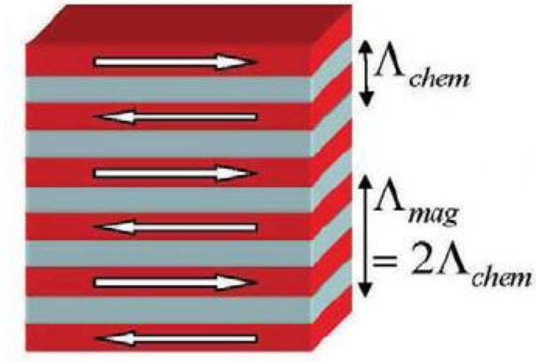
Australian Government

Reflectometry Instrumentation

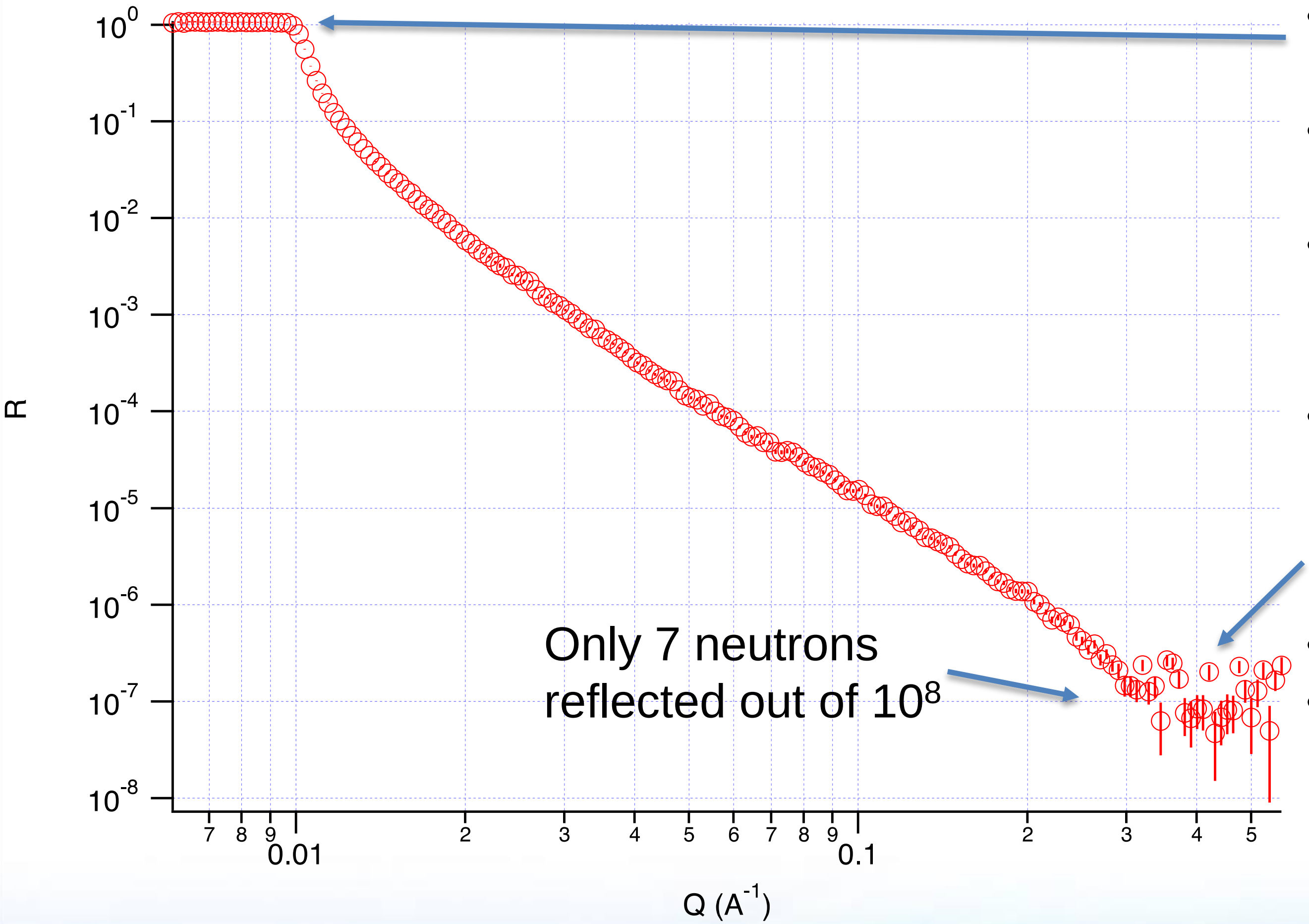
Andrew Nelson

“Citius, Altius, Fortius”

- Faster measurements
- Higher Q
- Lower reflectivity
- Wide dynamic Q ranges
- Smaller samples
- More complicated sample environments



Overarching Reflectometry Requirements



- Detector copes with high count rate
- Detector copes with high local count rate
- Detector efficient for relevant wavelengths

