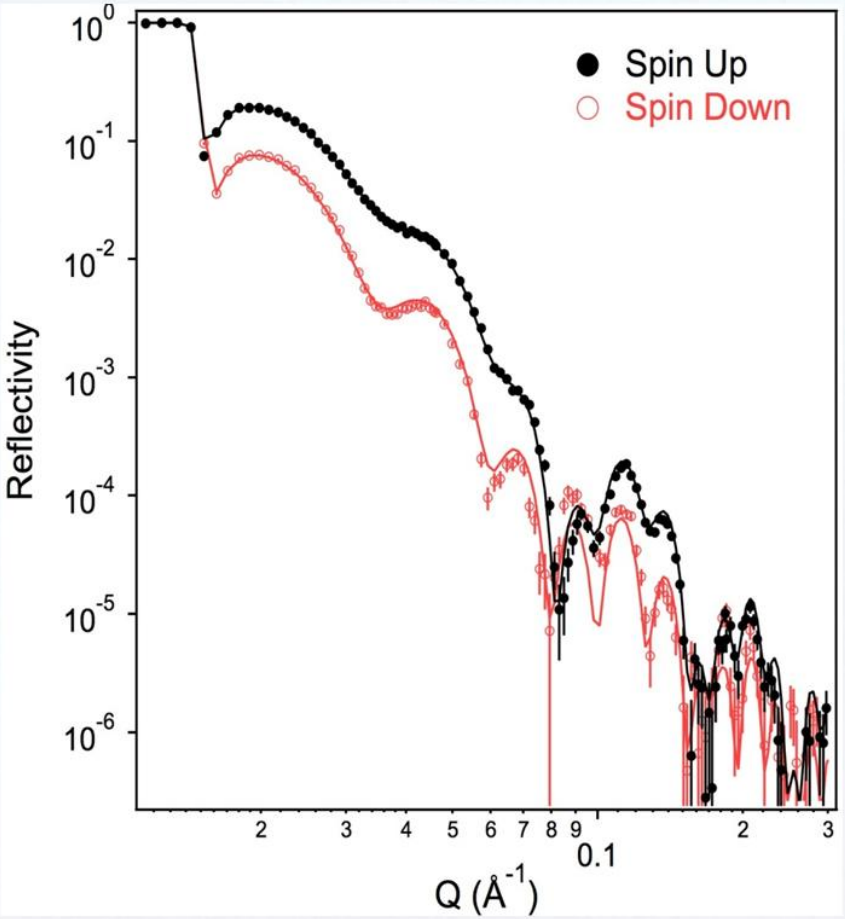
1% h-DDAB on D_2O

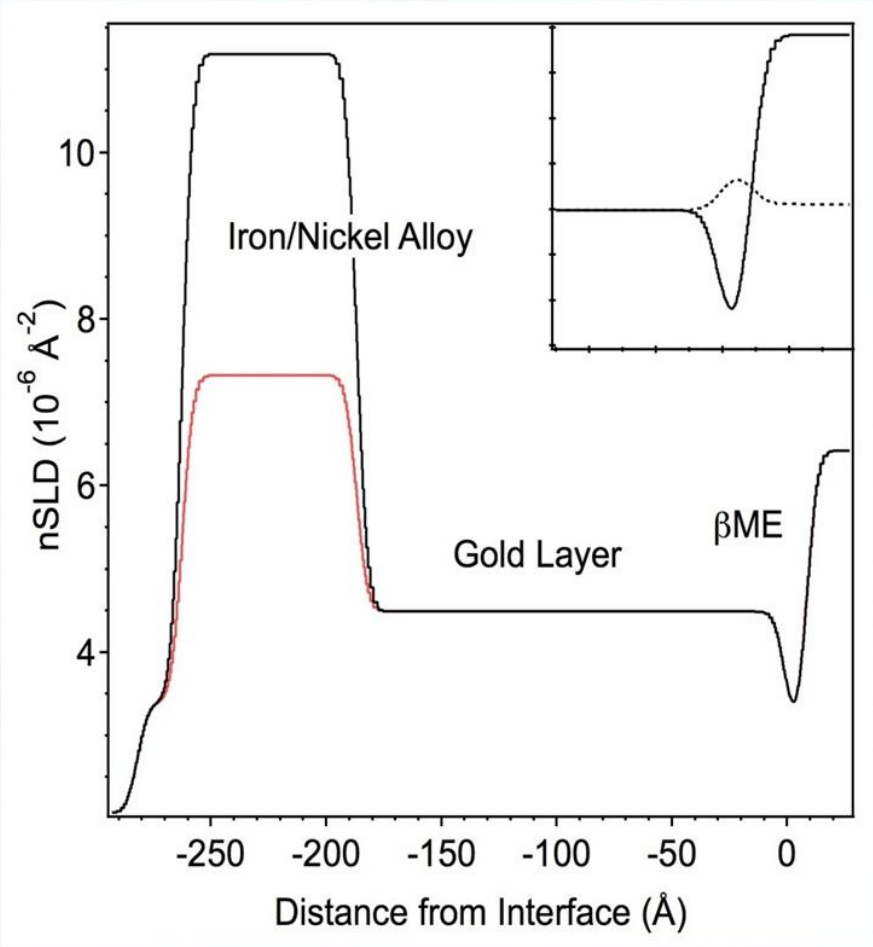
Thomas et al, 2001 (see ISIS annual report)

Magnetic Reference Layers



- Data from NIST
- The use of magnetic reference layers to study biomolecular materials





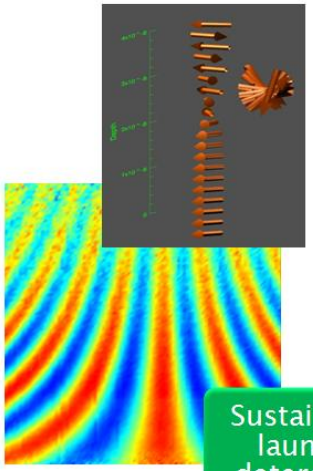
- Layer 8.2 ± 0.6 Å
- V_f 0.5 - 0.8

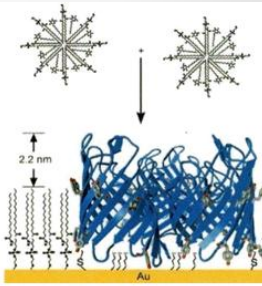
S.A. Holt et. al.

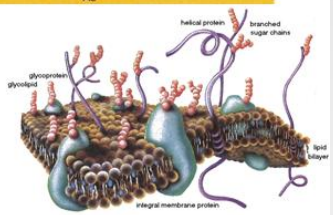
ansto
Nuclear-based science benefiting all Australians

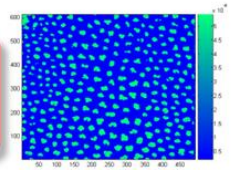
Science Themes

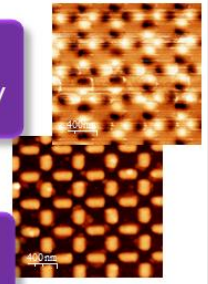
Fundamentally driven, *technologically relevant*











Protein resistant surfaces

Organic light emitting diodes

Lung surfactant

Surfactant adsorption biosensors

Atmospheric chemistry

Ionic liquids

Sustainable laundry detergents

Nanoscale hydrogen loading

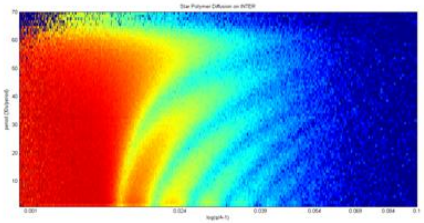
Gene delivery

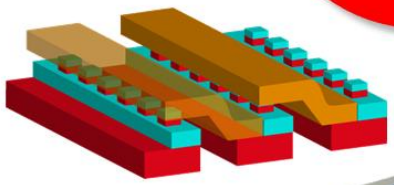
Nanoscale superconductivity

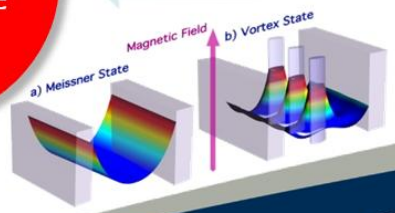
Inorganic templating

Neutron nanoscience

Spin transfer torque







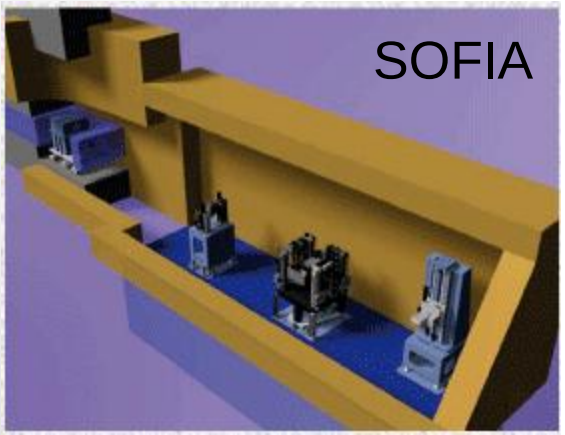
Science & Technology Facilities Council
ISIS

Current Instrumentation

- New sources in the past 5-10 years (FRM II, SNS, J-PARC, Ansto and ISIS-TS2)
- All have (or are planning to have) more than 1 reflectometer.
- Reactors
  - Platypus at ANSTO in Sydney, Australia
  - C5 and D3 at Chalk River, Canada.
  - D17, SuperADAM and Figaro at the Institut Laue-Langevin (ILL), France
  - EROS and PRISM at Laboratoire Léon Brillouin (LLB), France
  - N-REX+, MIRA, TREFF@NoSpec, REFSANS and MARIA at FRM II, Germany
  - V6 reflectometer at HMI, Germany
  - PNR at the Dhruva reactor Mumbai, India
  - REFLEX and REMUR at Dubna, Russia
  - NG1, NG7 and AND/R at NIST, United States
  - Neutron Reflectometer at the University of Missouri Research Reactor
  - ROG at Delft, Netherlands

