# 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

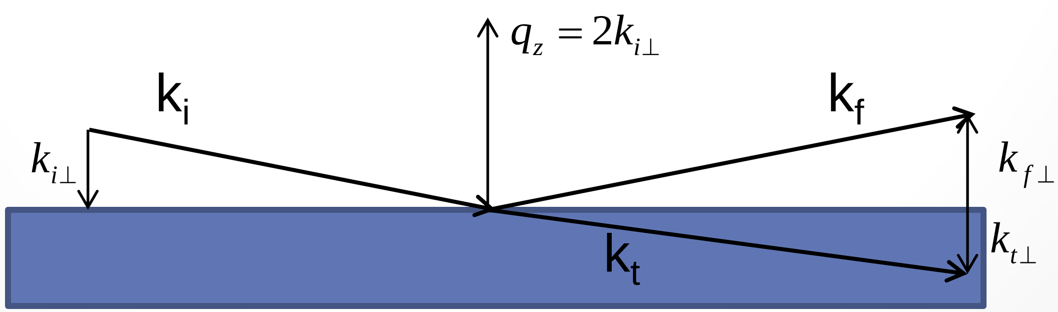
- 1946 - First observation of total external reflection of neutrons by Fermi and Zimm.
- 1947 - Fermi and Marshall performed detailed studies using monochromatic neutrons to measure scattering lengths of materials.
- Neutron Reflection then continued to be used to study bulk scattering lengths.
- 1976 - 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 $\theta$-$2\theta$ 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).

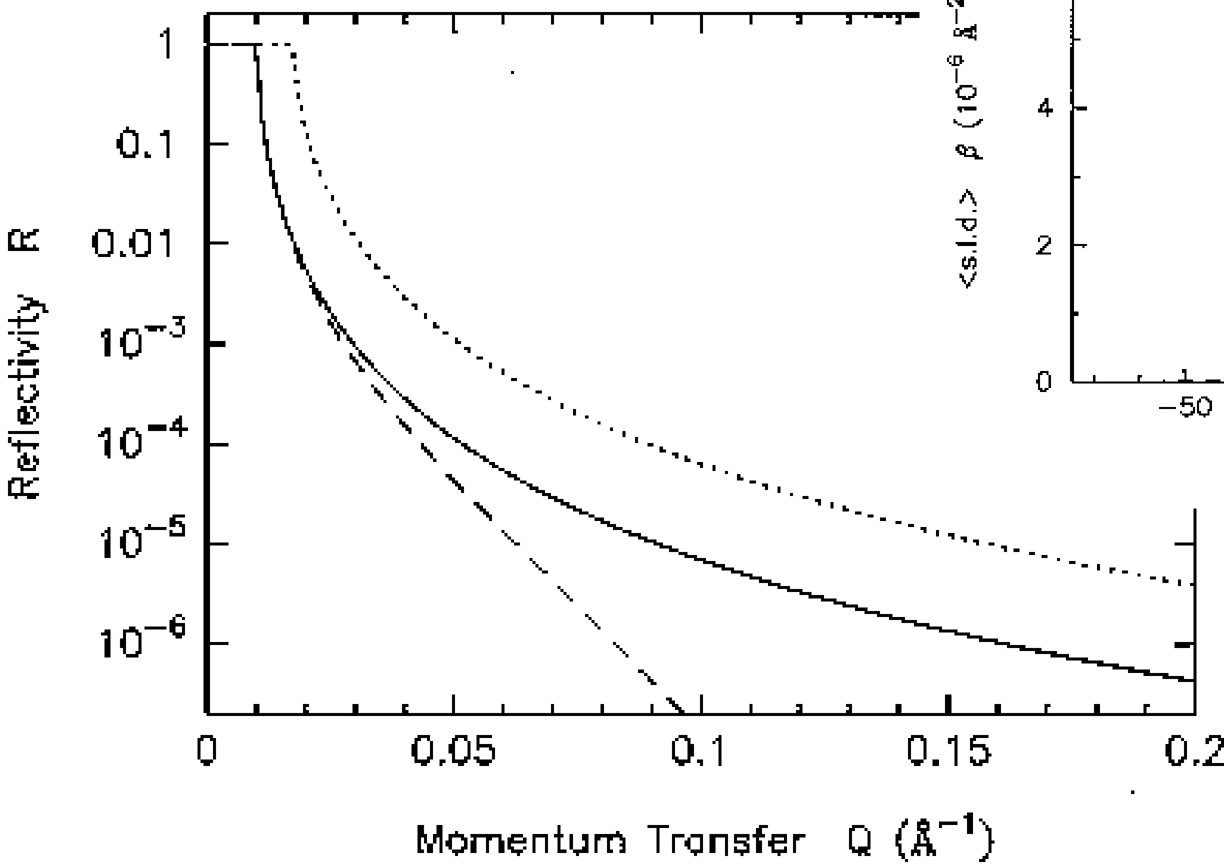
$$n \approx 1 - \frac{\lambda^2}{2\pi}\sum_i b_i N_i \qquad SLD = \sum_i b_i N_i$$

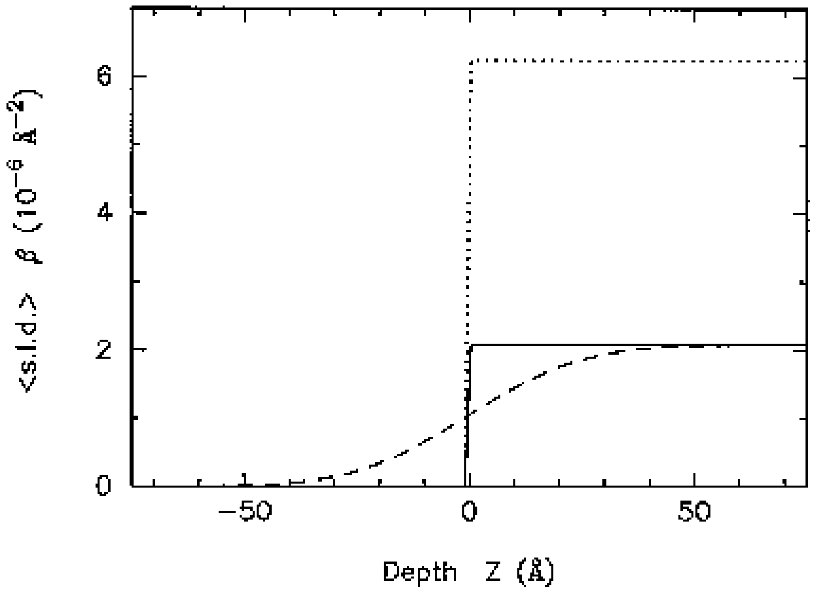
  - b is the scattering length and N the number density of a given atomic component and λ is the incident neutron wavelength.
  - The reflectivity, measured as a function of wavevector transfer perpendicular to the interface ($q_z$) be calculated using multilayer slab models of the SLD. Depending on the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



# Specular Reflectivity

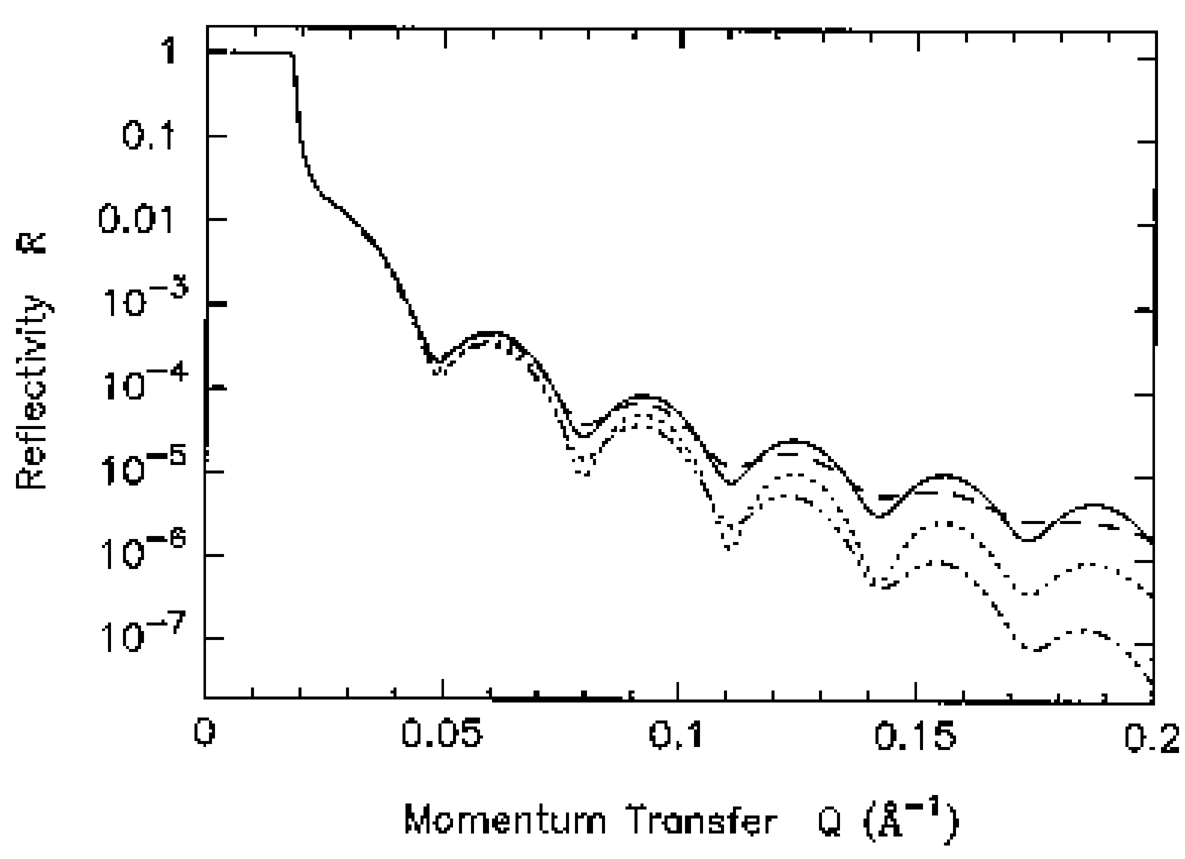
- Surface roughness and scattering length density changes are readily observed.