- Calibrated wavelength/angles

- Good source brilliance
- v. low background

Instrument types – angular vs energy dispersive

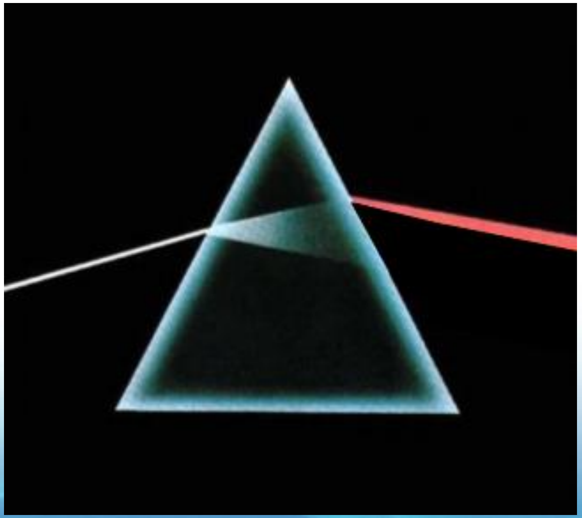
$$Q_z = \frac{4\pi}{\lambda} \sin \Omega$$

Monochromatic – Fixed λ, vary Ω (0 - 5°)

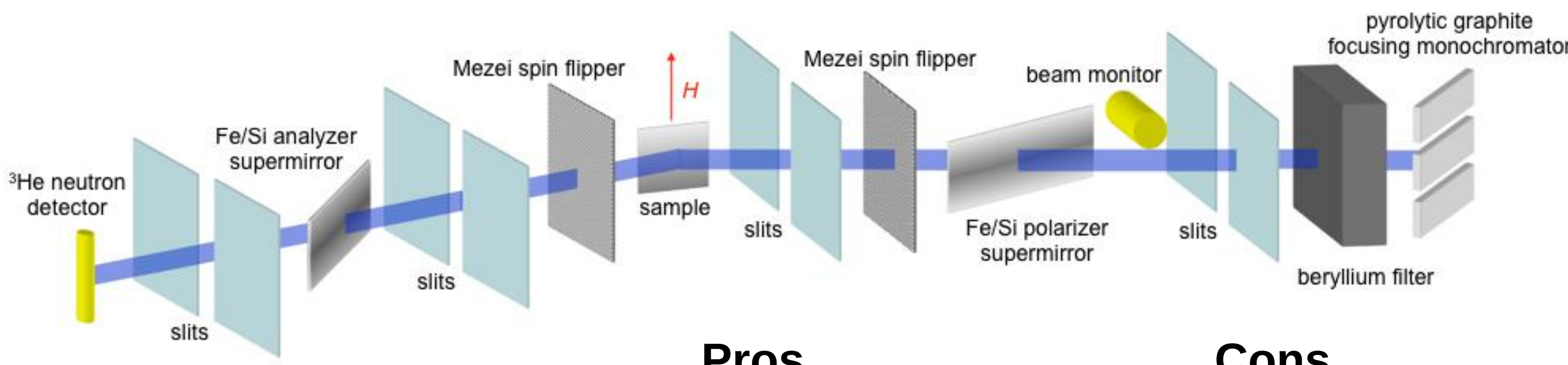
λ = 1.54056 Å (for X-rays)

λ = 4.75 Å (for neutrons)

Energy Dispersive – Vary λ (1.5 – 30 Å), fixed Ω



Conventional monochromatic at reactor

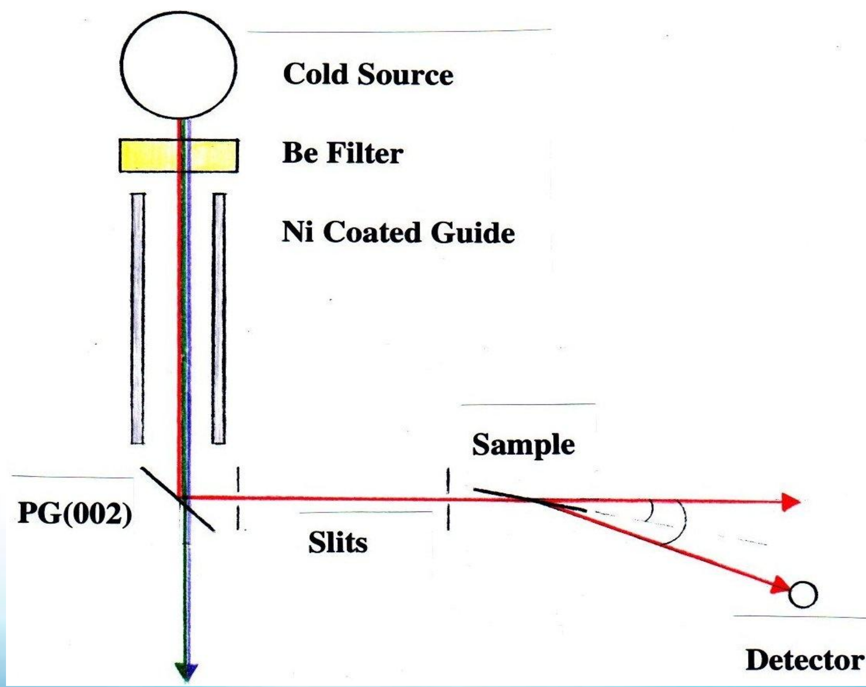


Pros

- Relatively simple to construct / calibrate / operate
- Well known resolution function, dQ/Q ~ 2%
- Efficient use of single wavelength
- Can vertically focus monochromator
- Doesn't require end guide position