Current Instrumentation

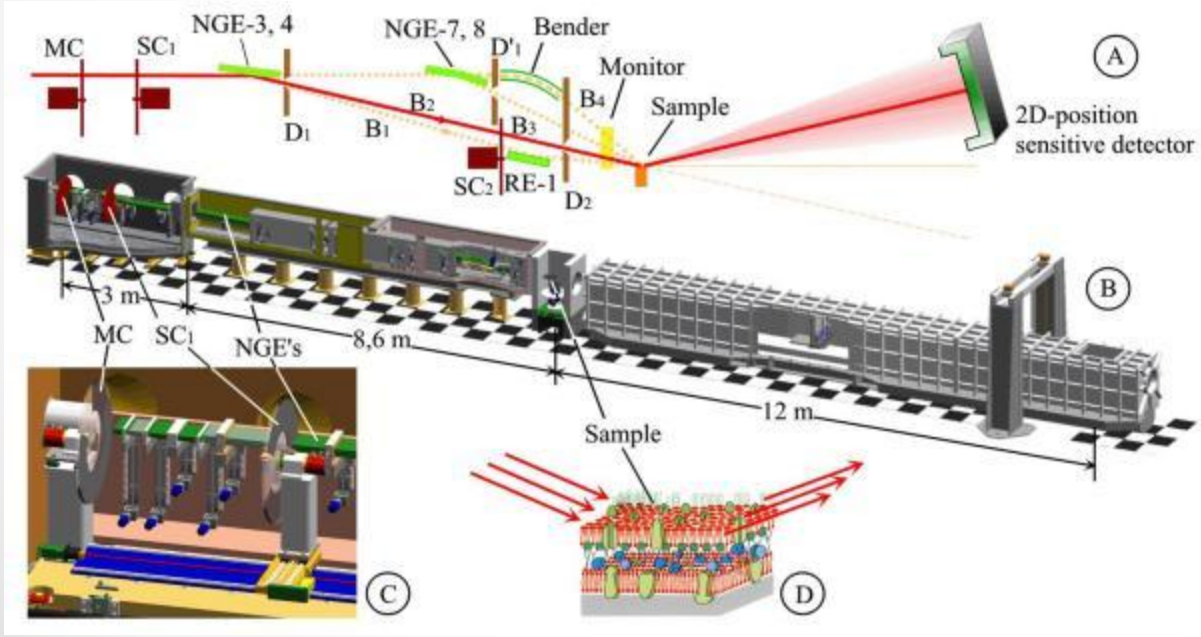
- Spallation Sources
  - ASTERIX and SPEAR at LANSCE, United States
  - AMOR at PSI, Switzerland
  - SURF, CRISP, INTER, Offspec and polREF at ISIS, United Kingdom
  - Liquids and Magnetic Reflectometers at the SNS, United States
  - BL16 (SOFIA) and BL17 (SHARUKU) at JPARC, Japan

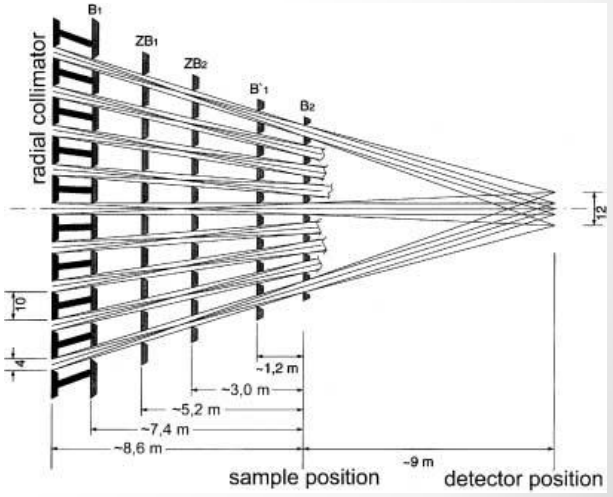


CRISP

Current Instrumentation

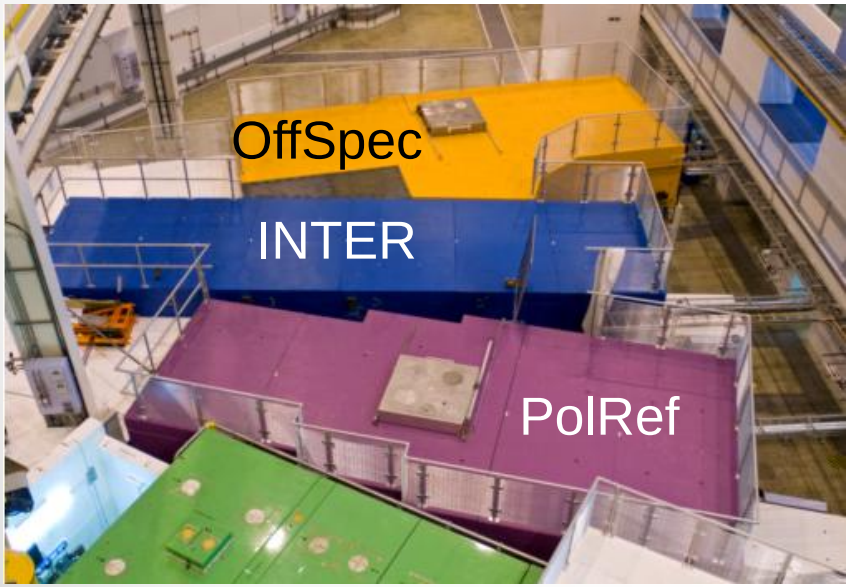
- Reflectometers are typically consider to be relatively inexpensive because they often have simple collimation and a small detector array.
- REFSANS at FRM 2 is possibly the most complex as it combines NR, SANS and GISANS.

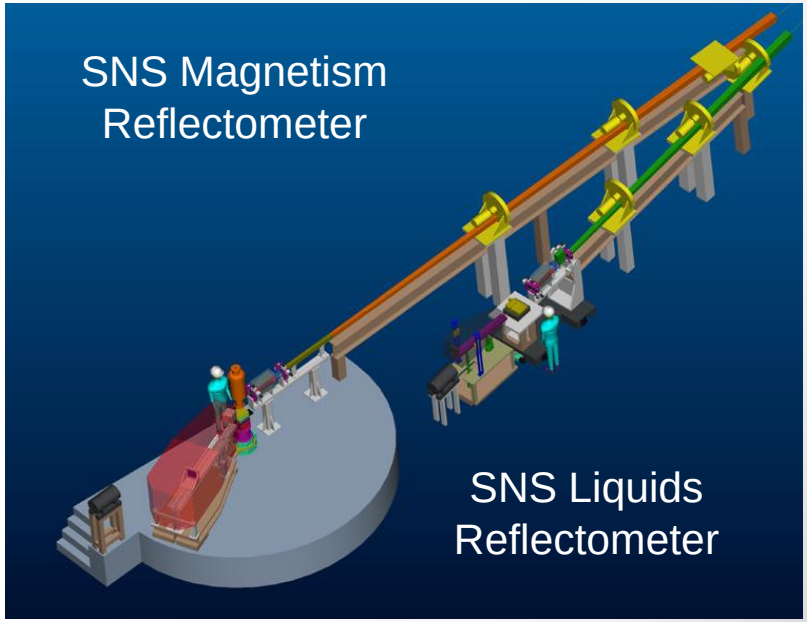




Primary Instrument Design Criteria

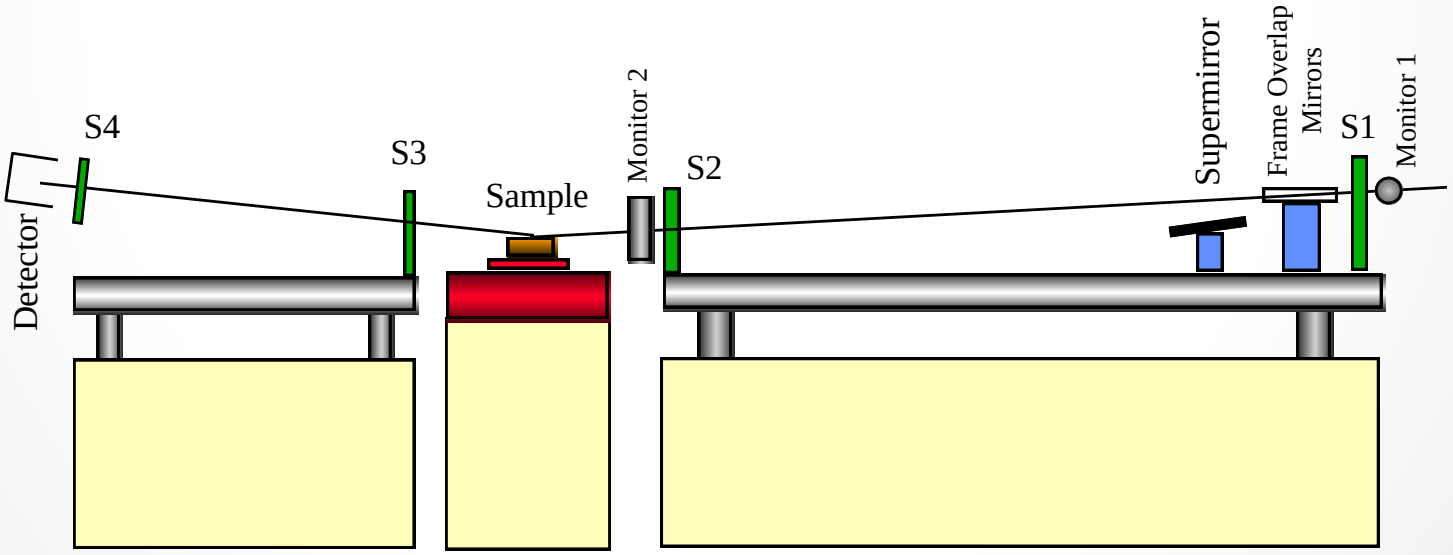
- Low Background.
  - This is required whatever the design and is the most important consideration
- What is the science case?
  - Is this is a general purpose instrument?
  - Are free liquid surfaces to be studied?
  - Magnetic systems?
- Design must be optimised to deliver science!