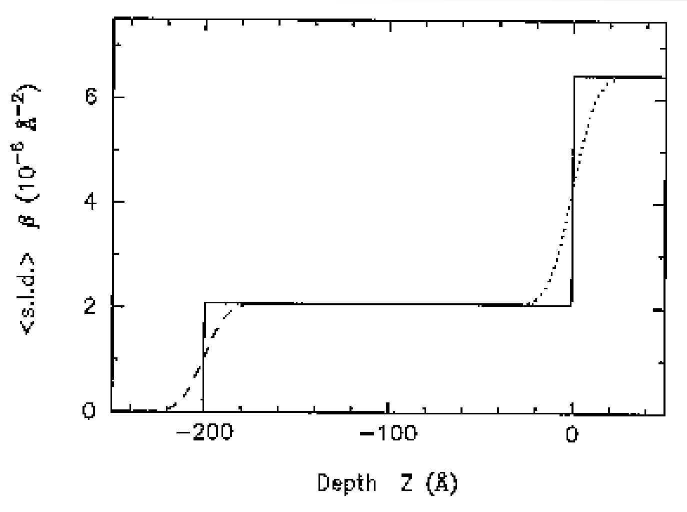


# Speular Reflectivity

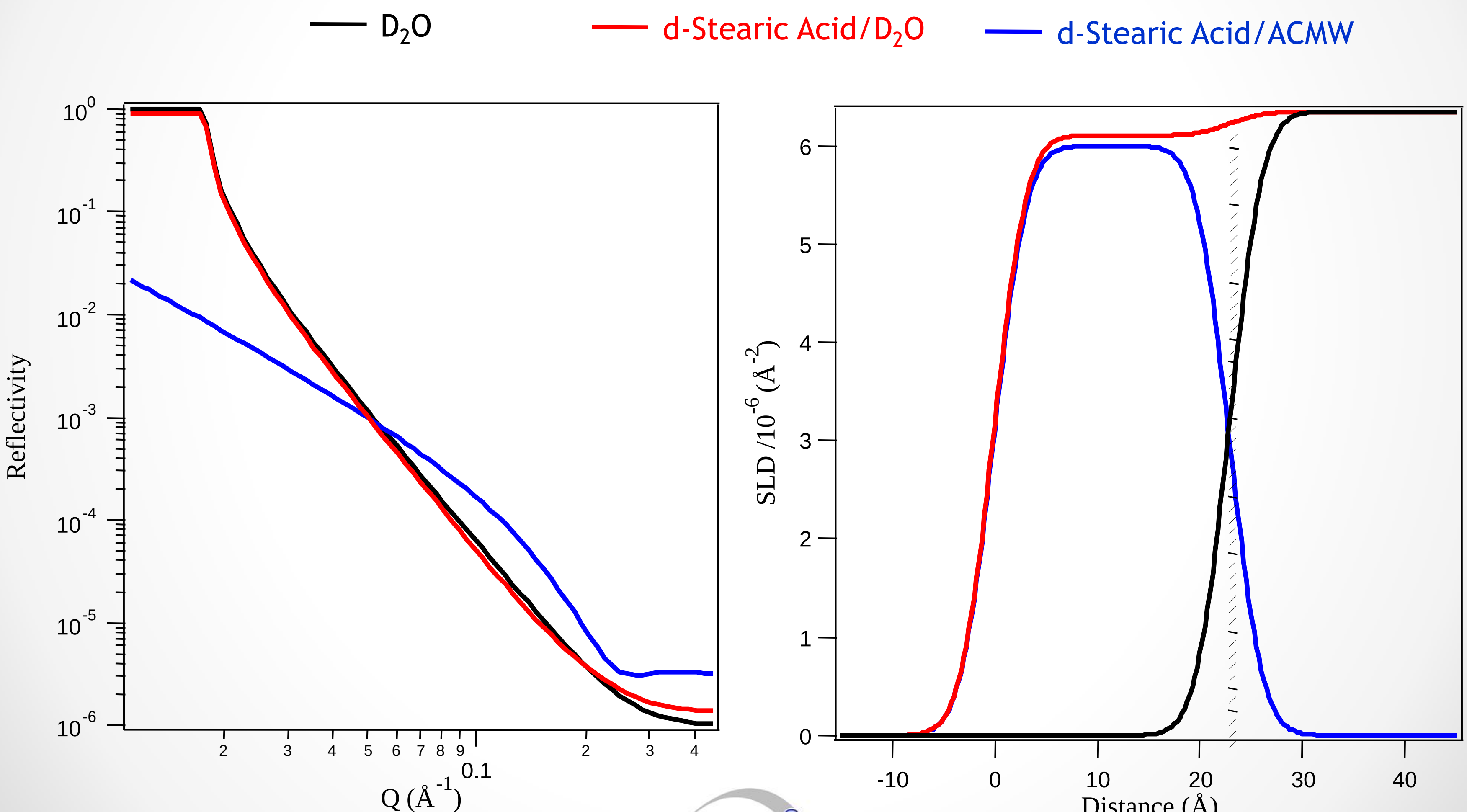
- Thin films at the interface lead to interference fringes.
- Interfacial roughness makes lie more complicated



# Specular Reflectivity

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

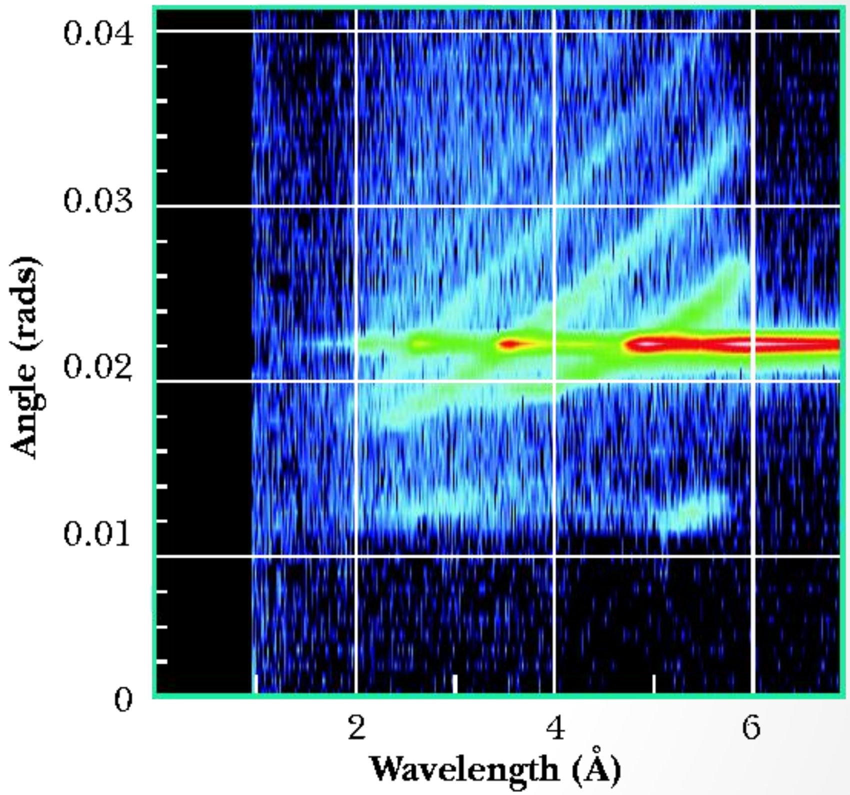


# Off Specular Reflectivity

- Any Scattering that does not satisfy:

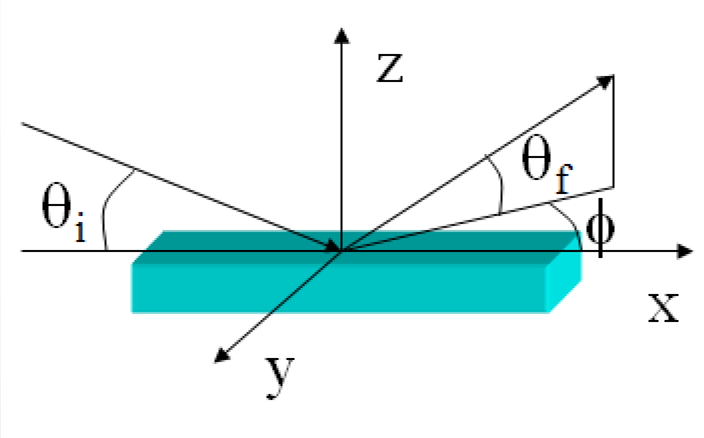
  $\theta_i = \theta_f$ and $\phi = 0$

- Reveals information about in-plane structure.

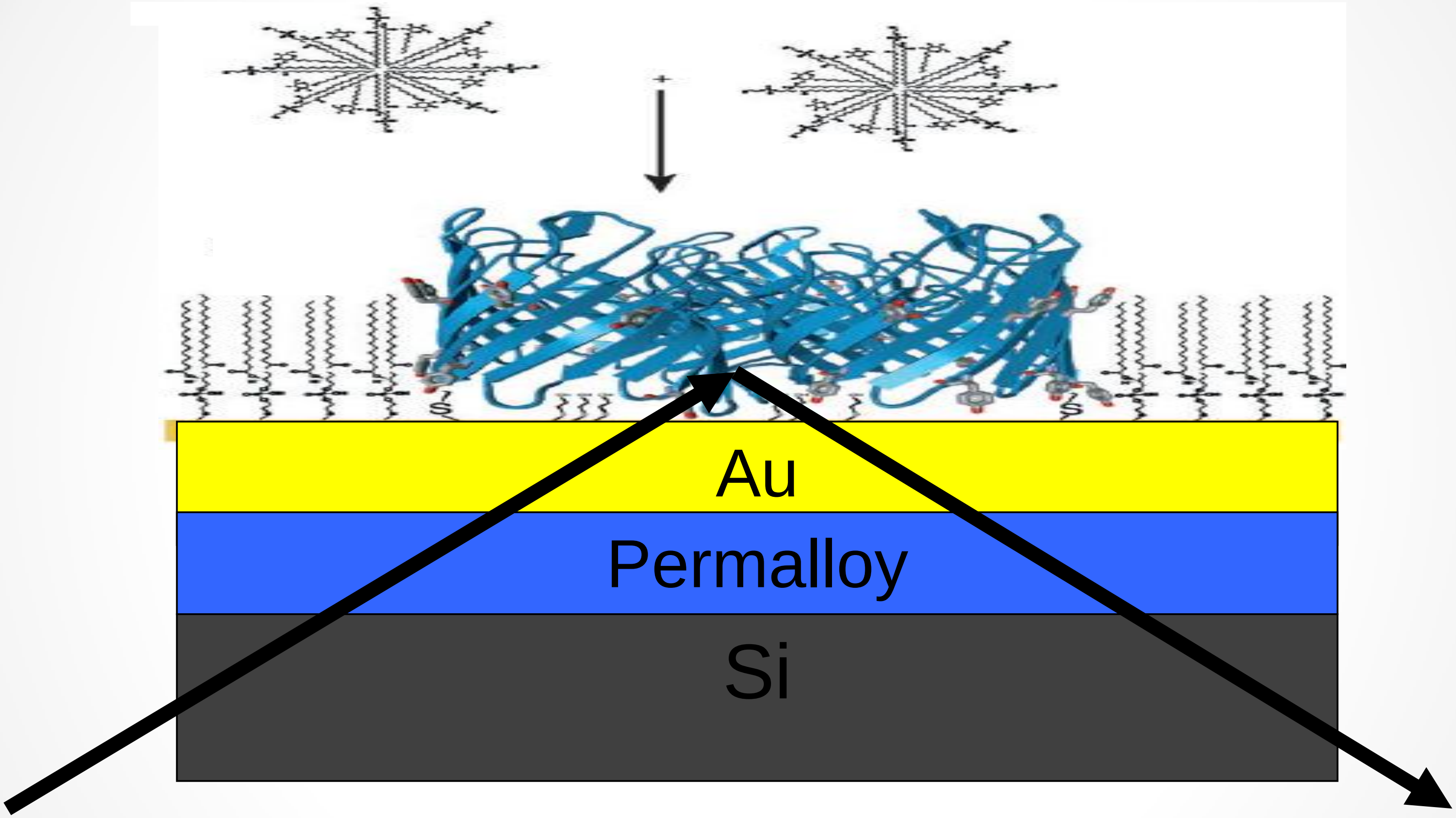



1% h-DDAB on $D_2O$

Thomas et al, 2001 (see ISIS annual report)