Cons

- Not suited to studying realtime processes (angular dispersive)
- Can't vary resolution
- Constant changing of slits for footprint (calibration + attenuators)
- Very small angles for low Q (hard to under-illuminate reproducibly)
- Free liquid surfaces are harder



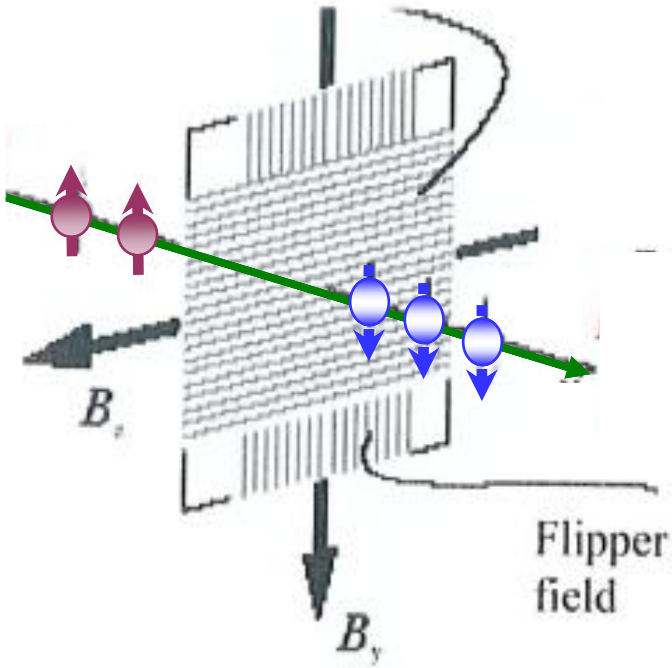
$\lambda = 4.75Å$
sample = 30mm
$\Omega = 0.22°$