Simple Instruments (At first Glance)

- 4-5 movable apertures
- Wavelength defining devices (Choppers/monochromators)
- A moveable single element detector
- An incident intensity monitor
- A sample positioner

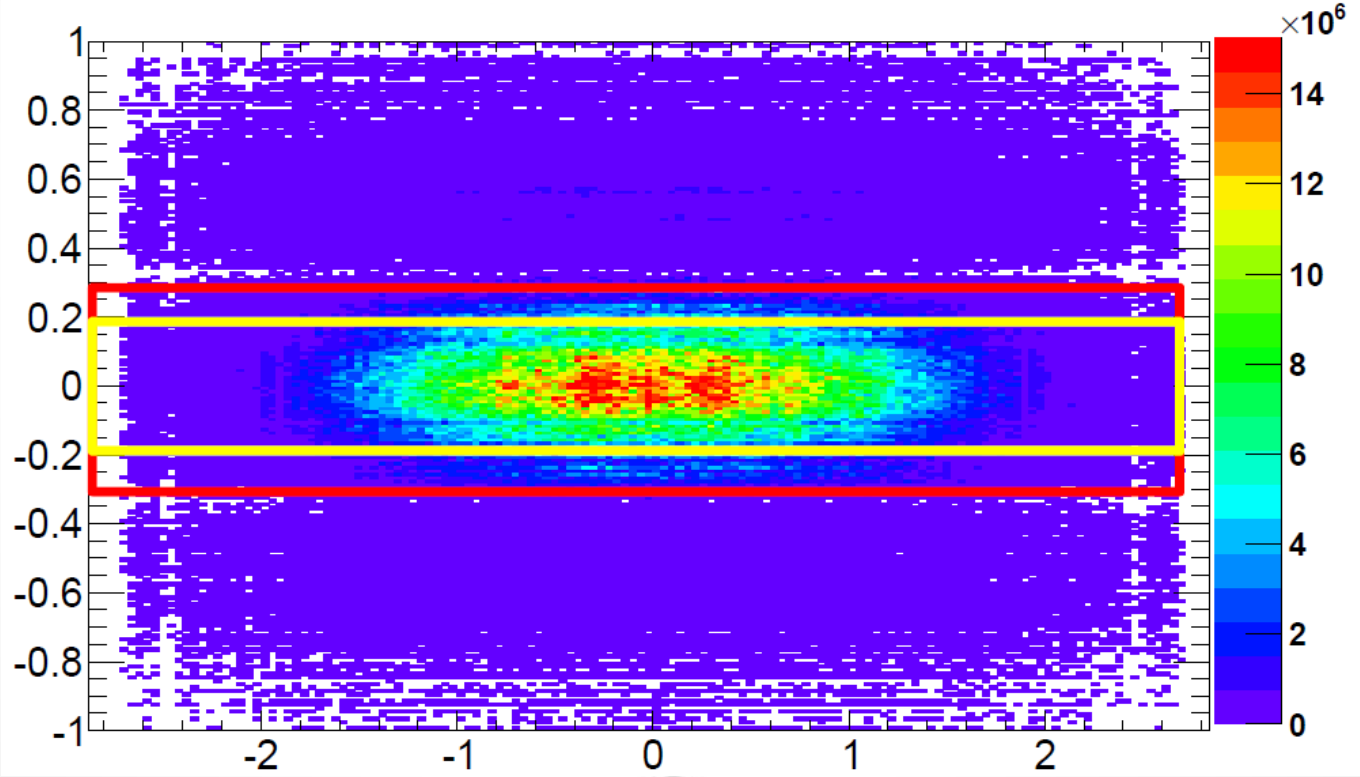


Resolution

- Because of the broad range of science and length scales studied resolution must often be varied between 1-5% dQ/Q.
- Physical optics at grazing incidence limit the angular resolution to >1% unless the instrument is very long or flux is sacrificed.
- Short Pulsed Spallation Source
  - Resolution dominated by physical optics, $\delta\lambda/\lambda<0.1\%$
- Reactor or Long Pulsed Spallation Source
  - Resolution function is more complicated θ or λ may be the dominant term.
- It is important to consider this and how to model it.

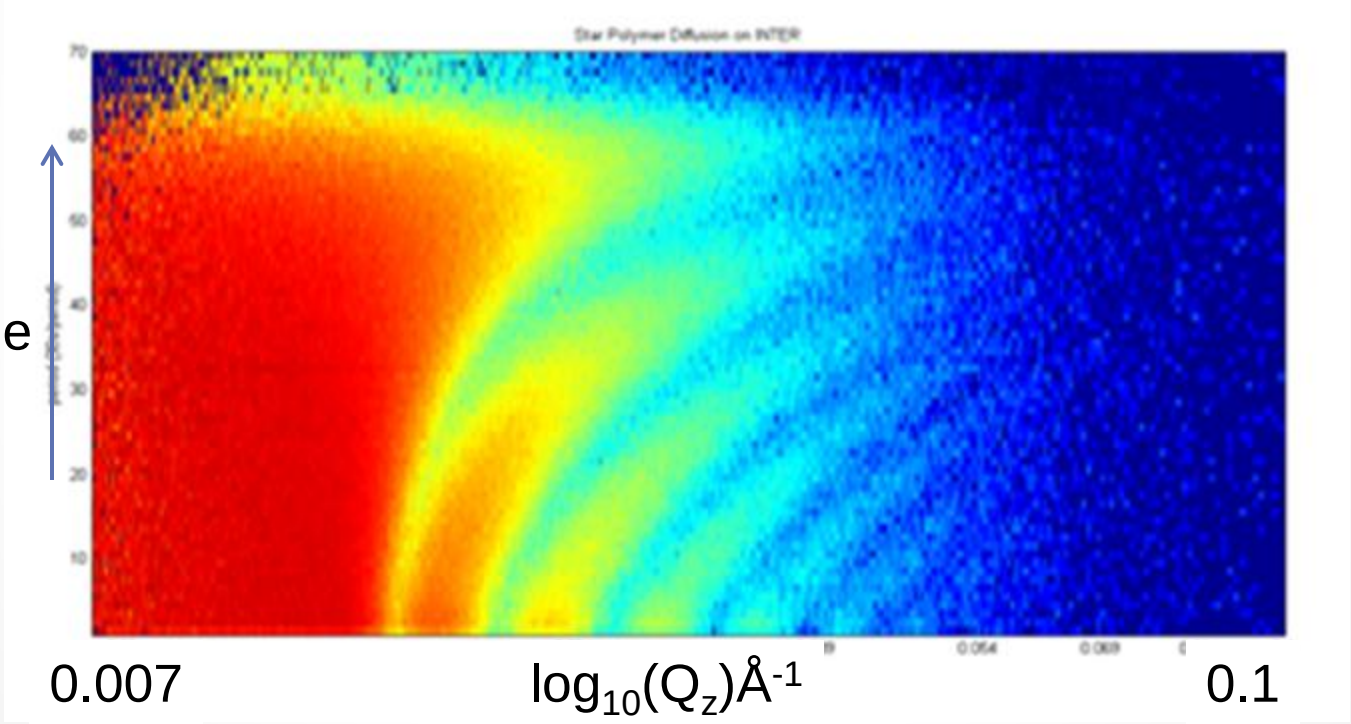
Beam Uniformity

- Beam flux profile should be as homogeneous as possible at all wavelengths.
- If it is not normalisation of data from different sample sizes becomes very difficult.

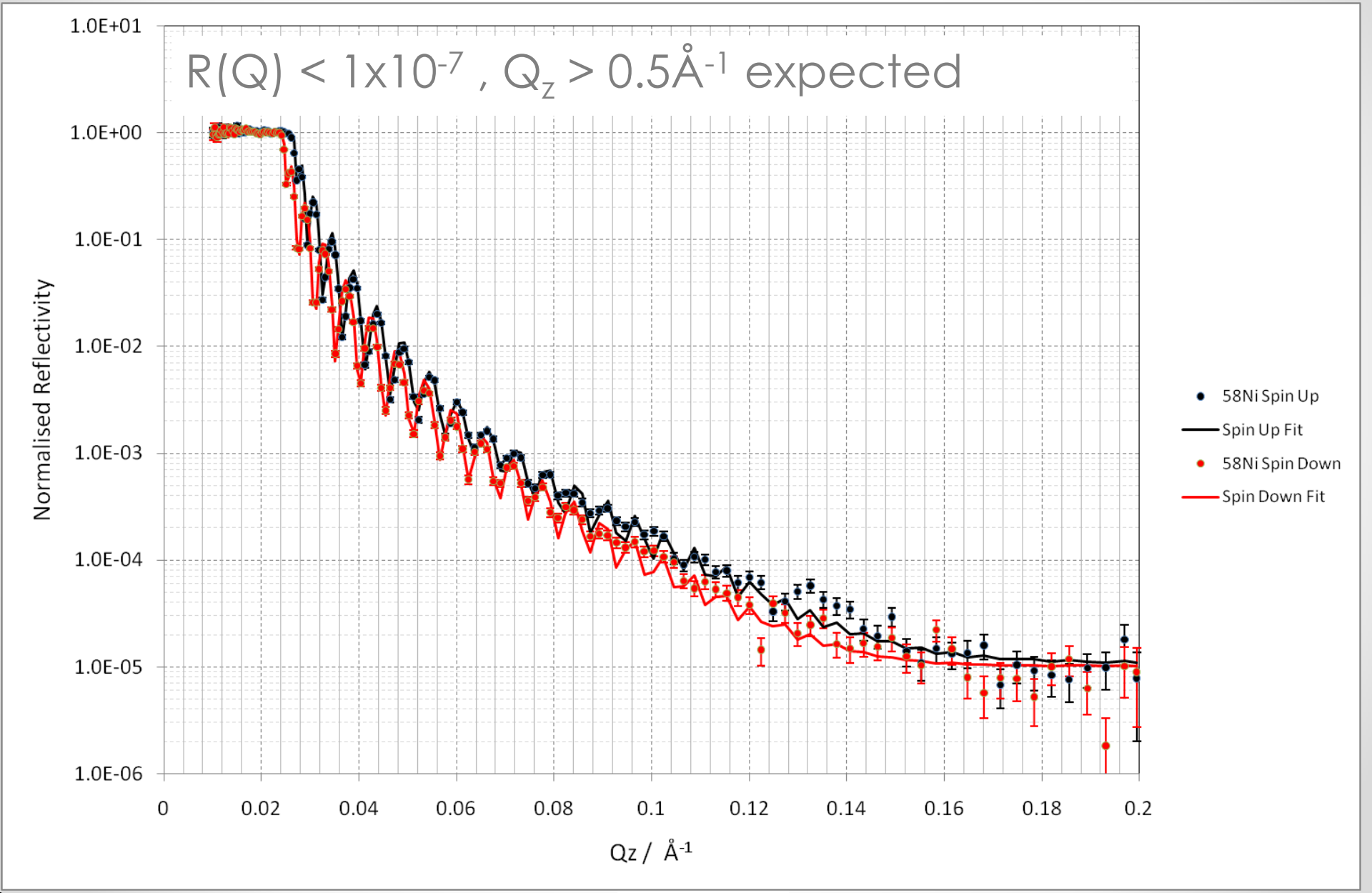


Bandwidth and Kinetics

- For kinetics measurement a wide simultaneous Q range is required.
  - INTER achieves this using a bandwidth of 0.8-20Å
  - D17 ~2-20Å.
  - Multiple simultaneous angles are difficult to use.
- $\lambda_{max}/\lambda_{min}$ is the important criteria. INTER and D17 >20.