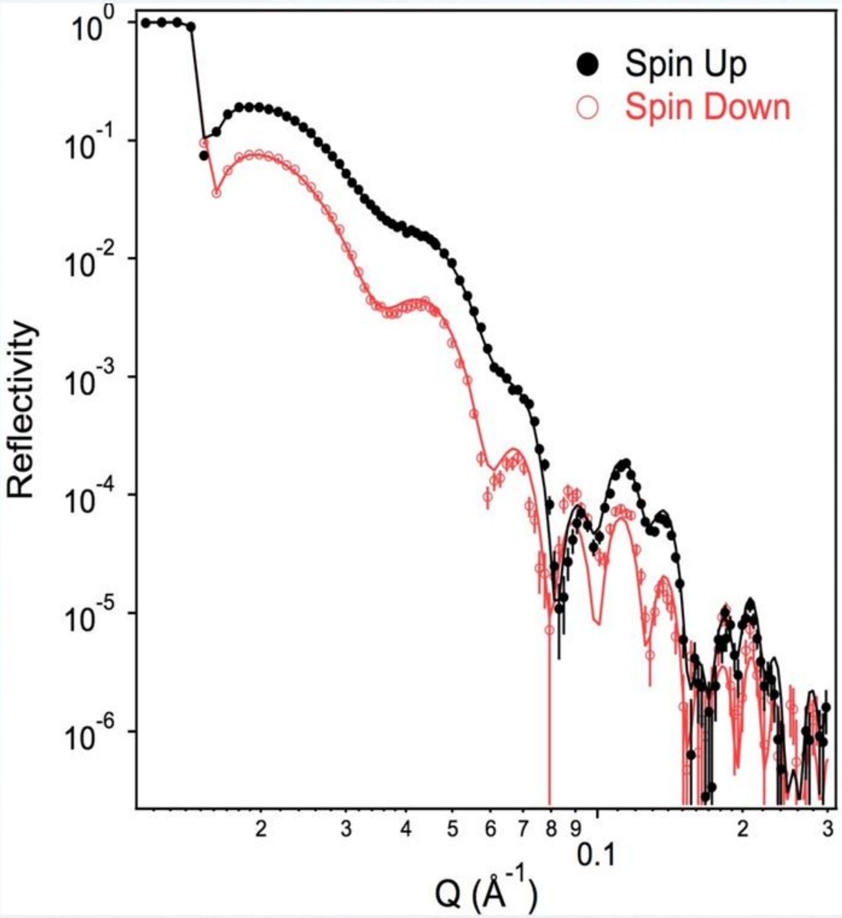


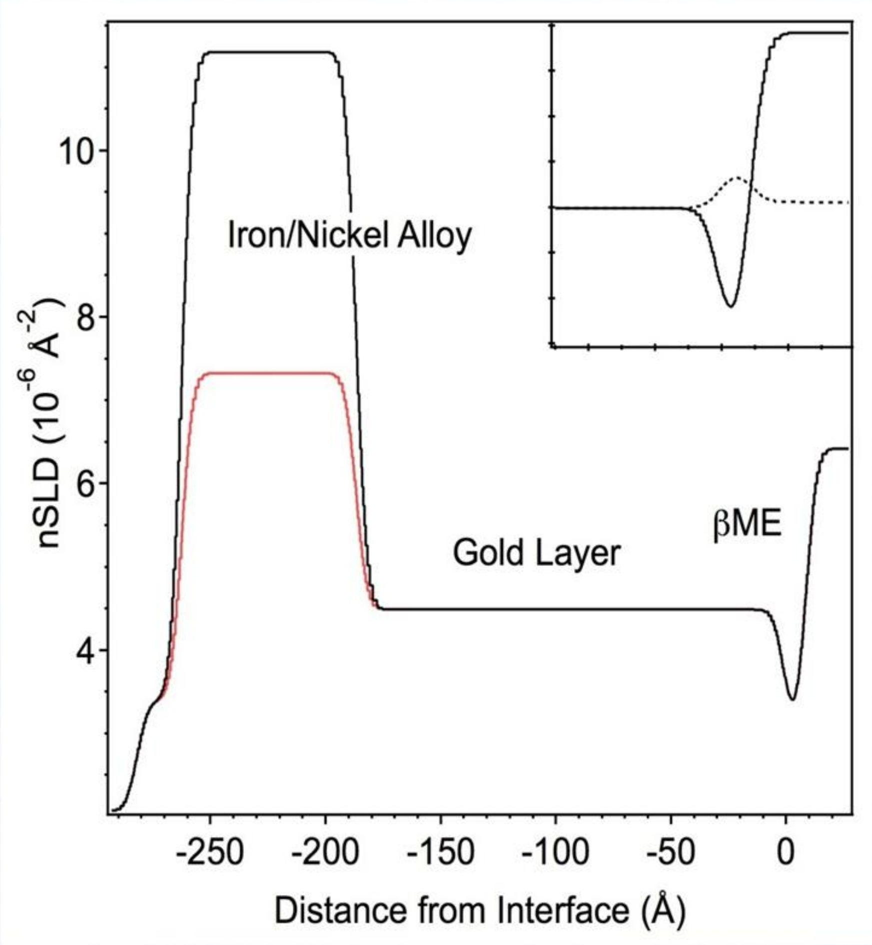
# Magnetic Reference Layers



•Data from NIST

•The use of magnetic reference layers to study biomolecular materials



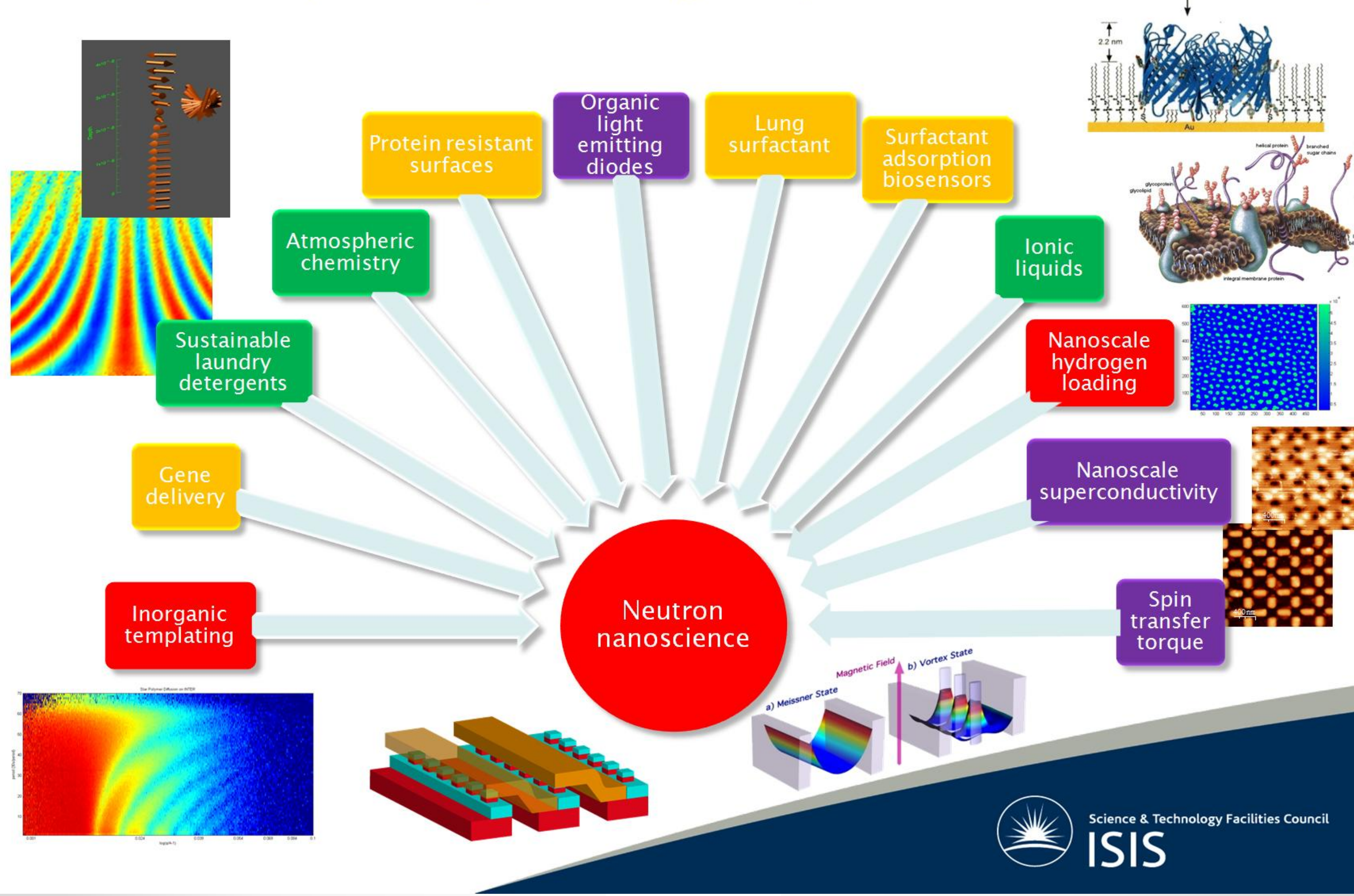

- Layer 8.2 ± 0.6 Å
- $V_f$ 0.5 - 0.8

S.A. Holt et. al.

ansto
Nuclear-based science benefiting all Australians

# Science Themes

## Fundamentally driven, *technologically relevant*

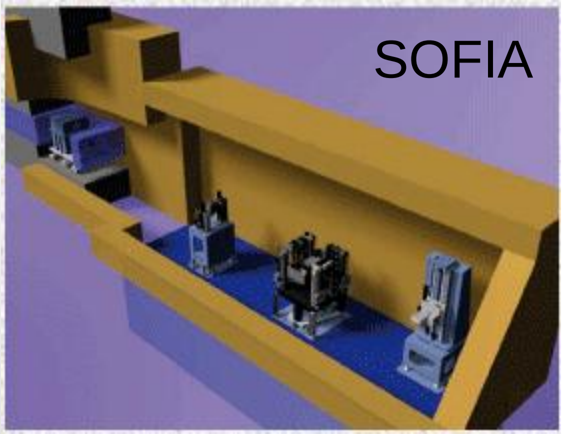


# Current Instrumentation

- 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

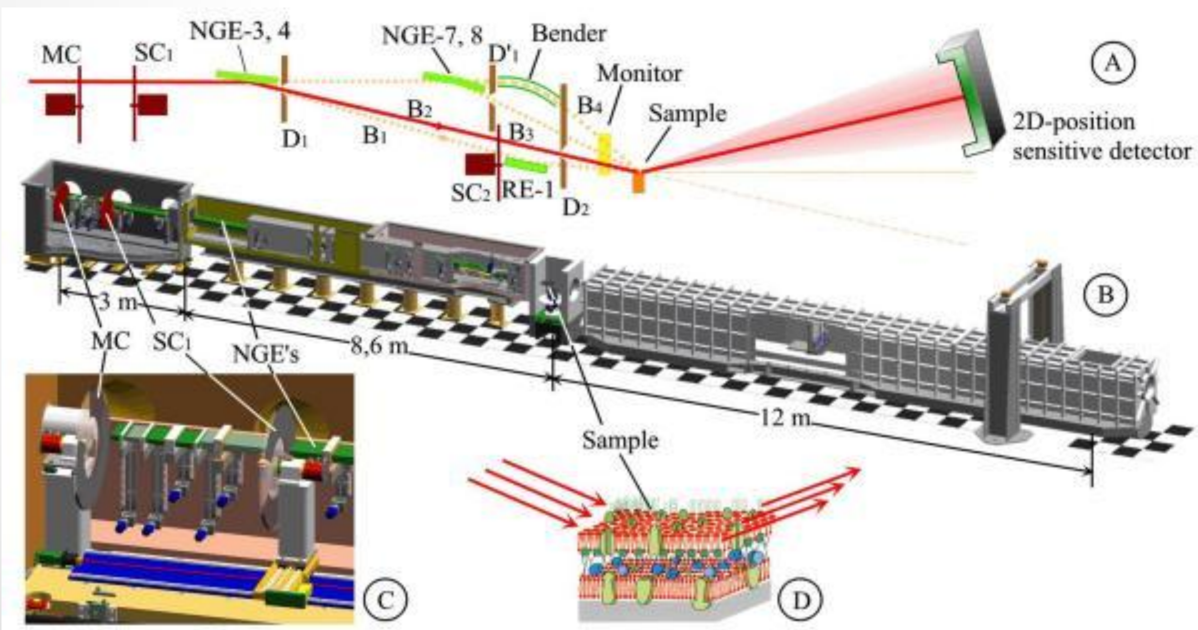


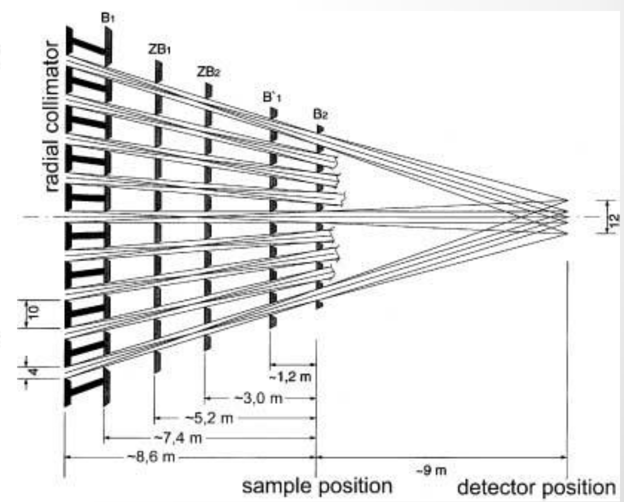


CRISP

# Current Instrumentation

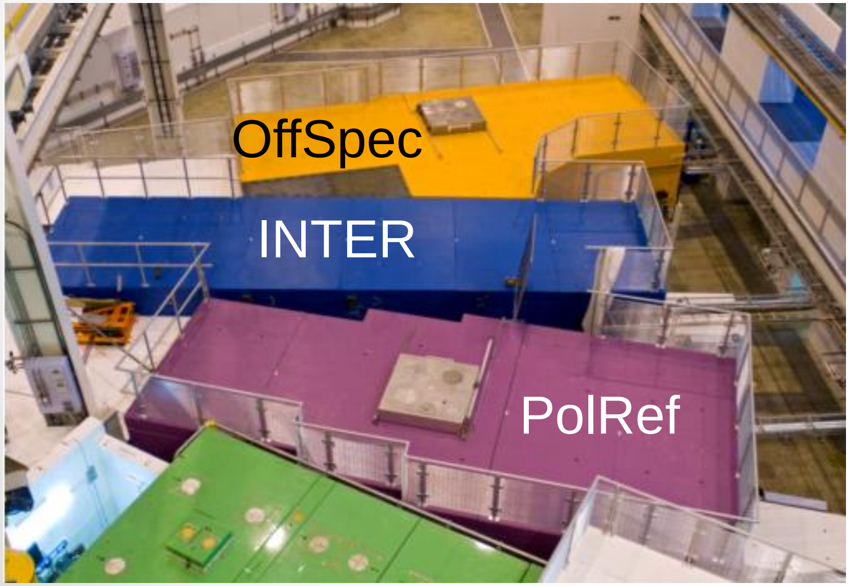
- Reflectometers are typically consider to be relatively inexpensive because they often have simple collimation and a small detector array.
- REFSANS at FRM 2 is possibly the most complex as it combines NR, SANS and GISANS.