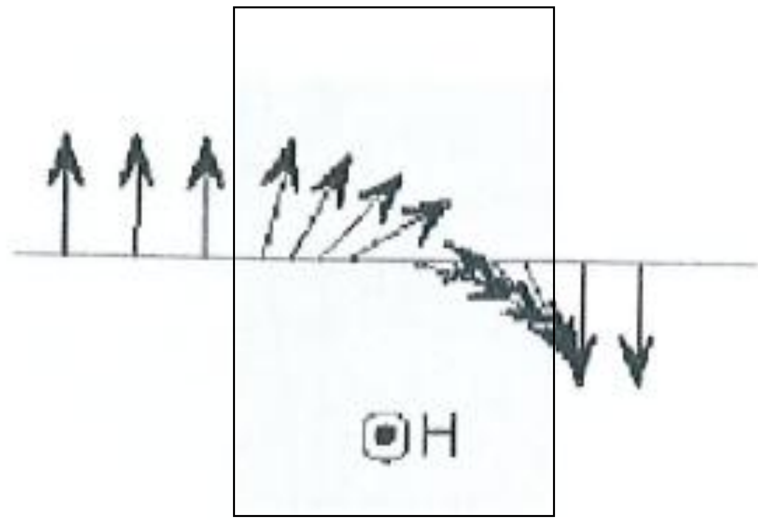
Footprint slit ~ 0.1 mm

https://ncnr.nist.gov/instruments/pbr/

Mezei Spin Flippers

Used to flip the spin state of monochromatic neutrons

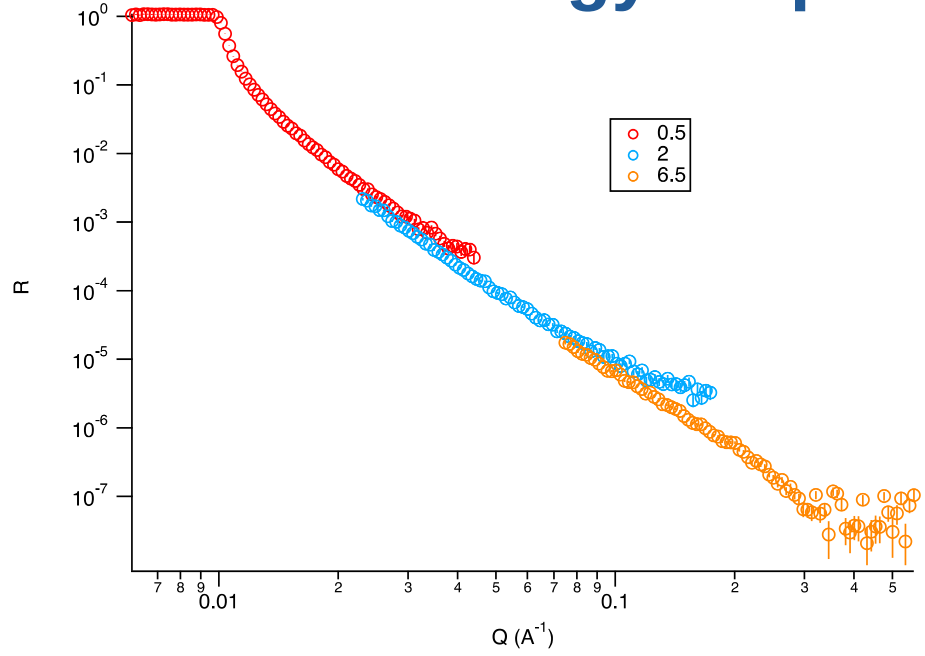




Current coils ⊥ to the beam induce a field in a solid foil that causes the spins to precess.

Sensitive to stray fields;
Current settings vary for every different wavelength

Energy Dispersive Reflectometry

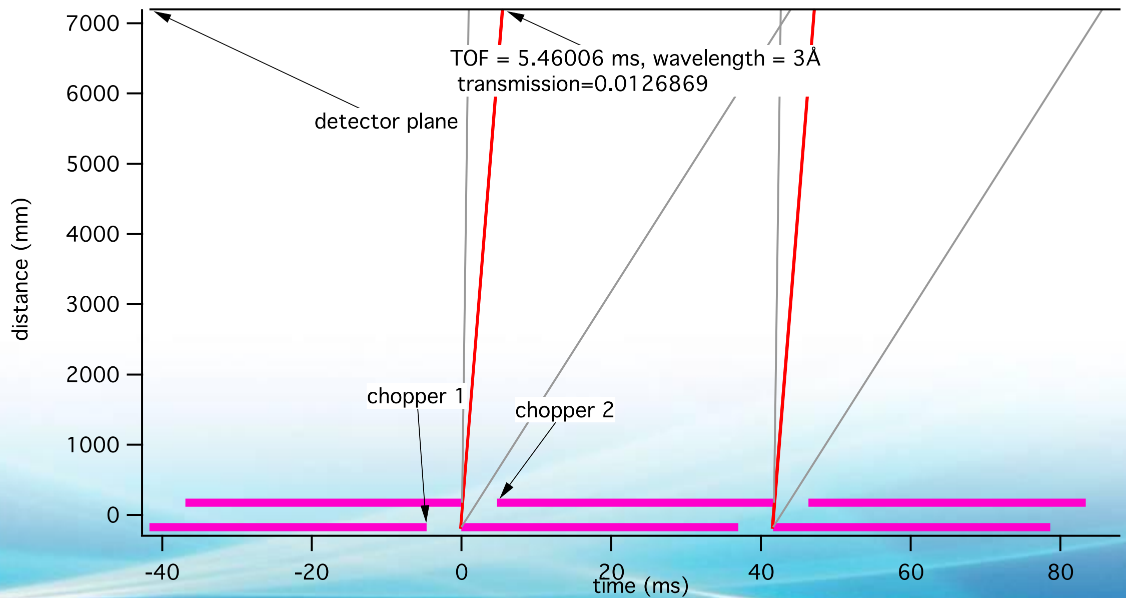


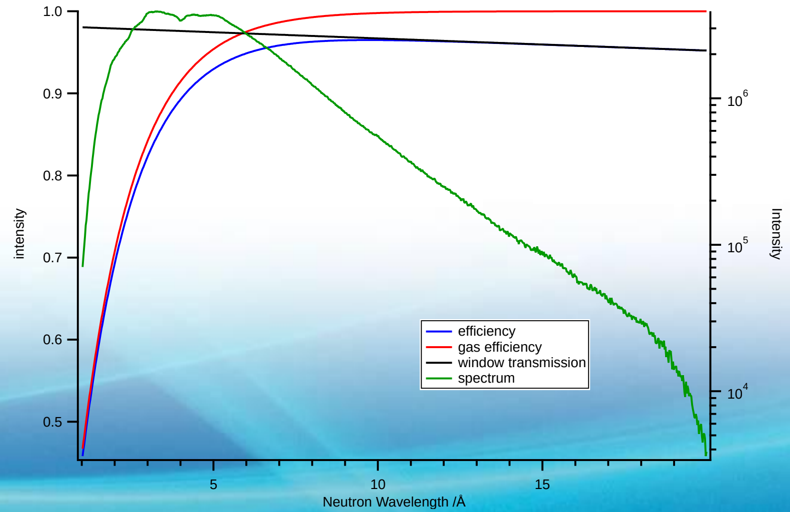
- Time of flight requires neutron pulses
  - Spallation – intrinsic pulse
  - Reactor – created by choppers
- Wavelength typically measured by time-of-flight:

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

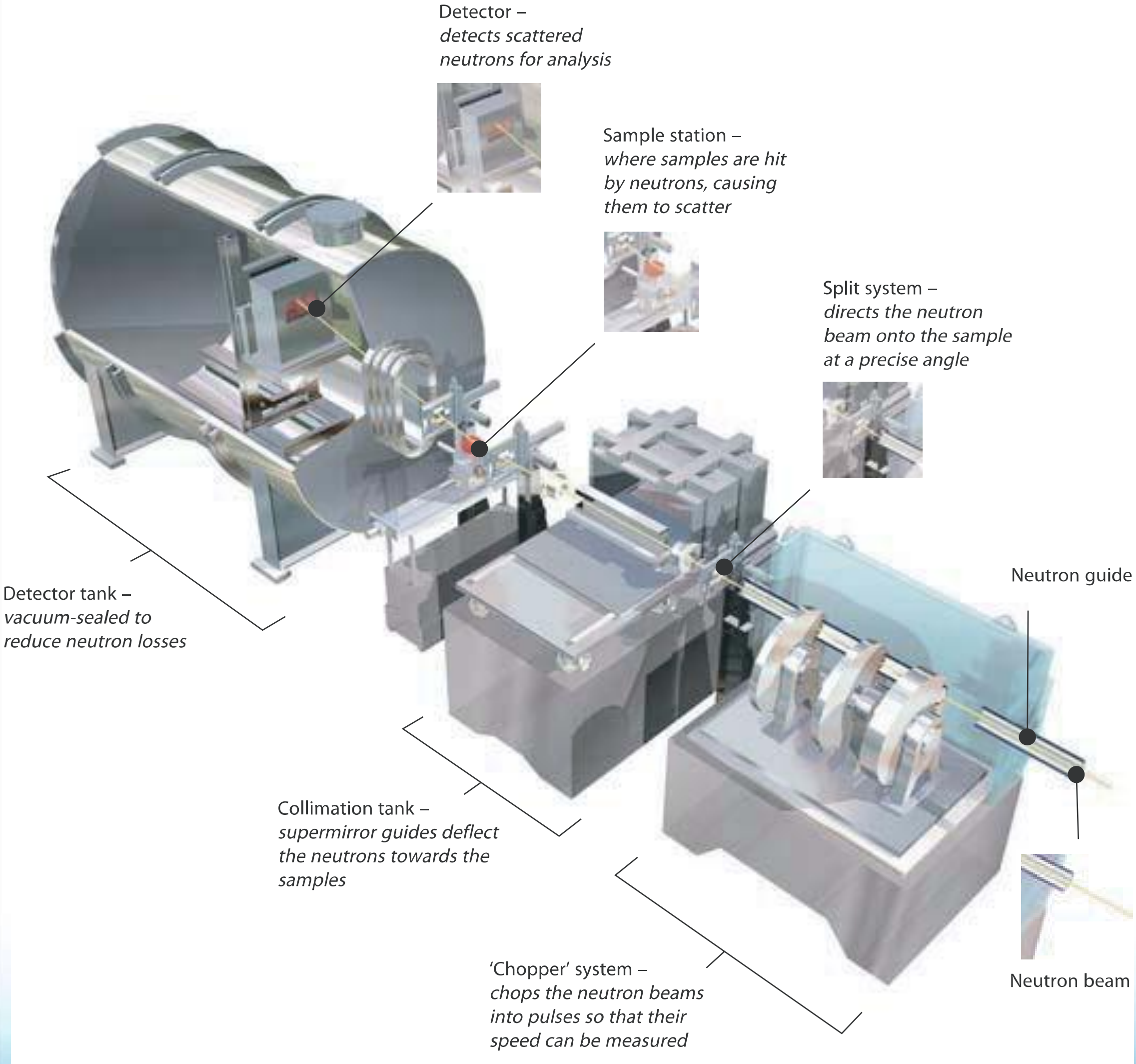
- **Dynamic Q range**: $\frac{\lambda_{max}}{\lambda_{min}}$