Dynamic Range

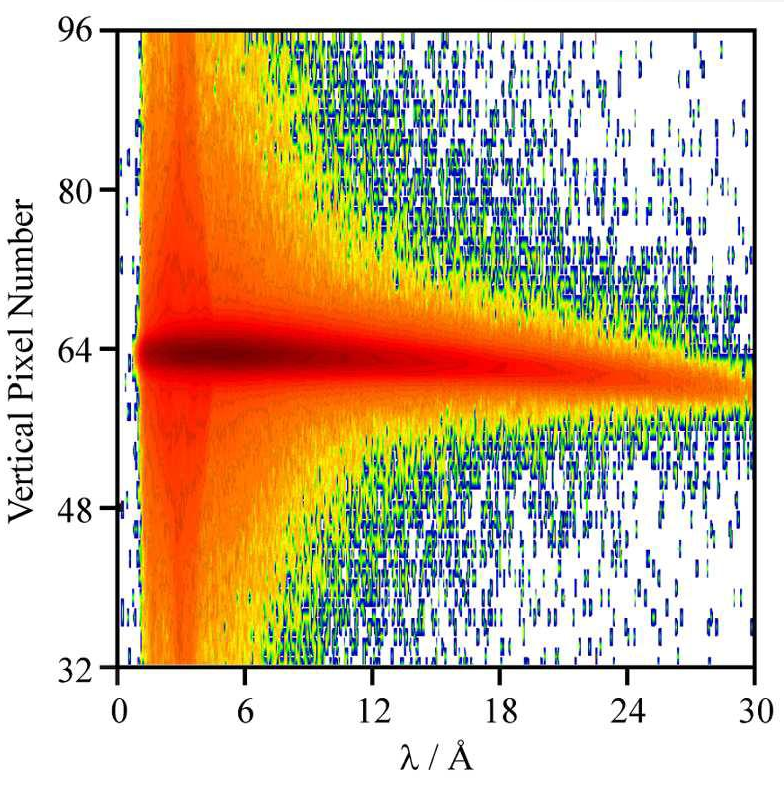


Gravity

- For a horizontal sample geometry gravity becomes a an issue.
  - <6-7Å effects are small
- Fine collimation (<0.5mm) required for high resolution measurement can introduce significant losses.
- Consider vertical geometry for small samples and focussing where possible.

Direct beam image from Figaro

Eur. Phys. J. Plus (2011) 126: 107



Major Components

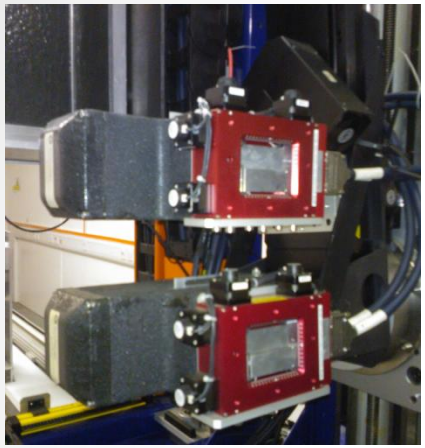
- Detectors
- Apertures/ Collimation Slits
- Choppers
- Polarisers and Analysers
- Sample Stage
- Focussing Elements

Detectors

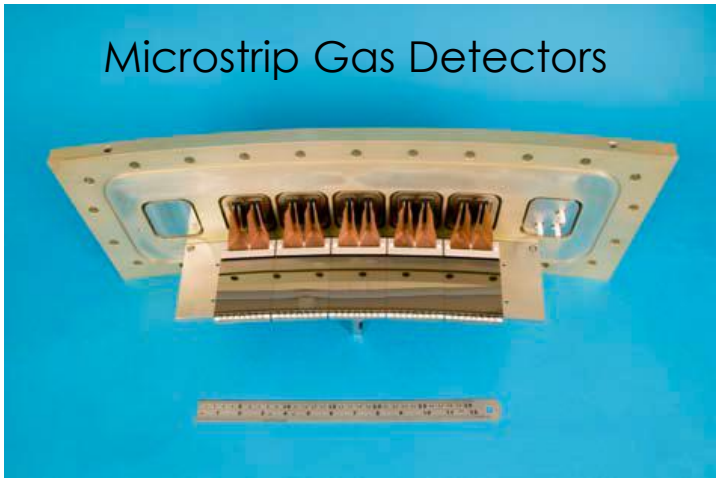
- Signal dynamic range is the most significant problem.
  - Detector must be able to count quickly in a small area (<2mm pixel size) but have low electronic background and good gamma discrimination.
- Saturation
  - SNS, JPARC, ILL and ISIS all have problems with detector saturation at the peak of the pulse if the direct beam hits the detector. (>>100kHz instantaneous rate)
  - Robust detector technologies must be chosen to allow for accidental exposure.
  - Filters may be required.
- Shielding
  - The detector must be very well shielded to only allow neutrons from the sample to reach the detection volume. This is difficult when large 2d arrays are required.

Some Possible Detector Types

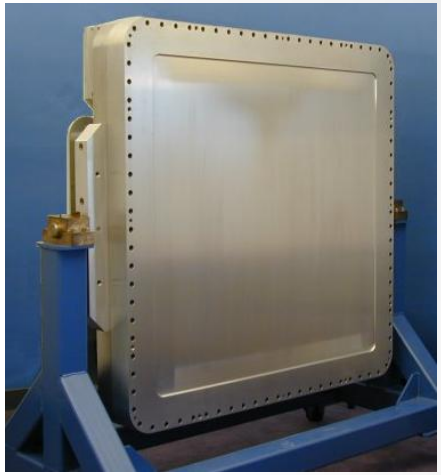
Single well shielded Helium tubes for specular reflectivity



Microstrip Gas Detectors



Multiwire proportional counters



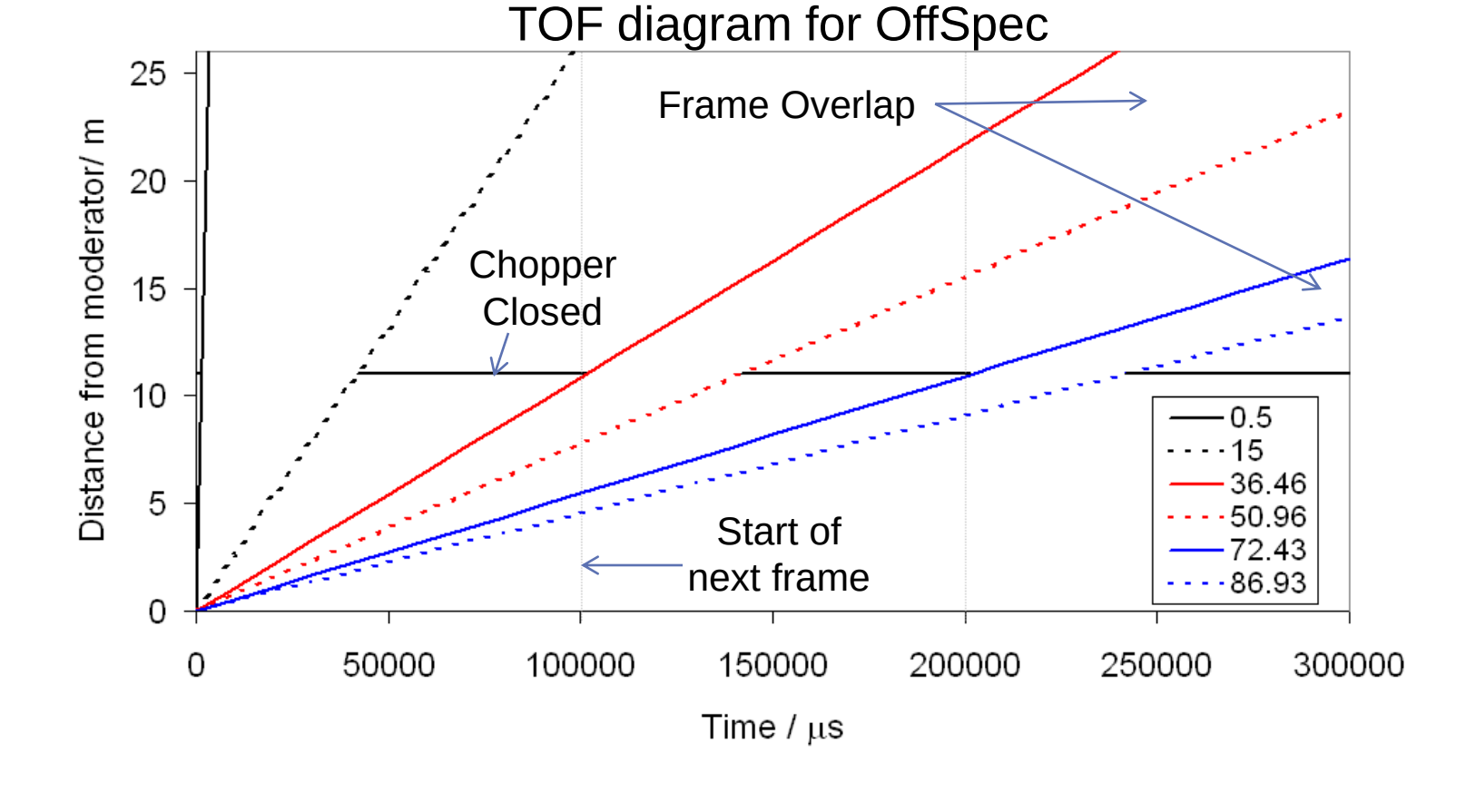
ILL resistive wire monoblock detector

Scintillator based detectors

- Direct scintillator to fibre coupled
- Wavelength shifting fibre coupled

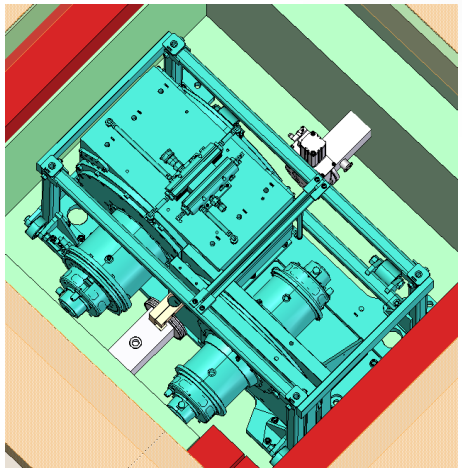
Choppers

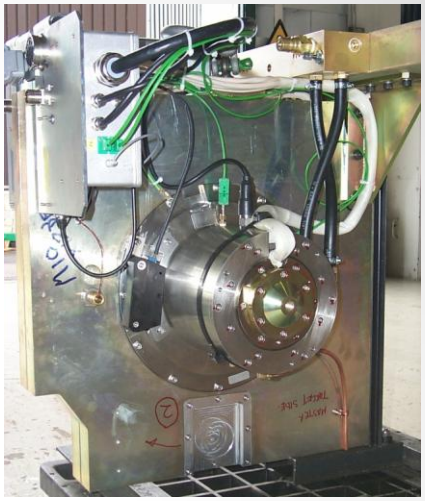
- Define the useful wavelength band or help to reduce background
- Slightly different problems on reactors and pulsed sources.