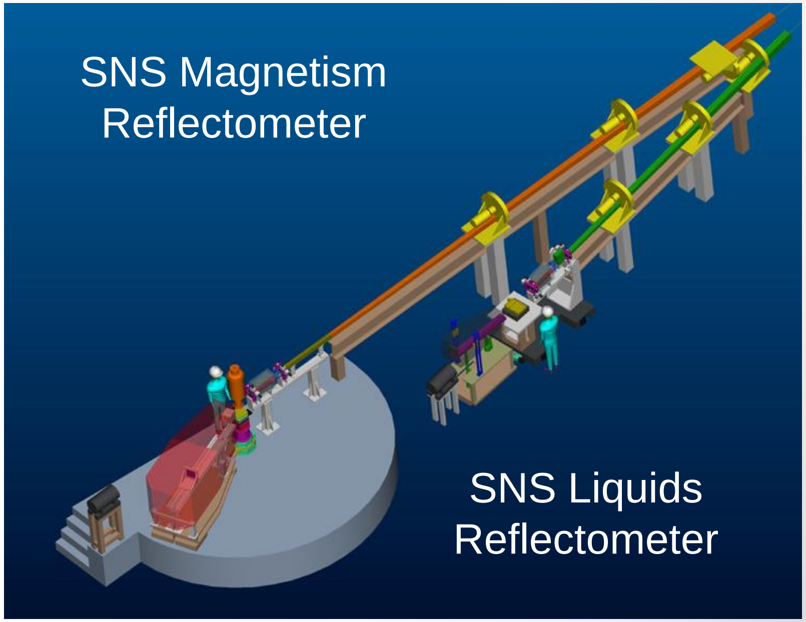


# Primary Instrument Design Criteria

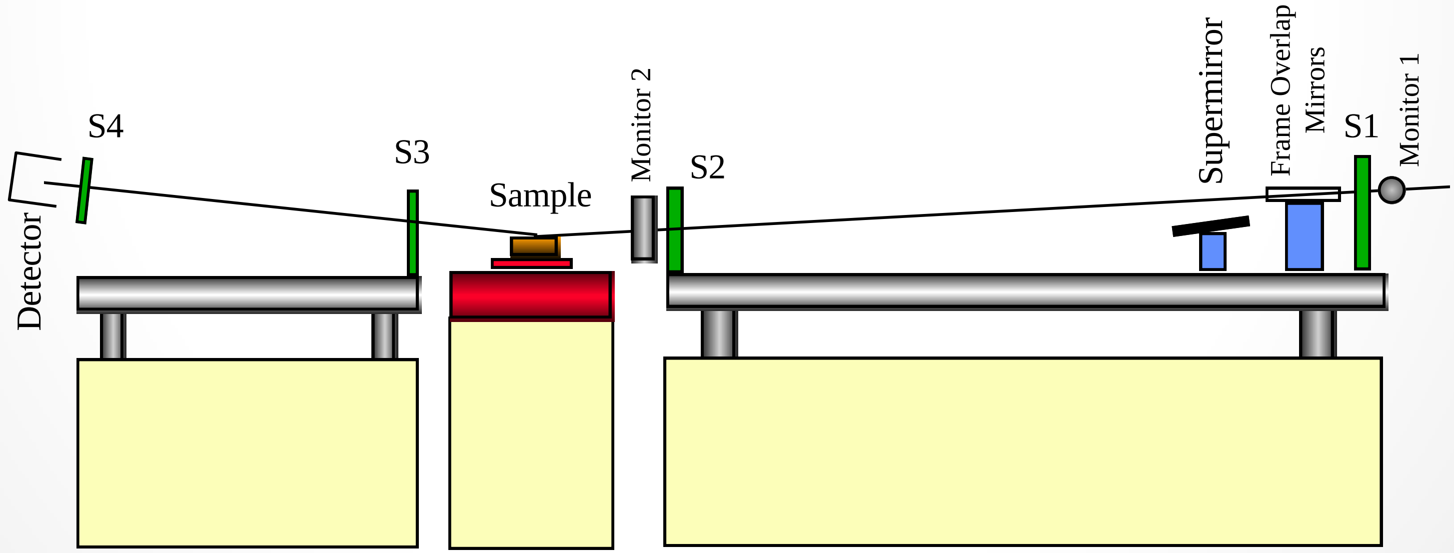
- Low Background.
  - This is required whatever the design and is the most important consideration
- What is the science case?
  - Is this is a general purpose instrument?
  - Are free liquid surfaces to be studied?
  - Magnetic systems?
- Design must be optimised to deliver science!



# 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

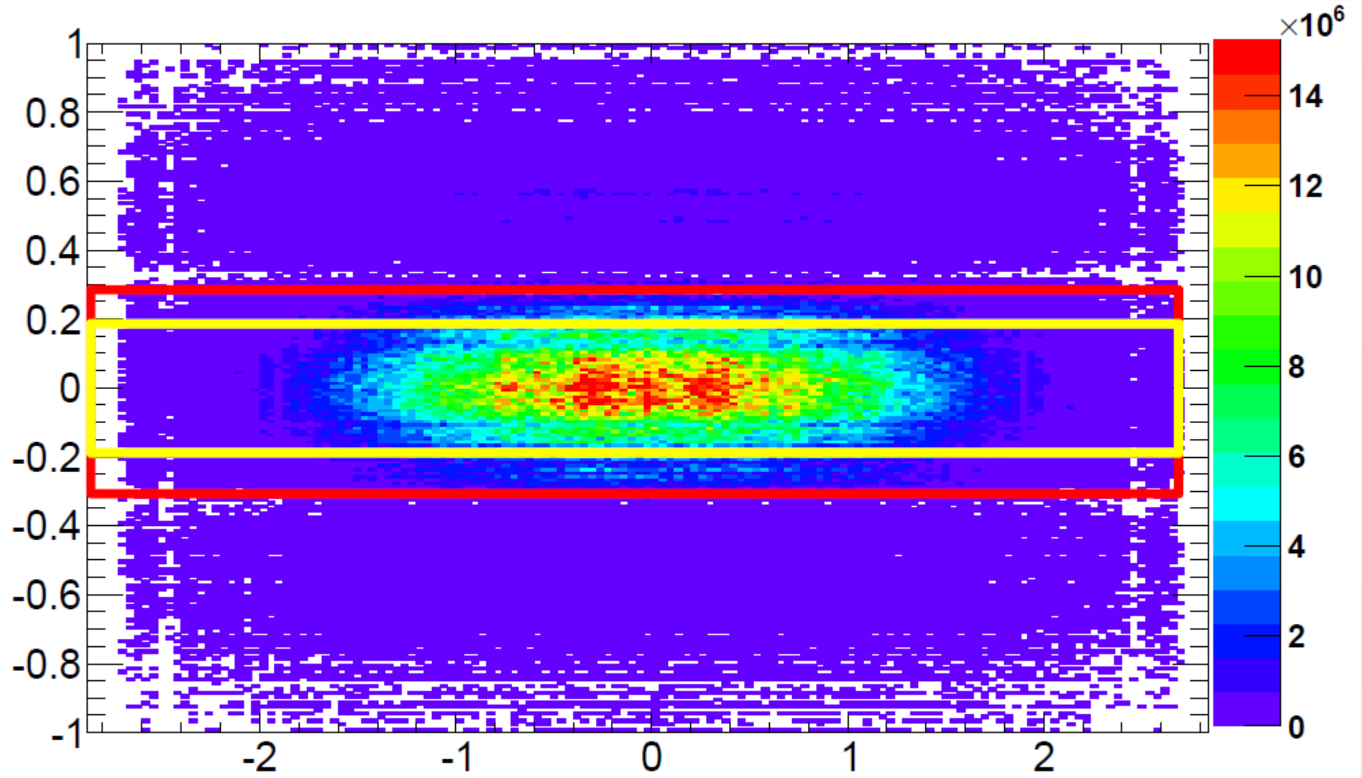


# Resolution

- 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 $\theta$ or $\lambda$ may be the dominant term.
- It is important to consider this and how to model it.

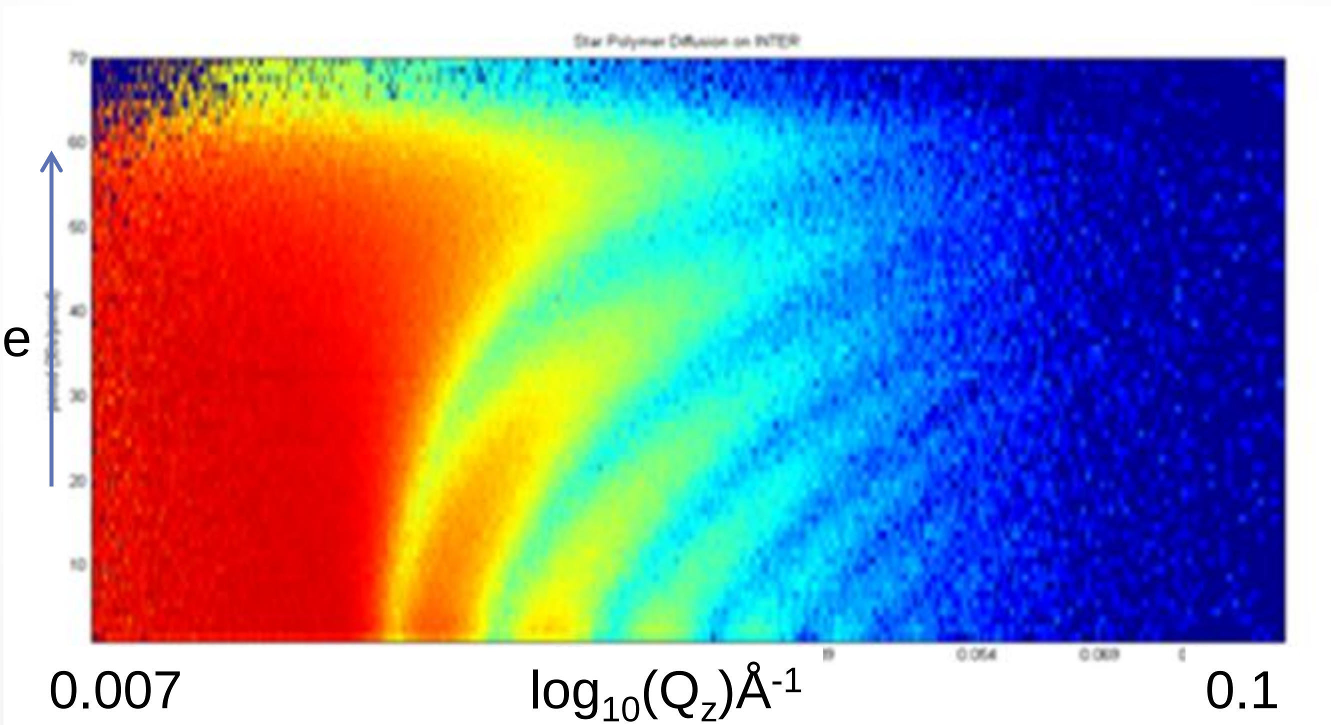
# Beam Uniformity

- 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.



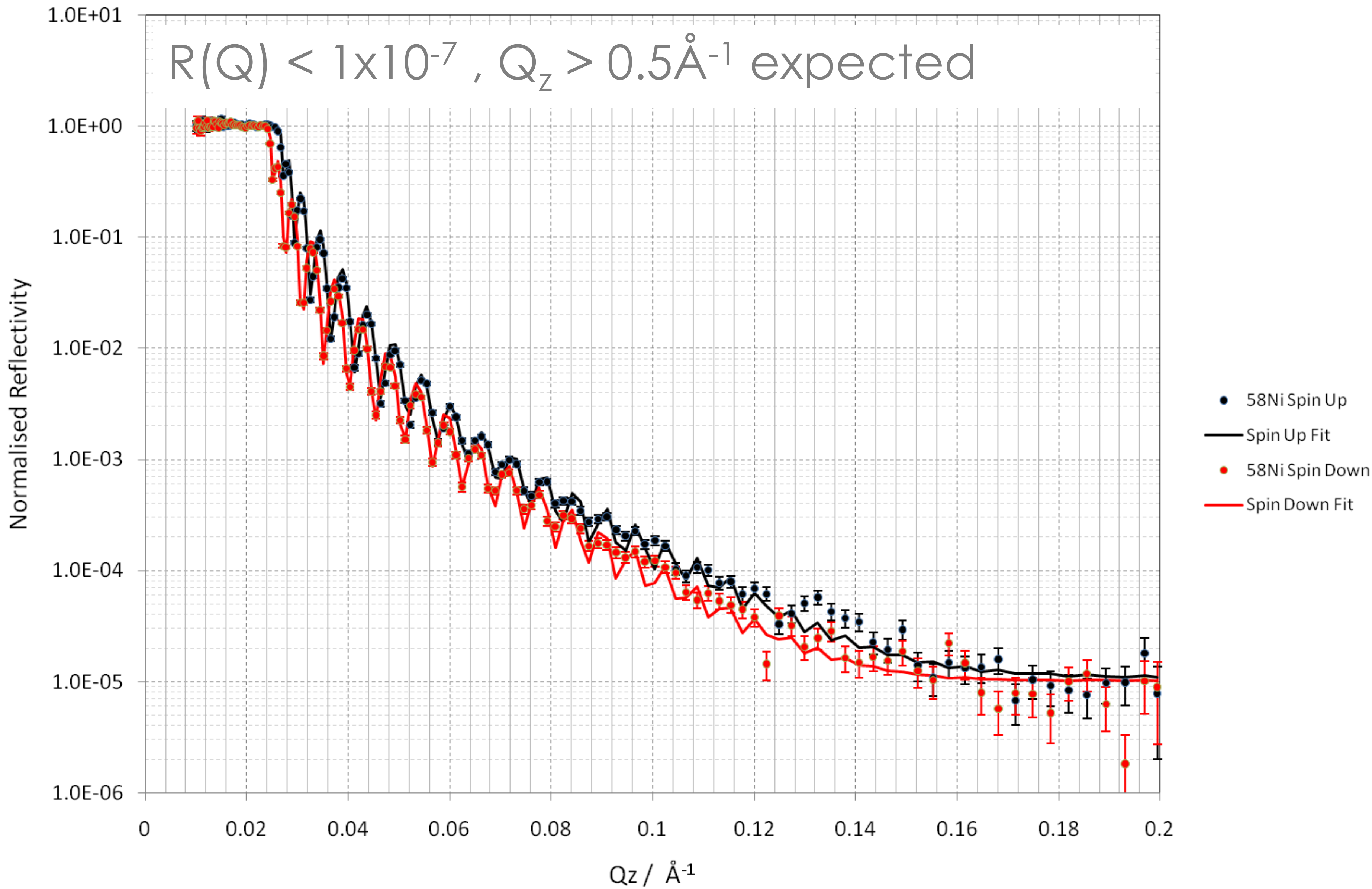
# Bandwidth and Kinetics

- 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.