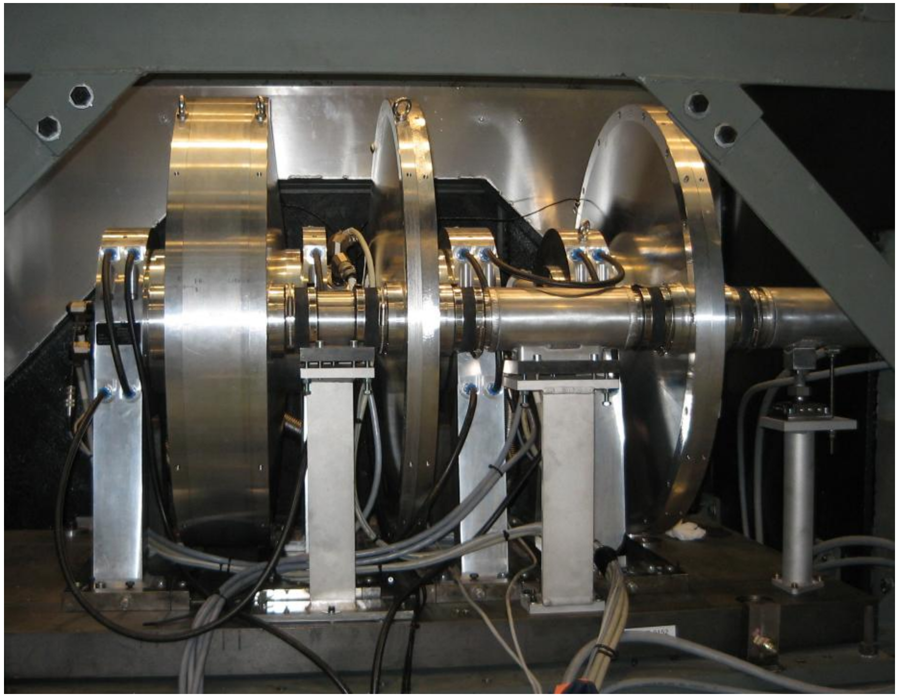
D17 (ILL): [2, 27] = 13.5
Platypus (ANSTO): [2.5, 19] = 7.6
INTER (ISIS): [1.5, 16] = 10.7





Time-of-flight at a reactor

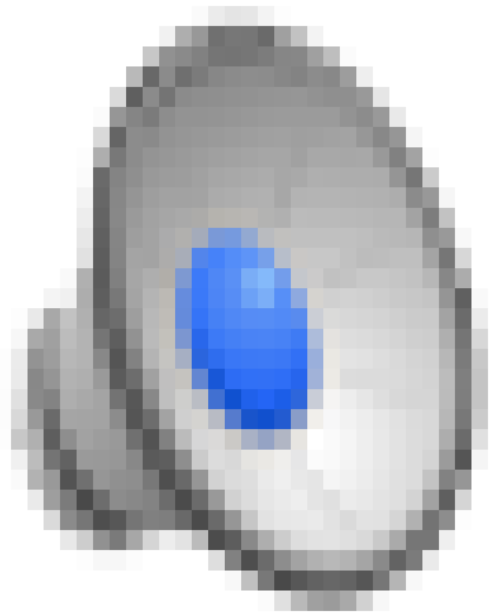




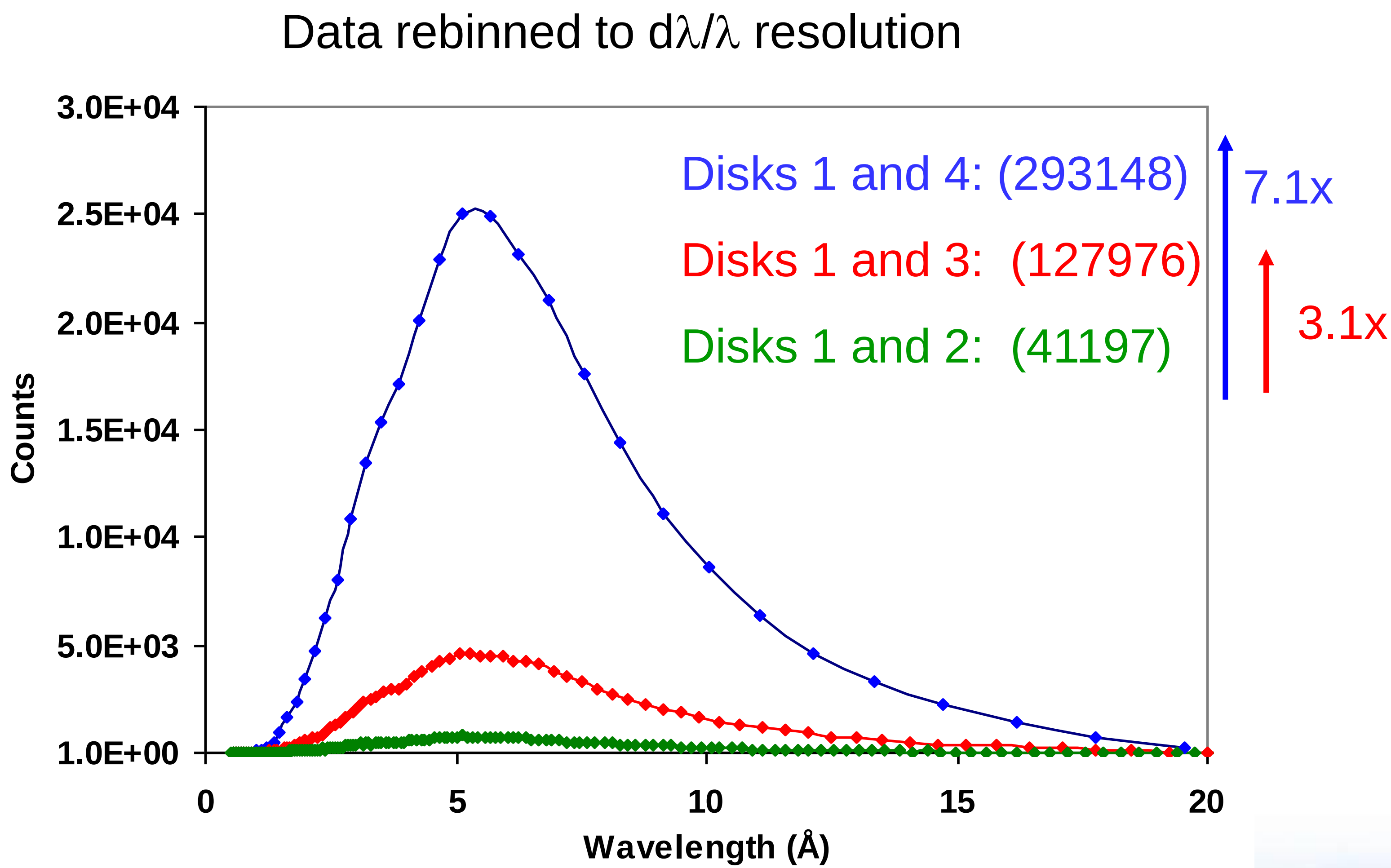
discs 1 & 2: $\Delta\lambda/\lambda$ ~1.1%