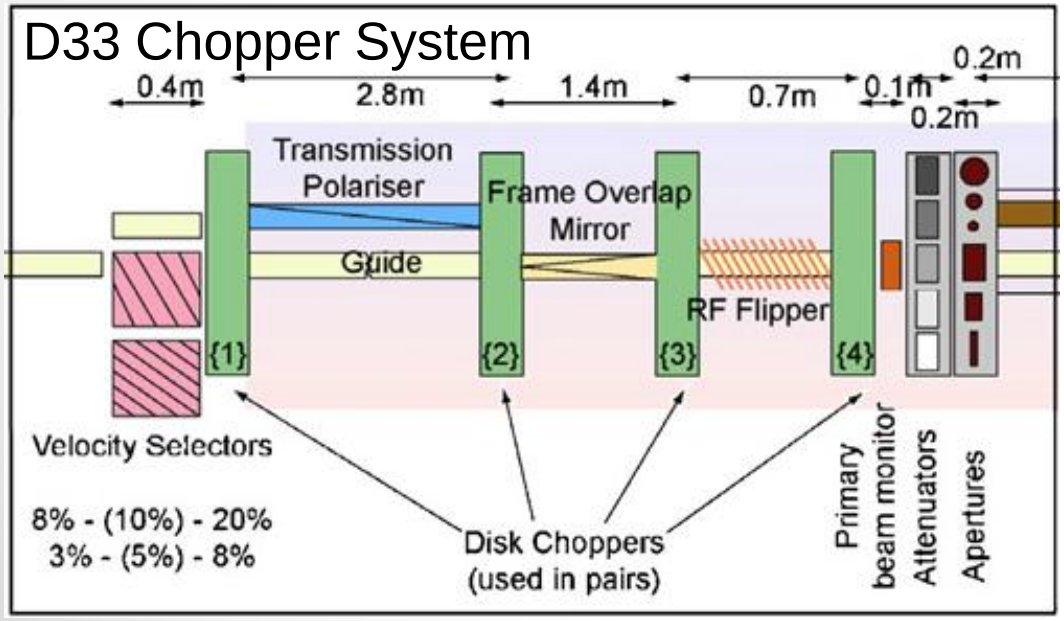


Choppers

- Pulsed Source
  - Large motors to keep the chopper in phase with the source.
  - Thick blades to absorb high energy neutrons (<0.5Å). (ISIS T0 Chopper is 700mm long and is made of nimonic alloy)



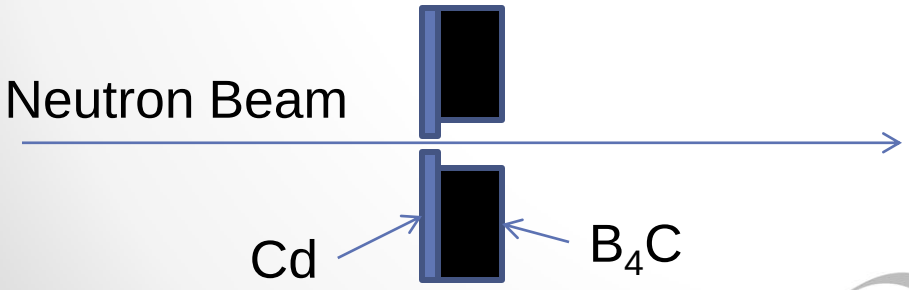


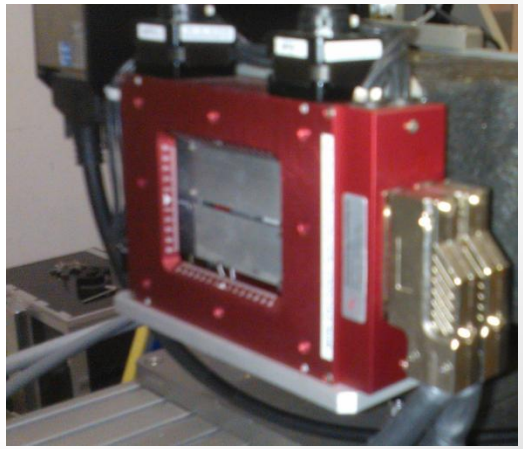


- Reactors
  - Smaller devices synced to each other to provide varying resolution
  - Thinner blades as instruments are typically on long guides using cold/slow neutrons that are easily absorbed.

Collimation

- Critical to performance
- Collimation length may be optimised to the science case e.g.
  - SURF – Short instrument (<5m) optimised for 5% resolution and high flux
  - CRISP – Longer flight path (~7m) 3% resolution optimum
- Absorbing material choice is crucial
  - Sintered B_4C – Difficult to machine and reflective, emits gammas
  - Cadmium – Soft, toxic, transparent at high energies, emits gammas
  - Lithium in a matrix – Soft, low absorption efficiency, no gamma emission
  - Gadolinium – expensive, difficult to machine.
- Cadmium backed with B_4C?





Polarisation

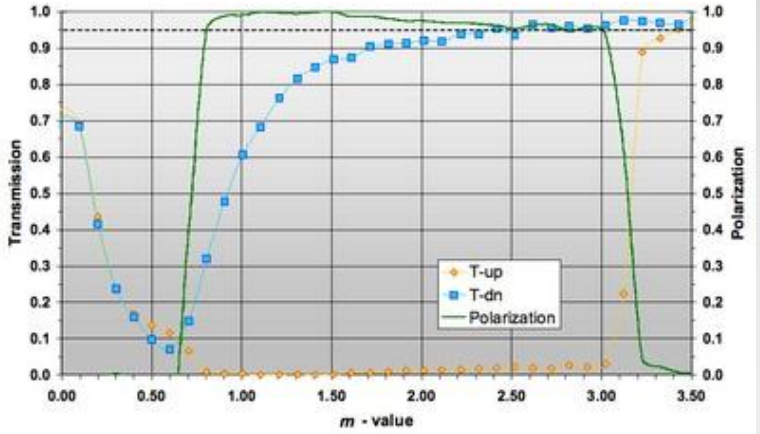
- Neutrons are spin ½ particles
- In an external field the neutrons align parallel (spin up) or anti-parallel (spin down) to the field.
- The polarisation is a scalar that expresses the relative population sizes of the two alignments

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

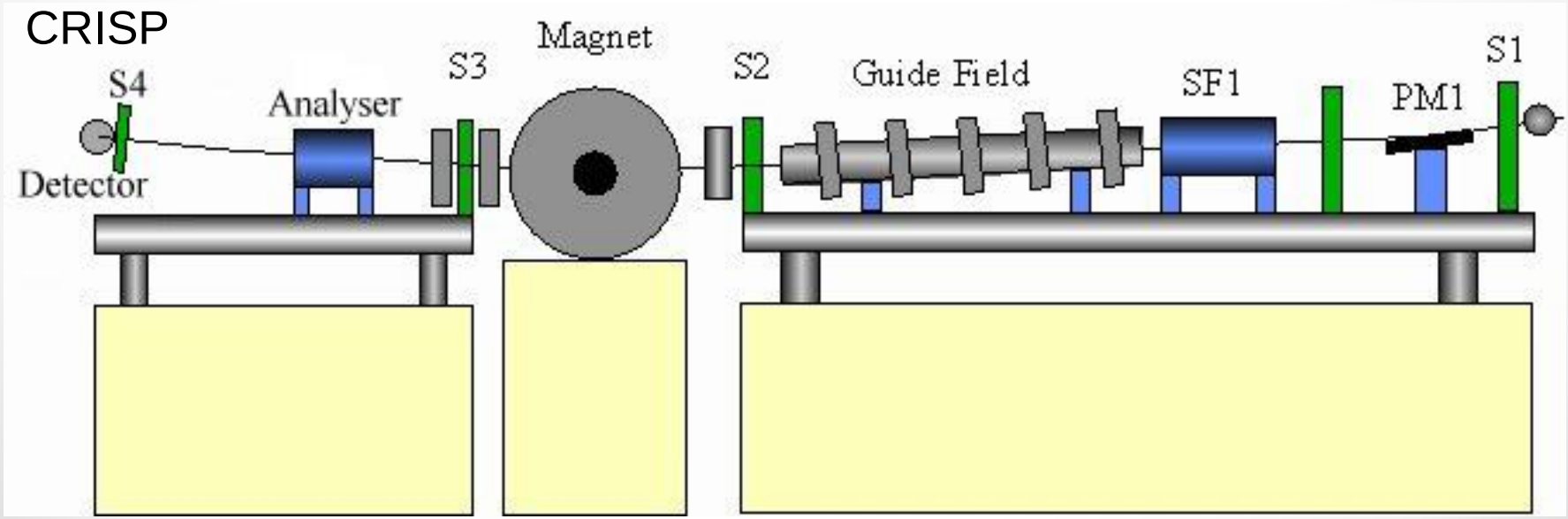
- Polarisation is never perfect and is usually wavelength dependent.

Polarisation

- Because of the use of ribbon collimation polarisation can be readily achieved using a polarising mirrors.