INSIS

# Dynamic Range

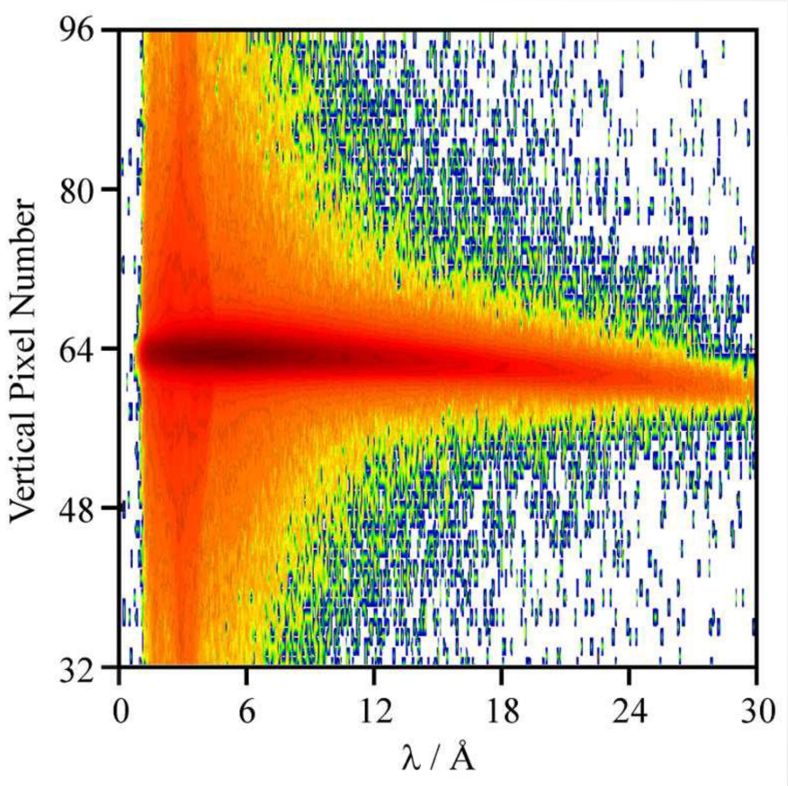


# Gravity

- 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

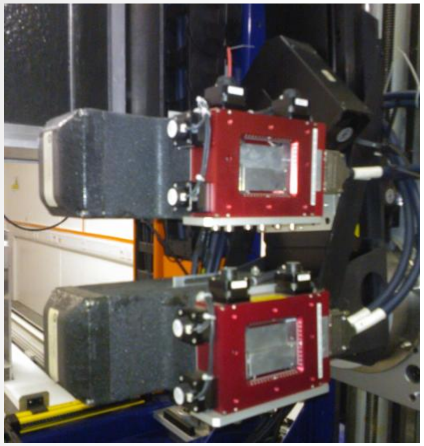
- Detectors
- Apertures/ Collimation Slits
- Choppers
- Polarisers and Analysers
- Sample Stage
- Focussing Elements

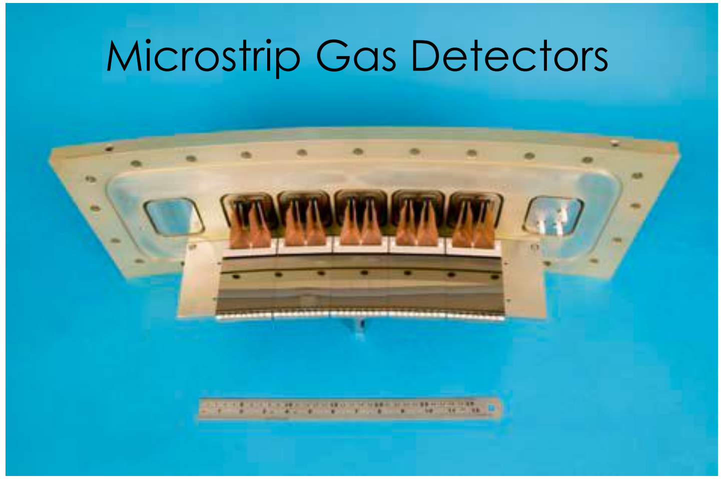
# Detectors

- 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.

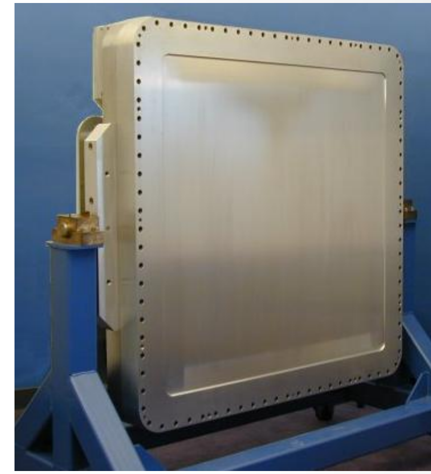
# Some Possible Detector Types

Single well shielded Helium tubes for specular reflectivity






Multiwire proportional counters



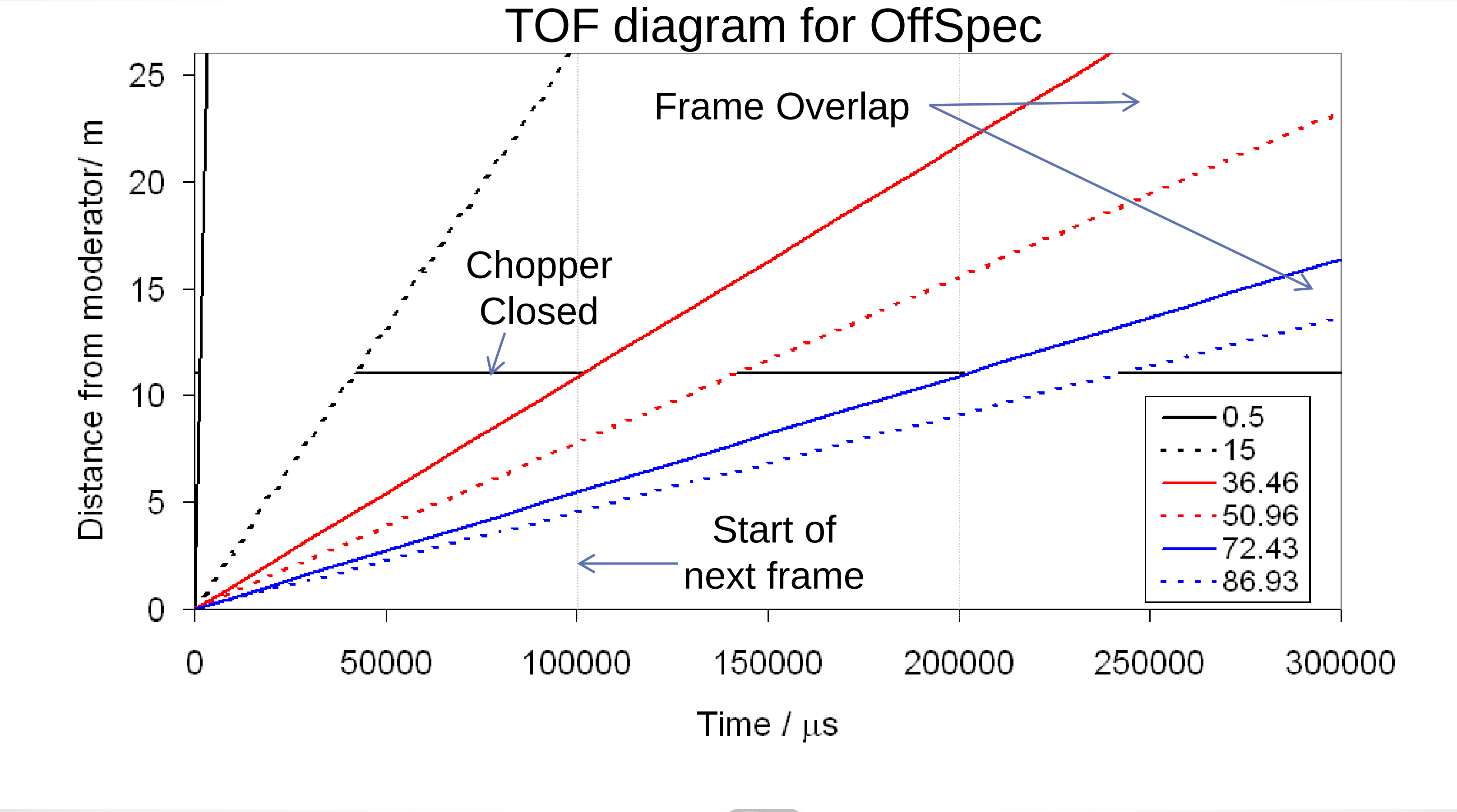
ILL resistive wire monoblock detector

Scintillator based detectors

- Direct scintillator to fibre coupled
- Wavelength shifting fibre coupled

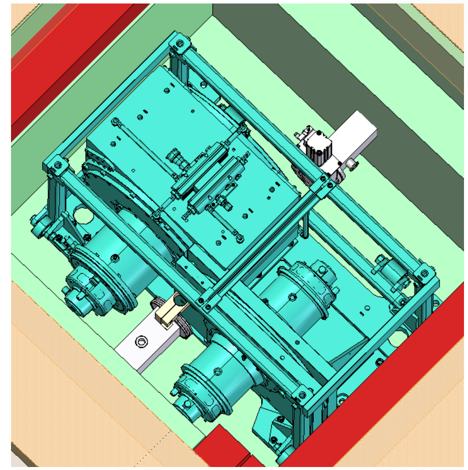
# Choppers

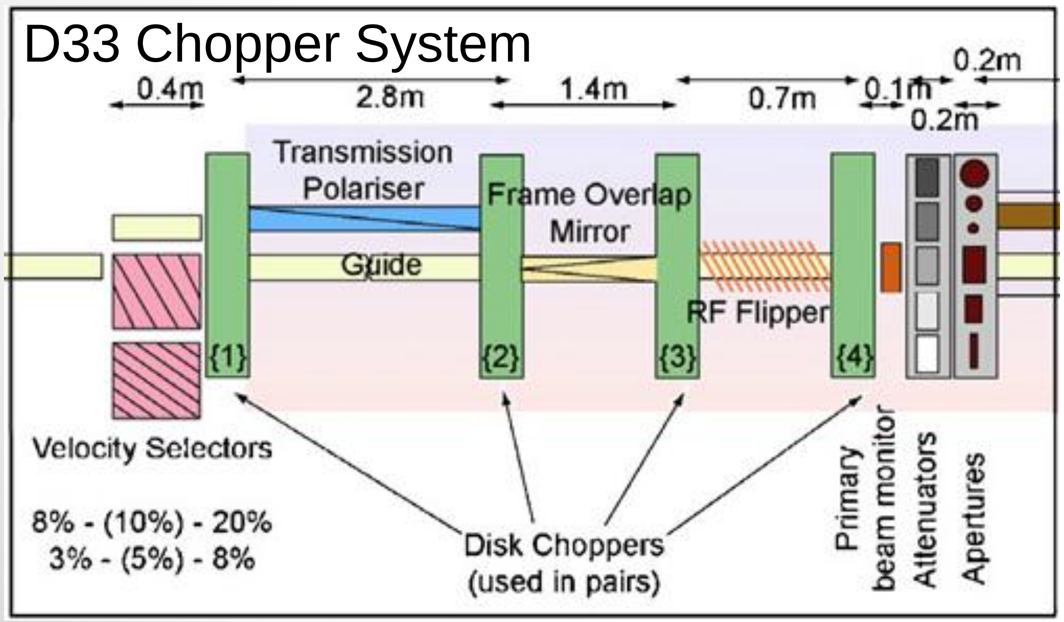
- Define the useful wavelength band or help to reduce background
- Slightly different problems on reactors and pulsed sources.