discs 1 & 3: $\Delta\lambda/\lambda$ ~3.3%

discs 1 & 4: $\Delta\lambda/\lambda$ ~7.7%



Incident Neutron Spectrum

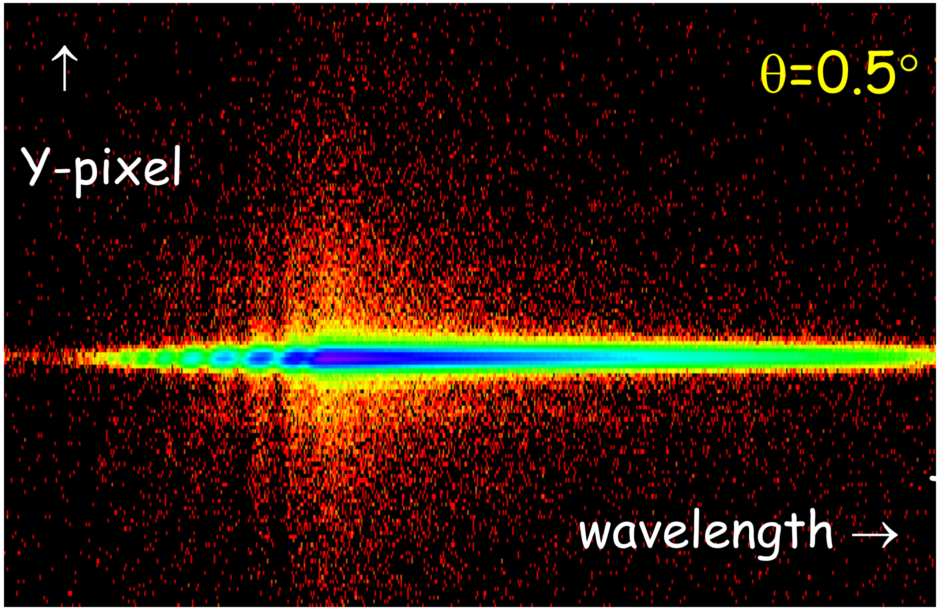


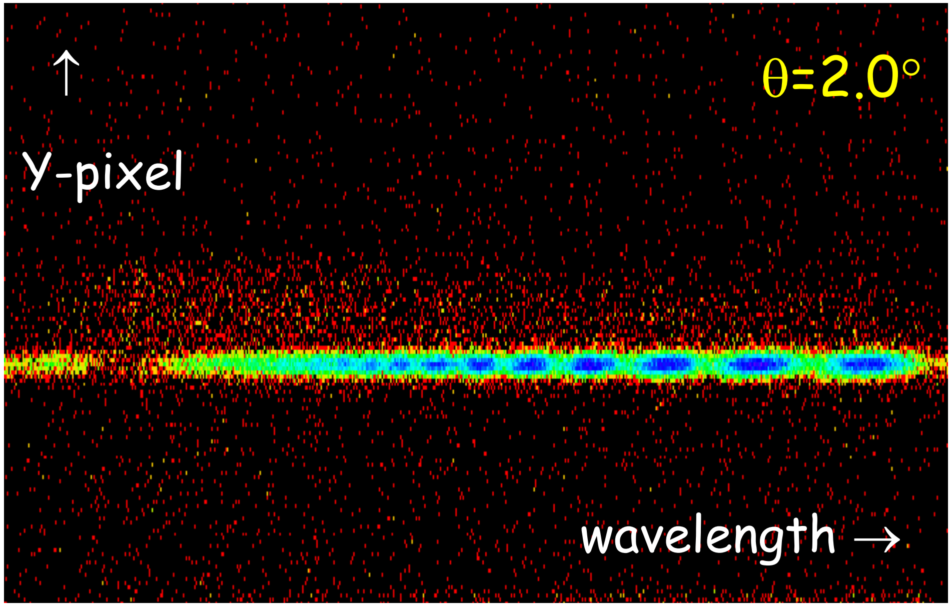