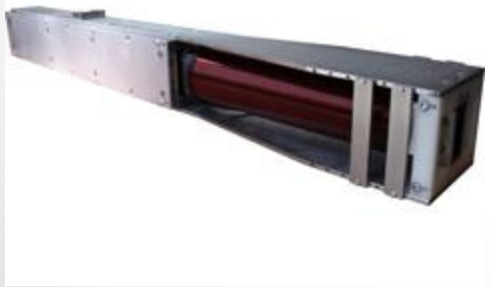


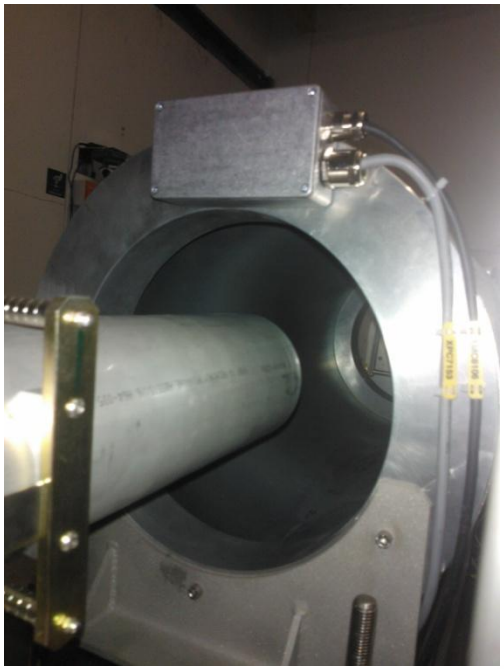
Beam Direction

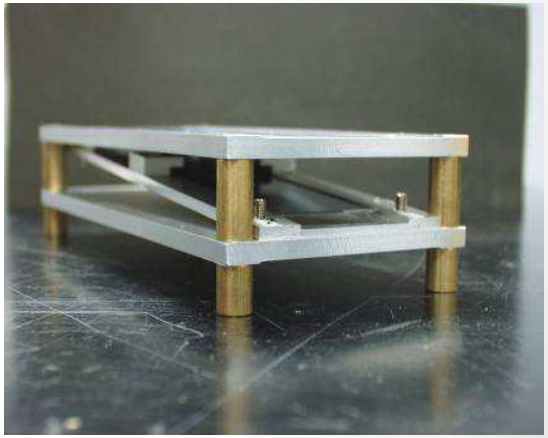


Spin Flippers

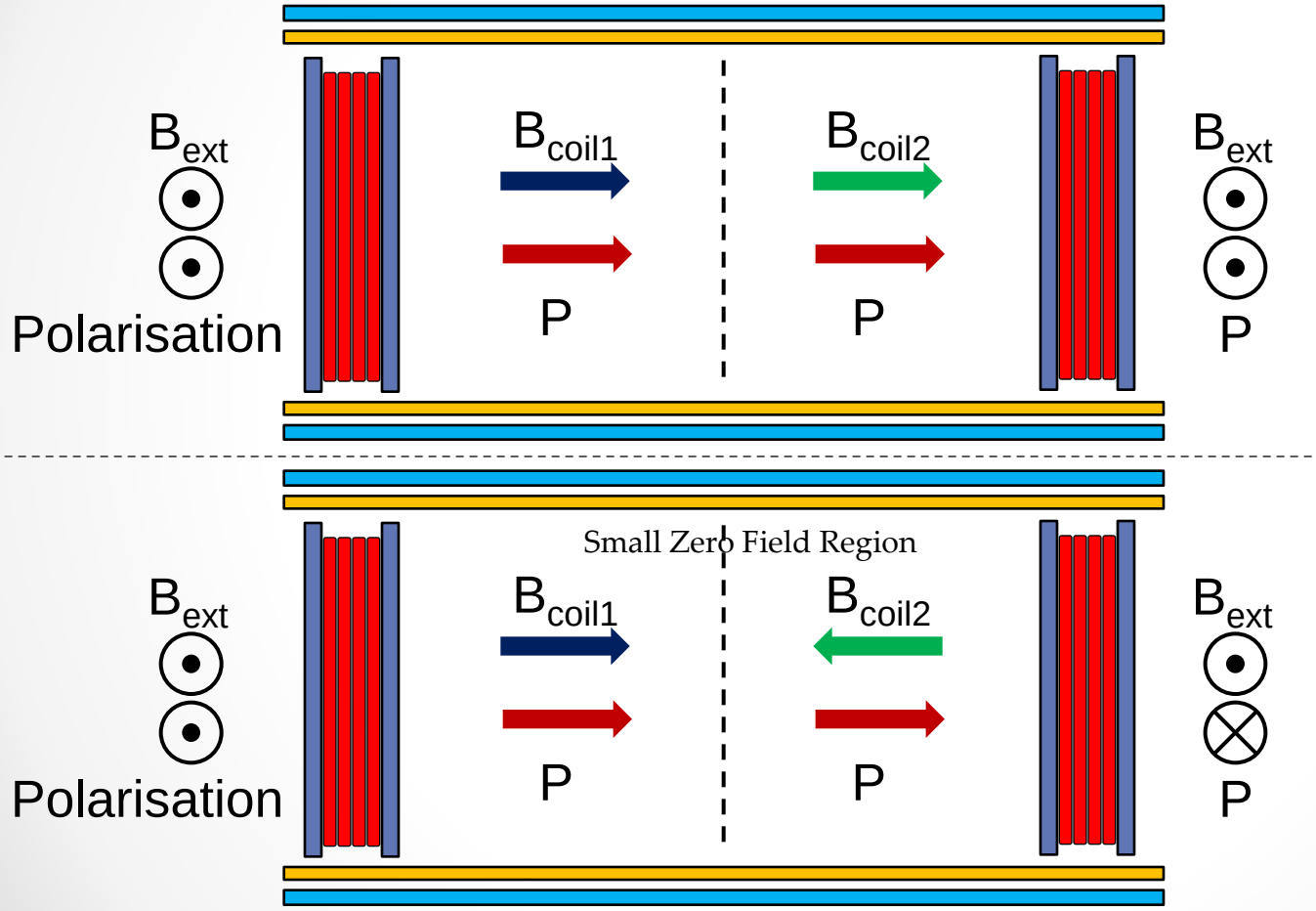
- Ideally no material in the beam for SANS and NR
  - Drabkin Flipper, RF Flippers
- Numerous types of design with material in the beam
  - Current Sheets, Mezei flippers, V-coils, Magnetised thin films...
- Or Flip the polariser/analyser
  - Remnant field mirrors, RF flip polarised ^{3}He







Drabkin Spin Flipper

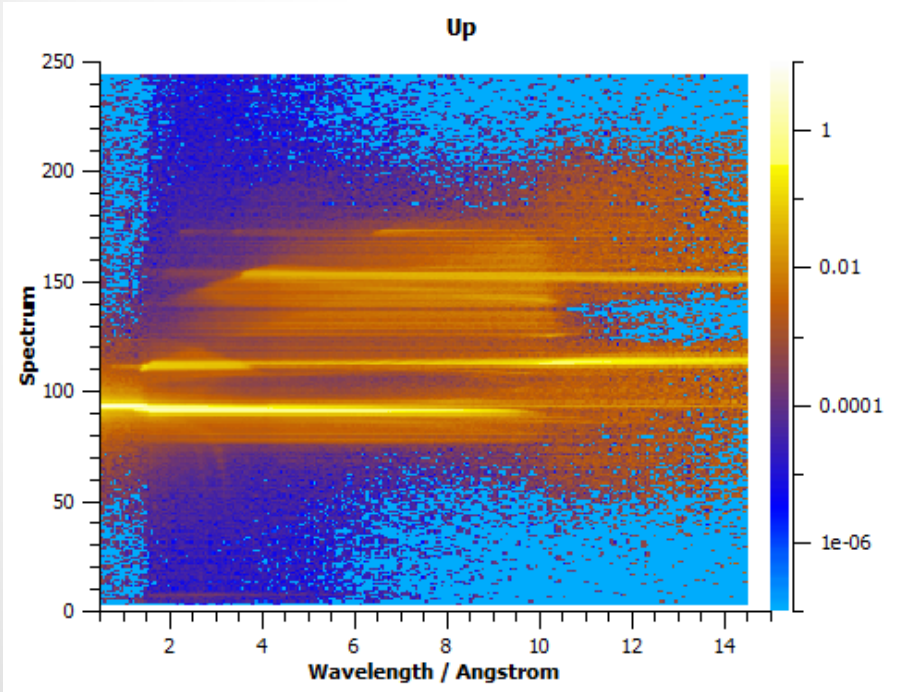


Polariser and Analyser

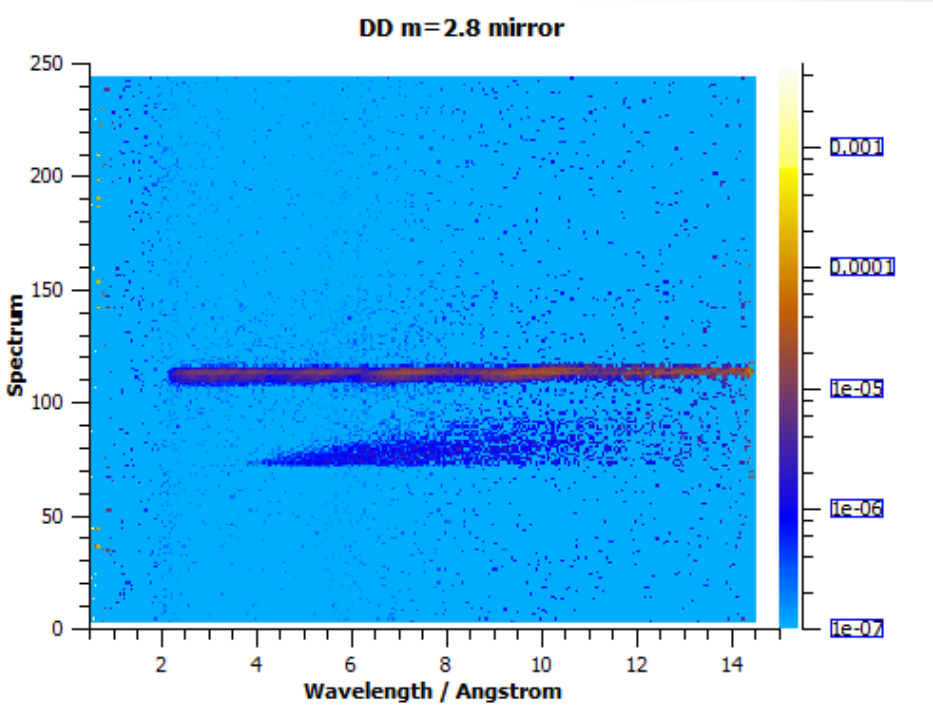
- Choice depends on application, wavelength band and budget.
- Polarised ^{3}He
  - Ideal for monochromatic beams or where only a short bandwidth is needed
  - Polarisation can be traded for intensity and bandwidth.
  - On beam pumping (SEOP) can provide stable polarisation
  - Essential if an undistorted 2D polarisation detector image is required.
  - Does require expertise and significant capital if not already available.
- Supermirrors
  - Provide stable polarisation over a broad bandwidth.
  - 99% polarisation can be achieved readily
  - Large area coverage is expensive
  - Can introduce parasitic scatter.