# 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)

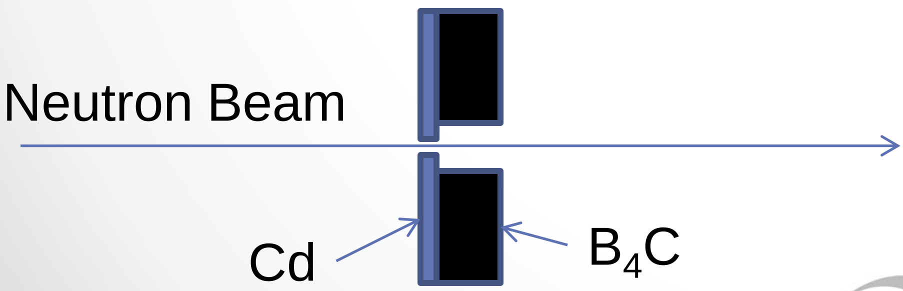


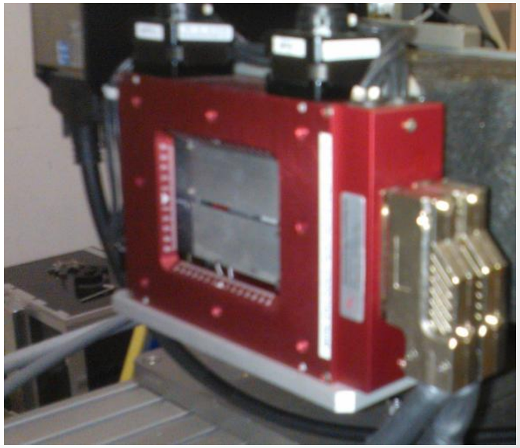



- Reactors
    - Smaller devices synced to each other to provide varying resolution
    - Thinner blades as instruments are typically on long guides using cold/slow neutrons that are easily absorbed.

# 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

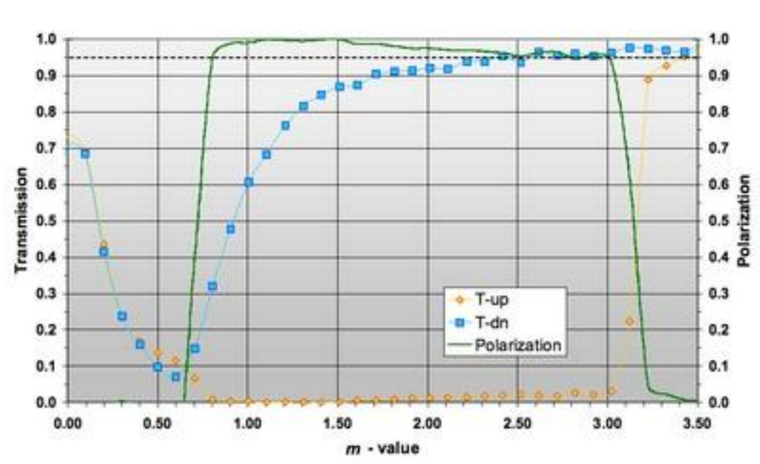
- Neutrons are spin ½ particles
- In an external field the neutrons align parallel (spin up) or anti-parallel (spin down) to the field.
- The polarisation is a scalar that expresses the relative population sizes of the two alignments

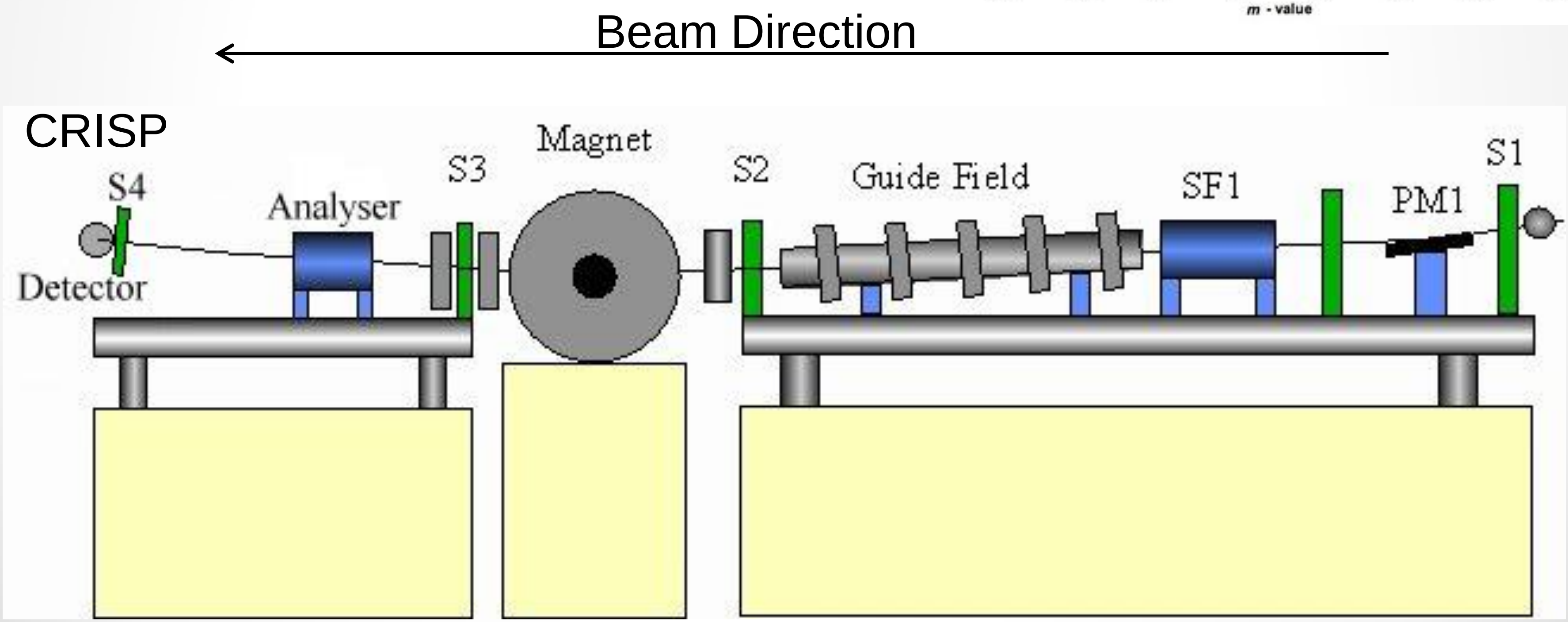
$$P(\lambda)=\frac{N_+(\lambda)-N_-(\lambda)}{N_+(\lambda)+N_-(\lambda)}$$

- Polarisation is never perfect and is usually wavelength dependent.

# Polarisation

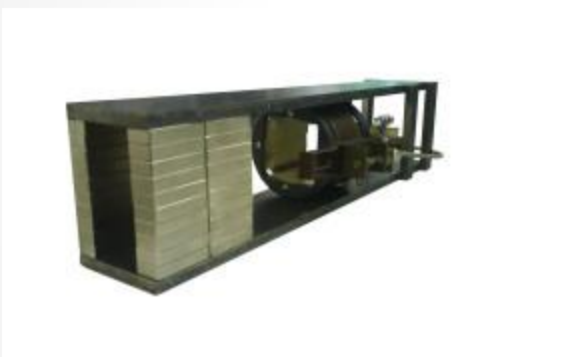
- Because of the use of ribbon collimation polarisation can be readily achieved using a polarising mirrors.

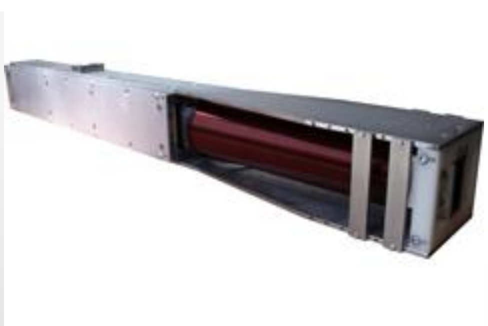


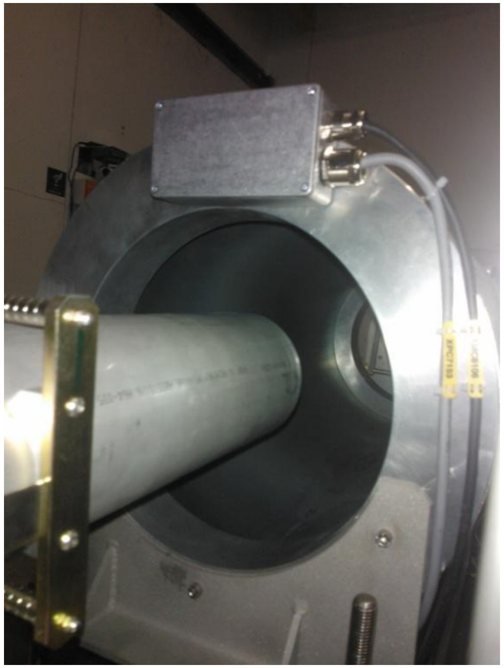



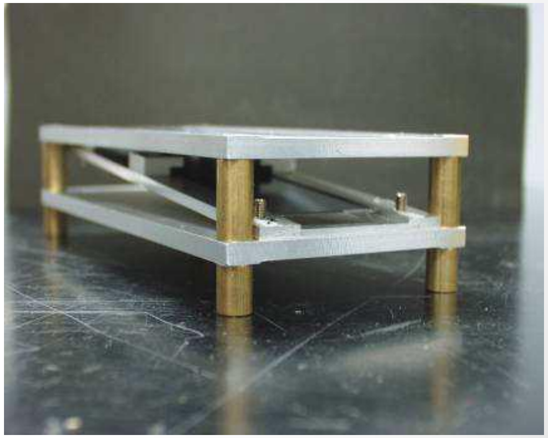
# Spin Flippers

- 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

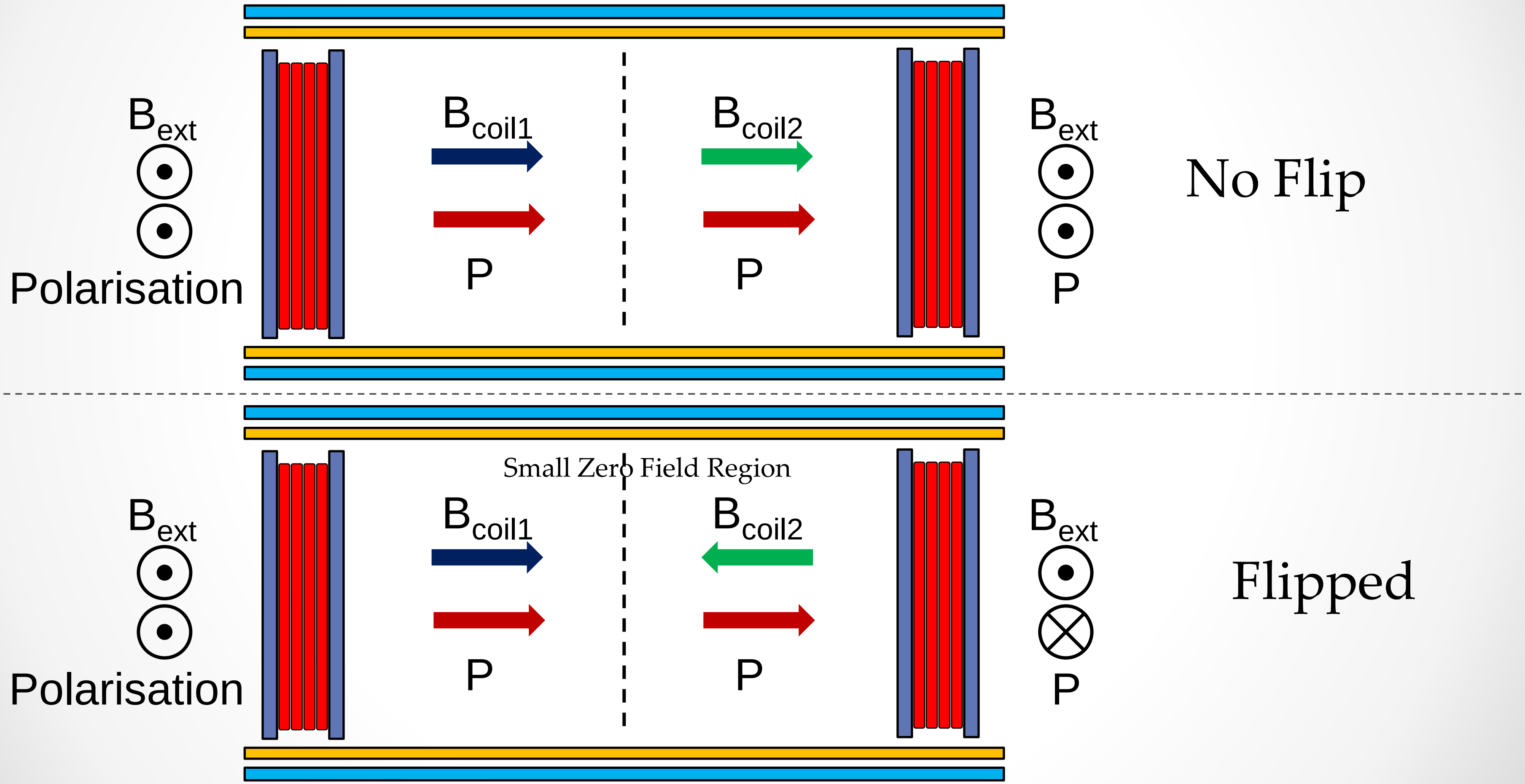








# Drabkin Spin Flipper

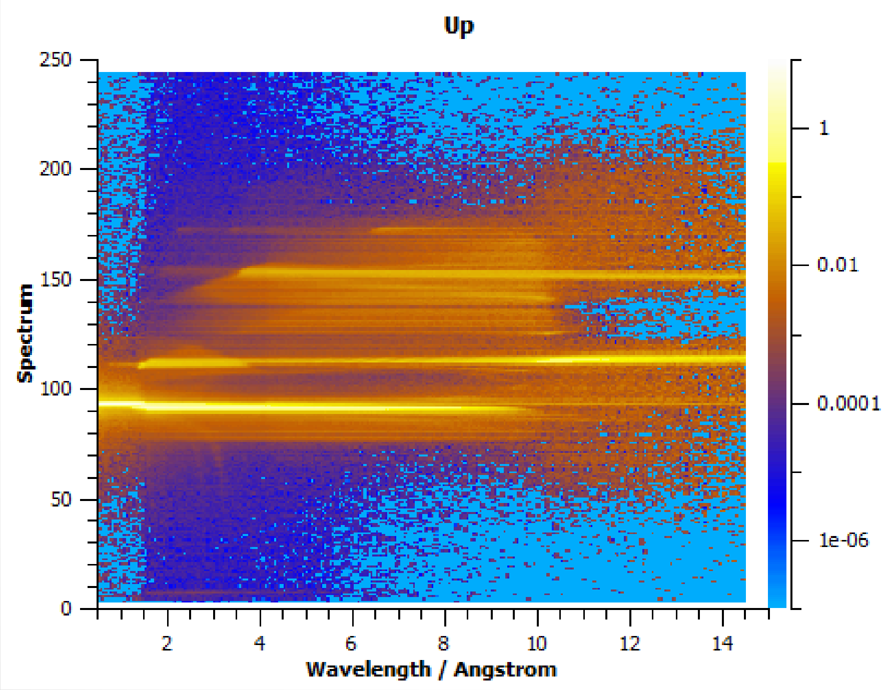


# Polariser and Analyser

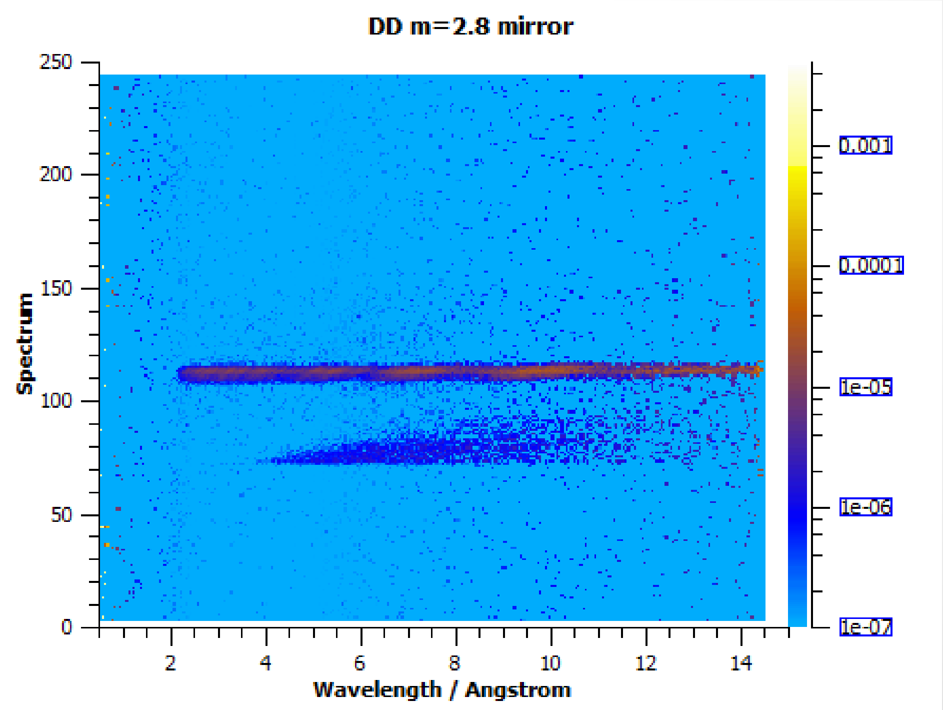
- Choice depends on application, wavelength band and budget.
- Polarised $^3He$
  - Ideal for monochromatic beams or where only a short bandwidth is needed
  - Polarisation can be traded for intensity and bandwidth.
  - On beam pumping (SEOP) can provide stable polarisation
  - Essential if an undistorted 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

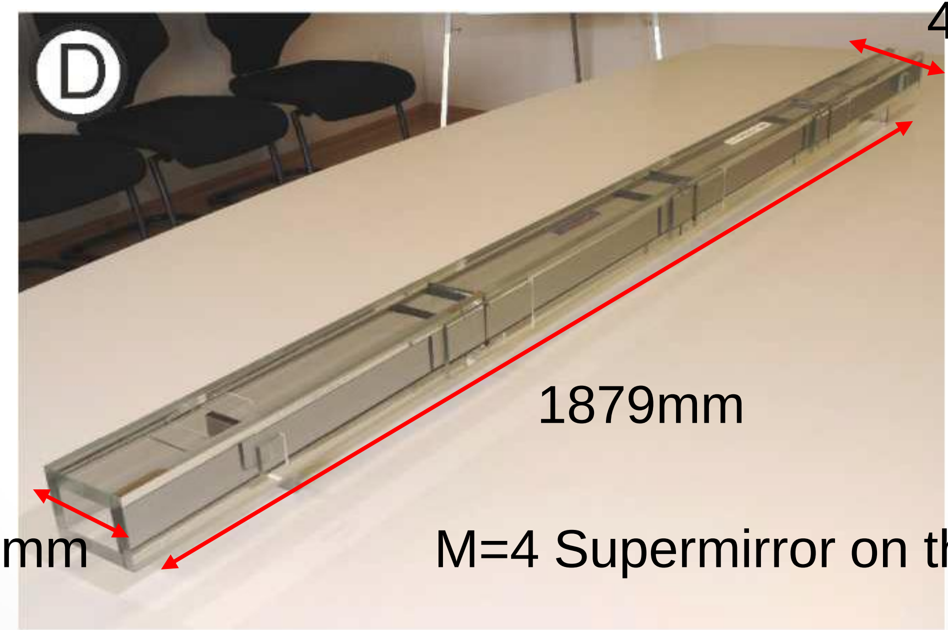


# Sample Stage

- 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.

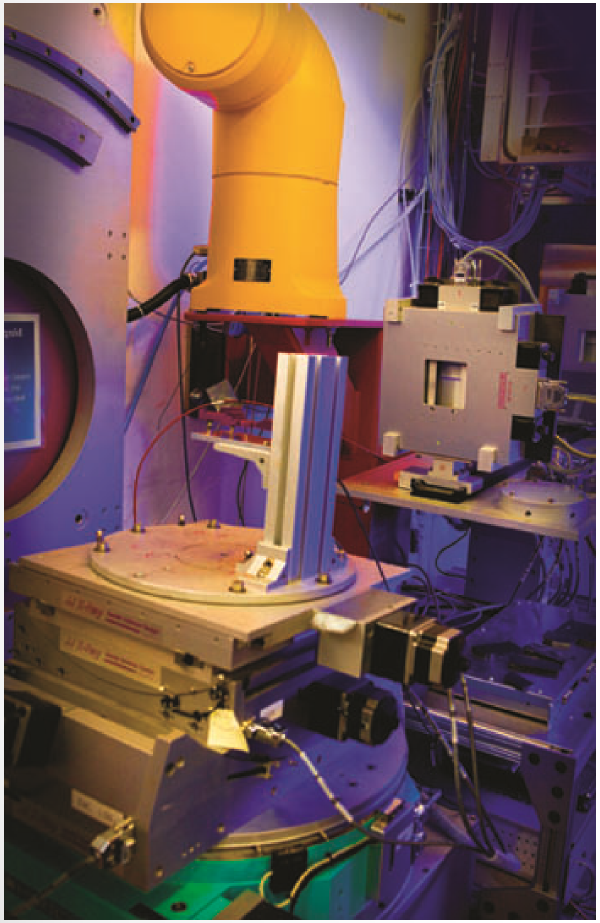
# Focussing

- 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.