Polariser Induced Structure

Domain Scatter from a transmission polariser



Reflection Polariser with absorbing underlayer

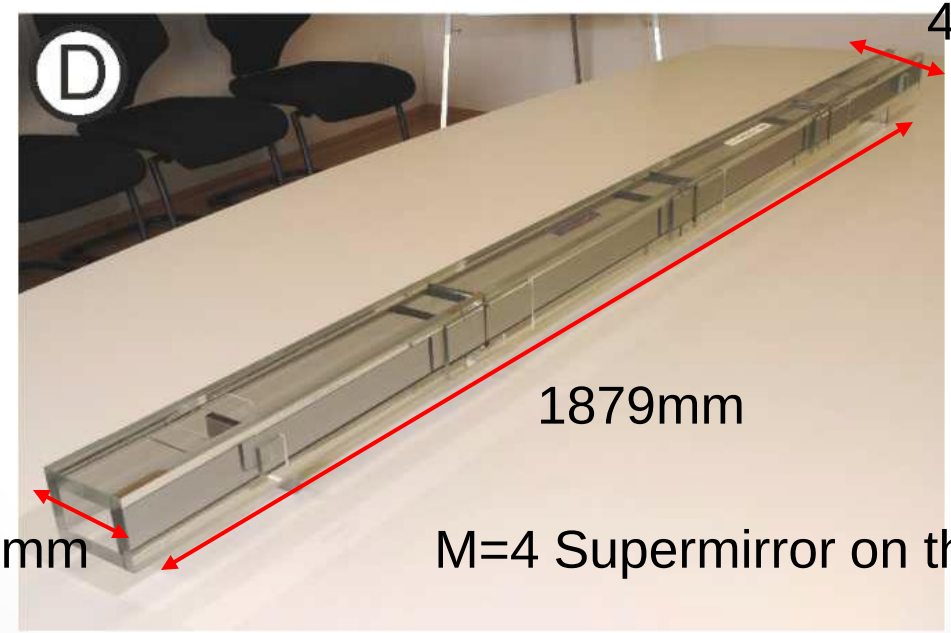


Sample Stage

- Very accurate positioning required.
  - <0.001° repeatability on arcs
  - <0.01mm resolution on height
  - Accurate positioning on centre of rotation essential.
- Readily achieved for light loads
- More difficult with magnets and cryostats
- Multi axis designs or hexapods are now much more available.
  - Good equipment is useless without the software to drive it.

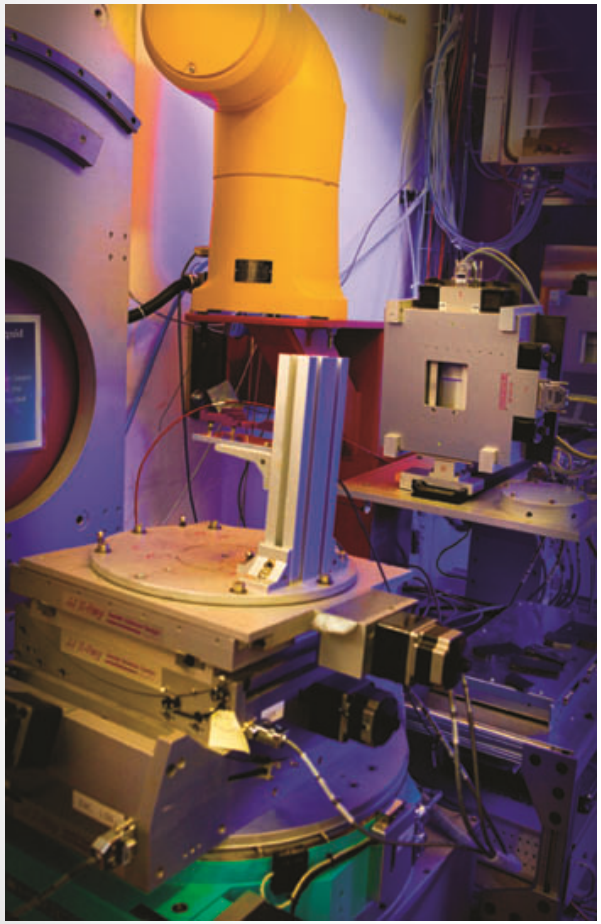
Focussing

- Focussing is readily possible in the non-scattering plane.
  - FIGARO (ILL) incorporated this into its optics
  - Focus from 59.4 to 40mm
  - Beam is broader at the detector so a larger integration region is required.



Sources of Background

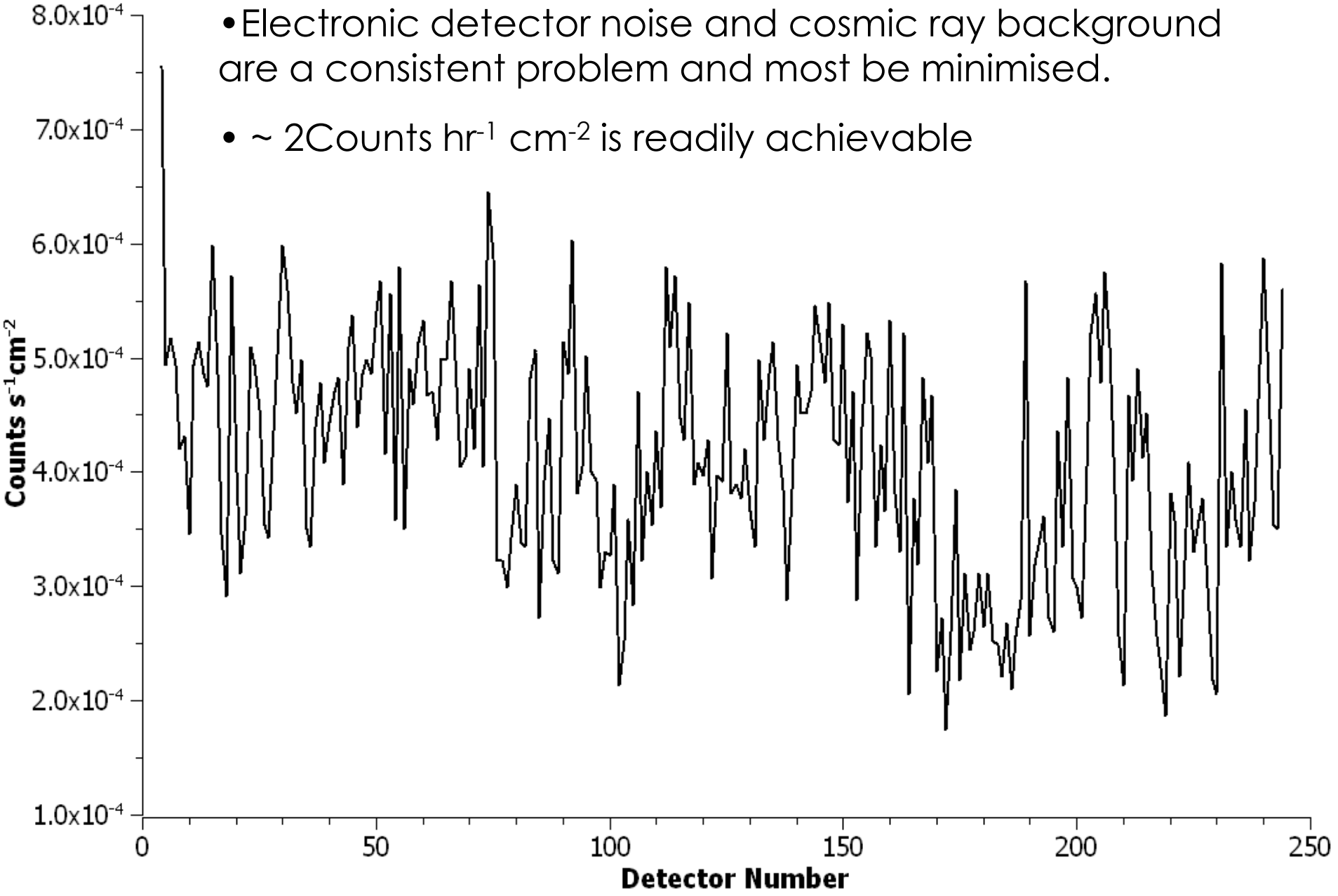
SNS Liquids Reflectometer



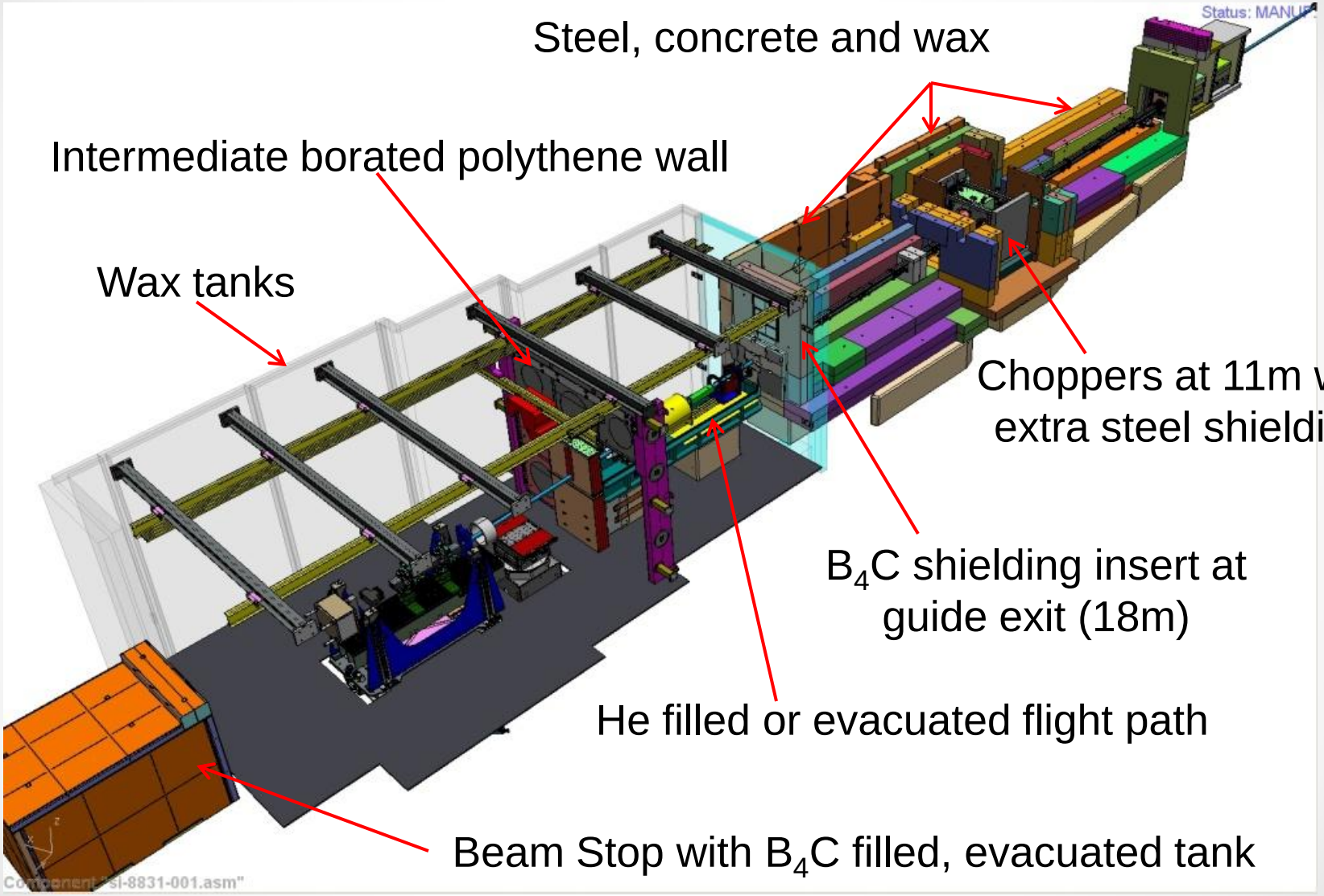
- The Sample
  - Incoherent scattering from the sample is the most significant limitation to measuring reflectivities below $1x10^{-7}$.
  - Need to remove hydrogen and reduce sample volumes.
- Air Scattering
  - Evacuate flight paths (REFSANS, D17)
- Stop high energy neutrons as early as possible.
  - ISIS TS1 and the SNS instruments suffer from backgrounds originating from the moderation of fast neutrons in the shield at the front of their blockhouses.
- Stray Reflections
  - It is difficult to simulate all possible stray neutron paths and reflections from optics.
- Think the worst!