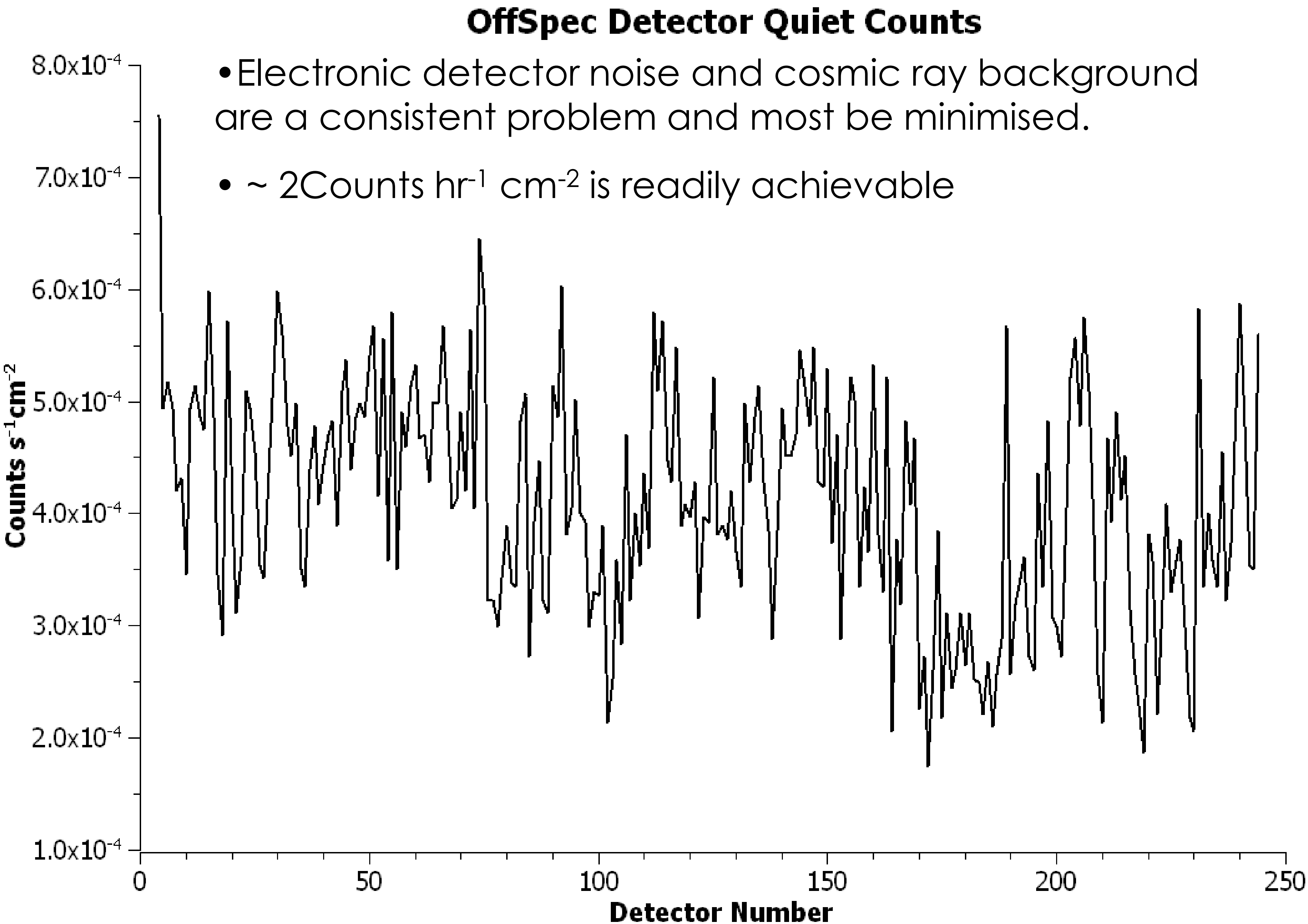
# Sources of Background

SNS Liquids Reflectometer

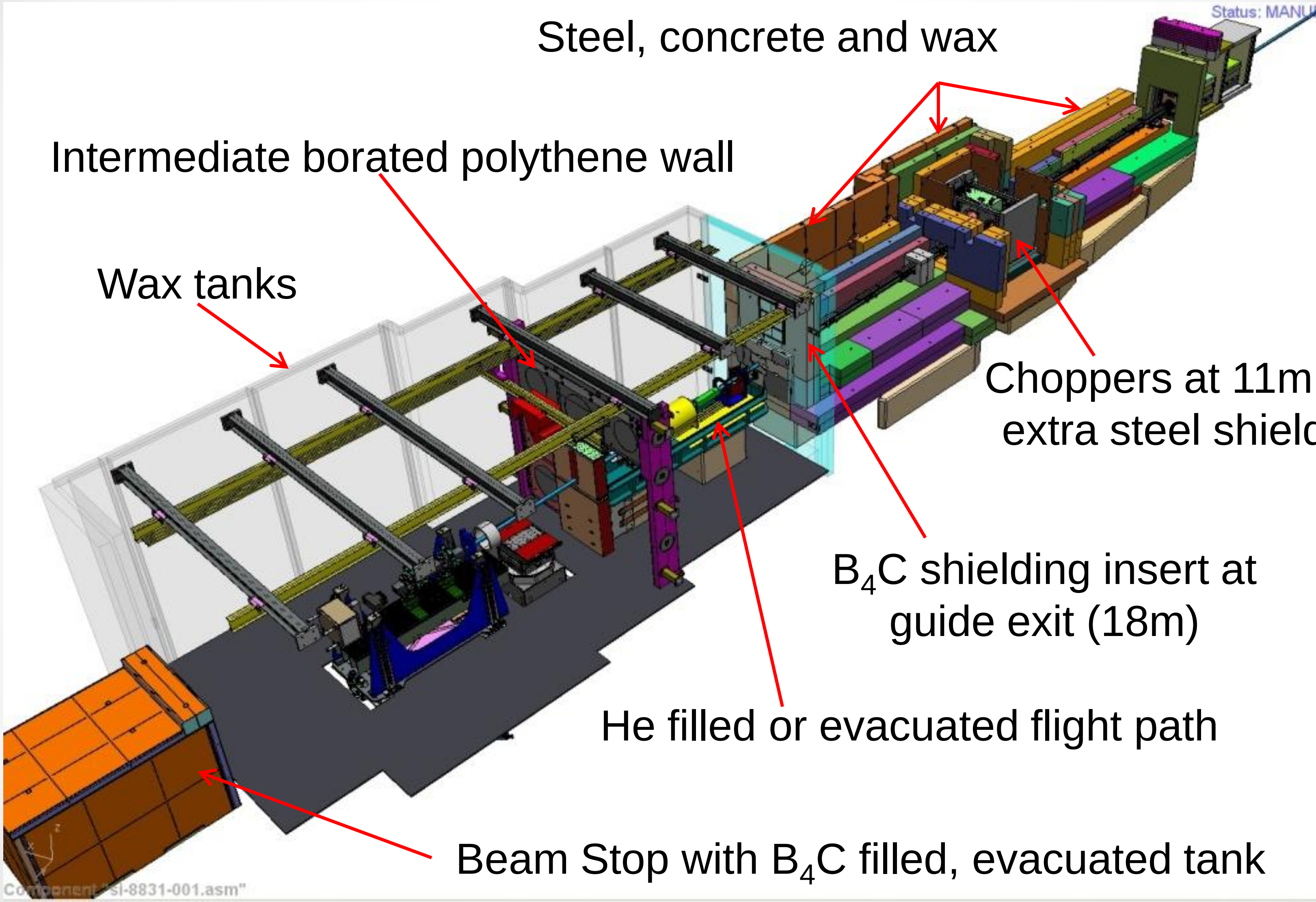


- The Sample
  - Incoherent scattering from the sample is the most significant limitation to measuring reflectivities below $1 \times 10^{-7}$.
  - Need to remove hydrogen and reduce sample volumes.
- Air Scattering
  - Evacuate flight paths (REFSANS, D17)
- Stop high energy neutrons as early as possible.
  - 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!

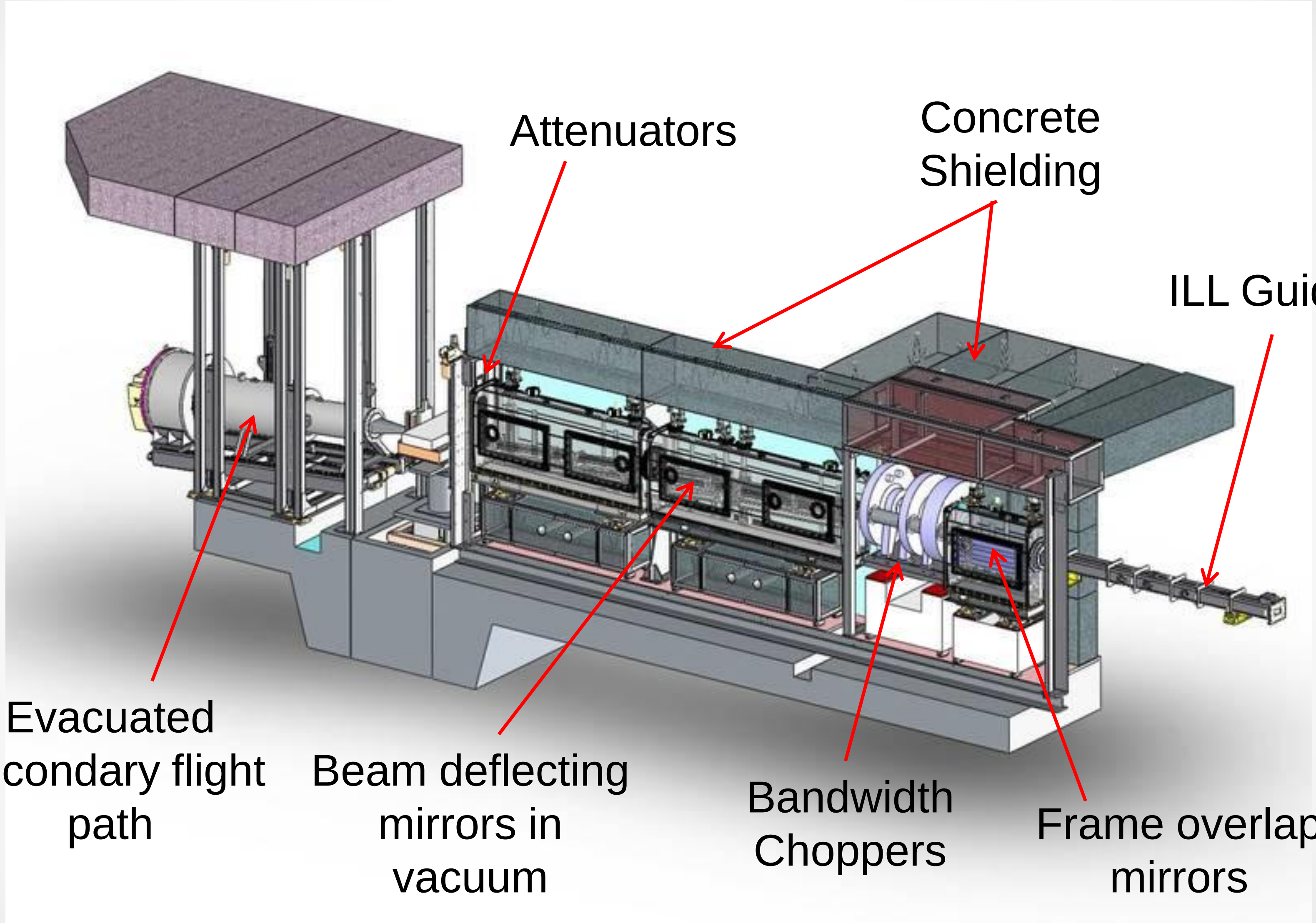
- Electronic detector noise and cosmic ray background are a consistent problem and most be minimised.
- ~ 2Counts $hr^{-1}$ $cm^{-2}$ is readily achievable



# PolRef ISIS TS2



# FIGARO, ILL

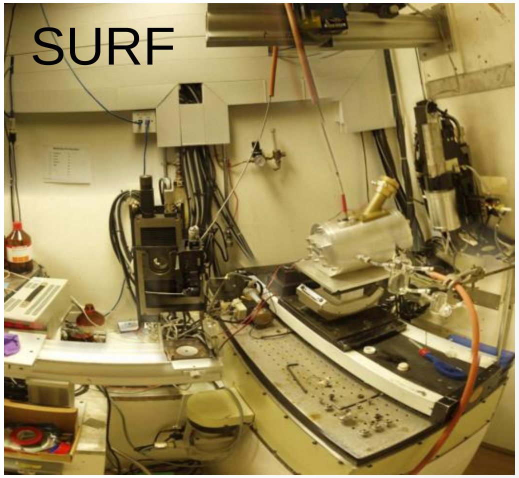

Attenuators
Concrete Shielding
ILL Guide
Evacuated secondary flight path
Beam deflecting mirrors in vacuum
Bandwidth Choppers
Frame overlap mirrors

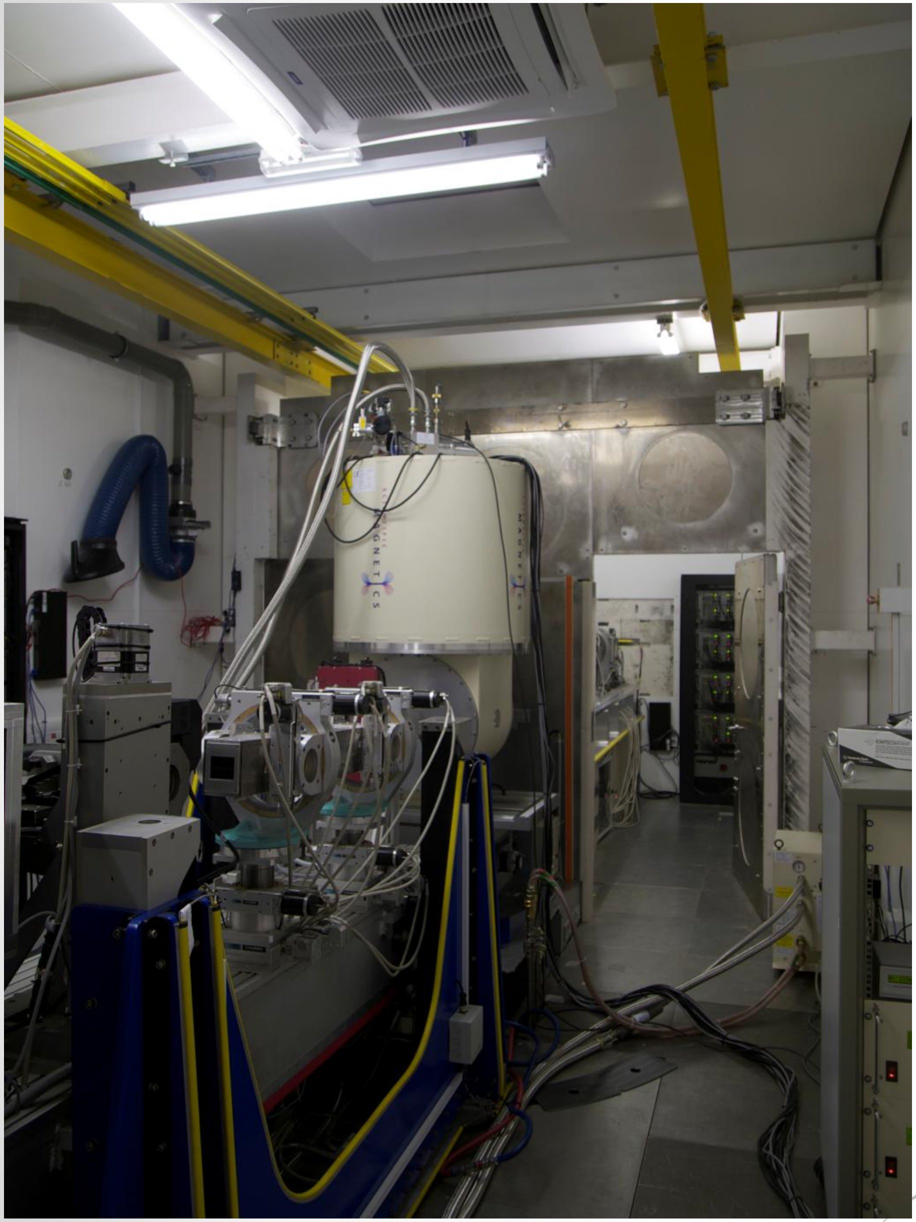
# Sample Background

- 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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