OffSpec Detector Quiet Counts

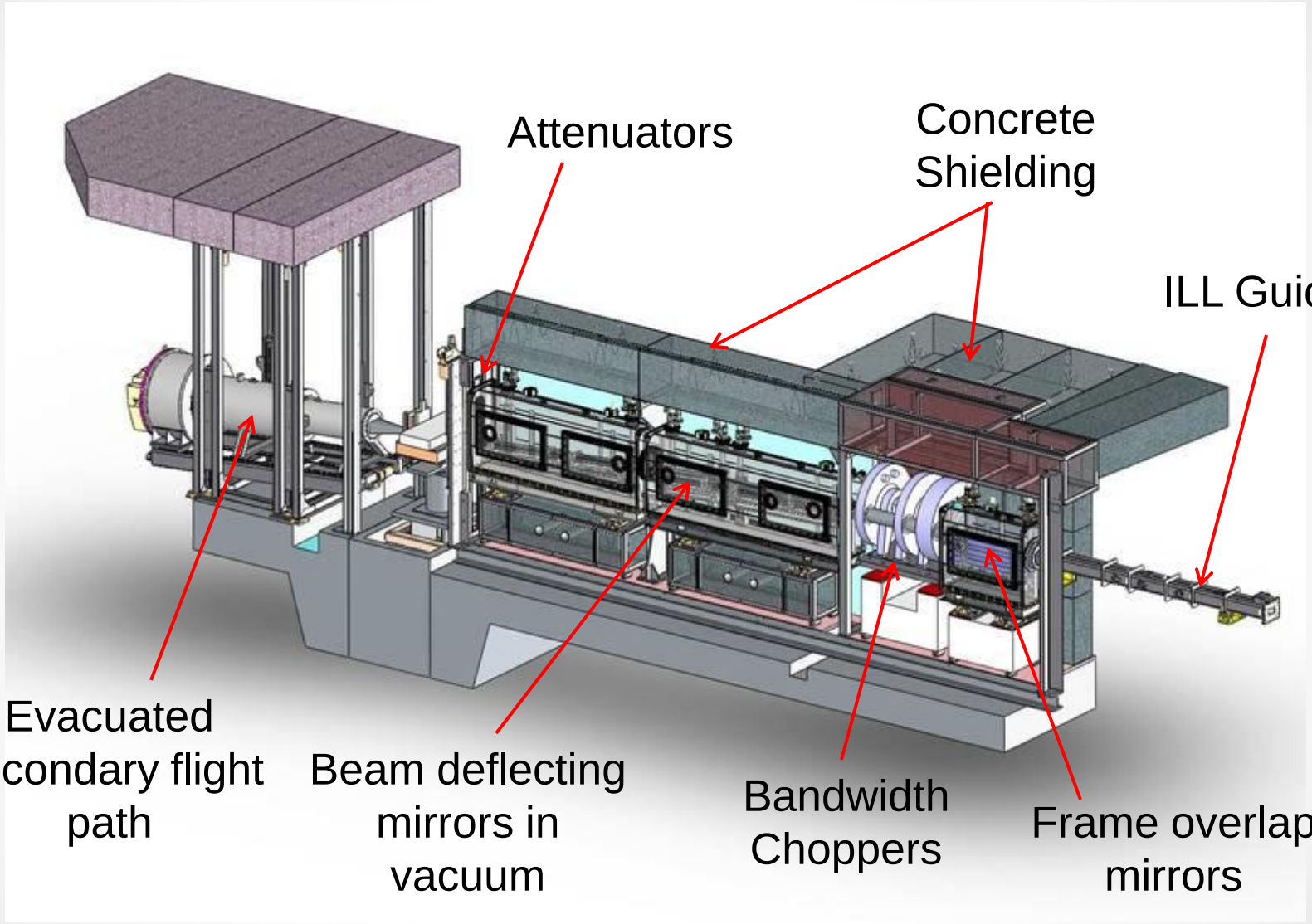
- Electronic detector noise and cosmic ray background are a consistent problem and most be minimised.
- ~ 2Counts hr^{-1} cm^{-2} is readily achievable



PolRef ISIS TS2



FIGARO, ILL

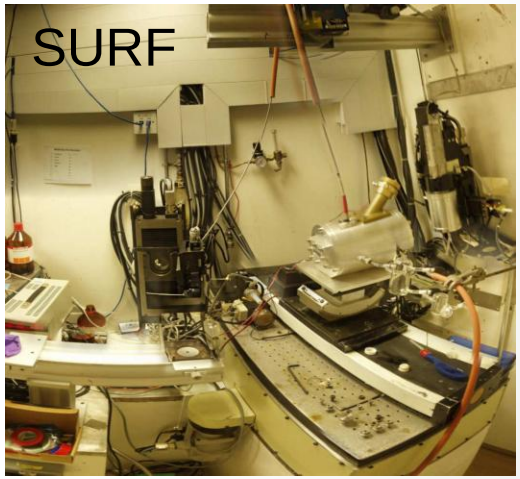

Attenuators
Concrete Shielding
ILL Guide
Evacuated secondary flight path
Beam deflecting mirrors in vacuum
Bandwidth Choppers
Frame overlap mirrors

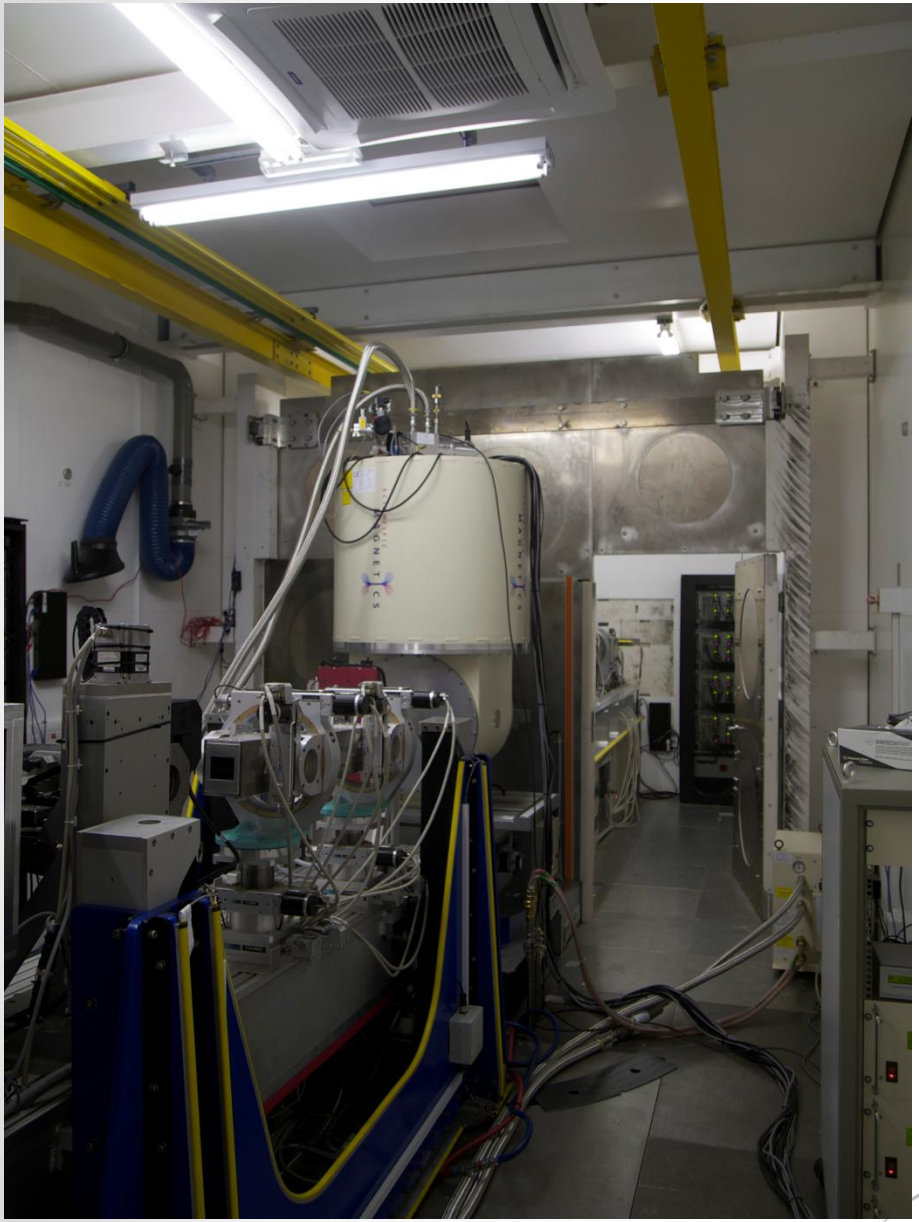
Sample Background

- Full polarisation analysis is often considered as a way to remove sample incoherent scattering.
- Be very cautious when considering this.
  - Factor of at least 3-4 loss in flux because of the polarisation process
  - Background signals are low intensity ($<10^{-4}$) so the mathematics is working against you because there are very few counts.
- Additional scattering from the polarising components may cause more problems.
- Phonon scattering in e.g. Silicon also produces a background at the level of $R(Q)=10^{-7}$. (Cubitt et al.) so it will always be difficult to reach a reflectivity of $<10^{-7}$.
- Consider different sample geometries that reduce the amount of e.g. Hydrogen.

Ancillary Equipment

- Hugely important
  - Need to consider what sample environment equipment will be required and allow space for it.
- Cryostats and Magnets
  - Space for pumps and control systems
  - Non-magnetic materials to be used to prevent strain on the magnet which can lead to quenching.





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