- $I \propto z_0 f \lambda$

To avoid frame overlap: $f_{max} = \dfrac{h}{mL(\lambda_{max} - \lambda_{min})}$

van Well, A., et al., *Physica B* **357** (2005) 204–207

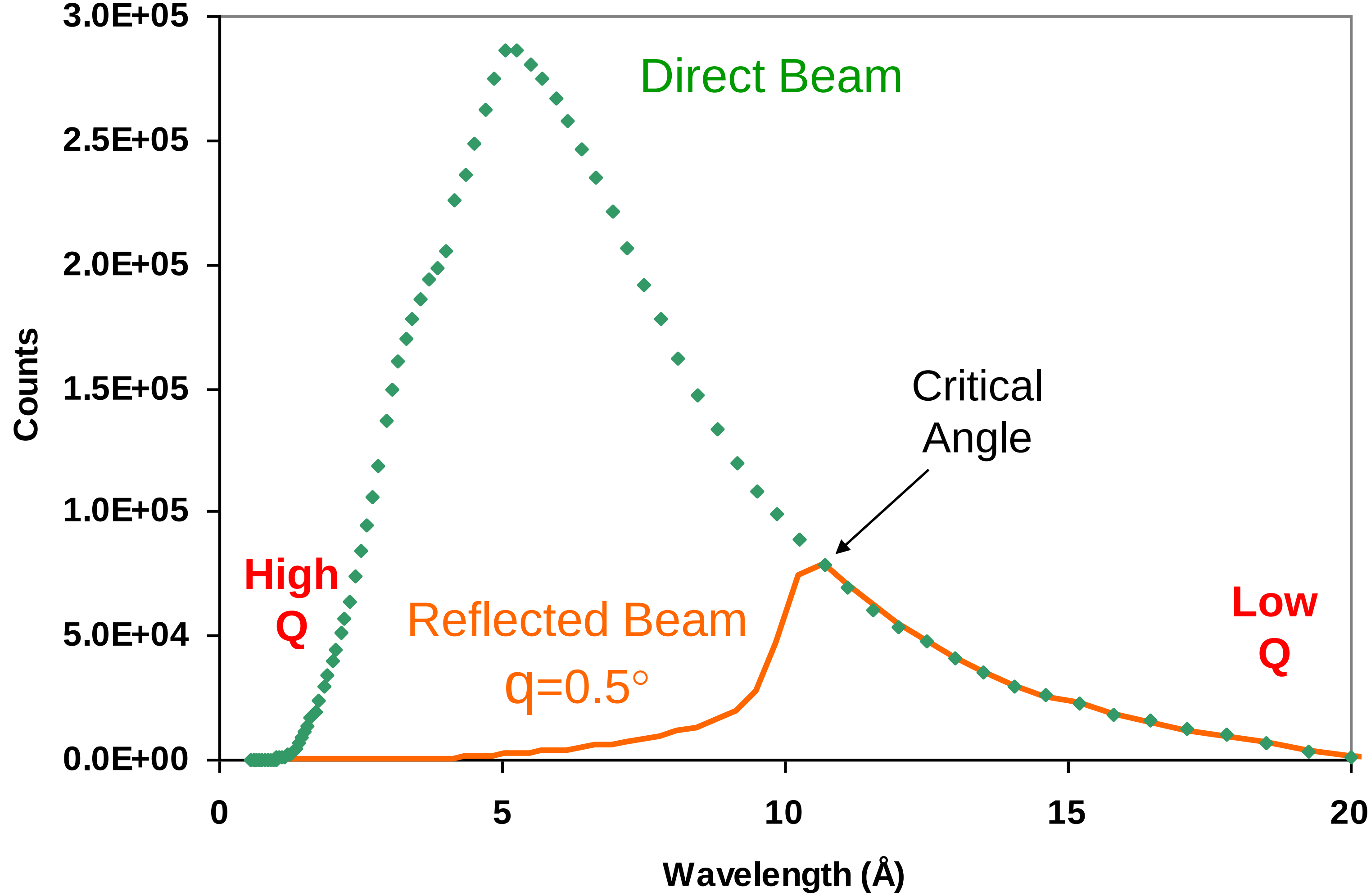
Spectrum



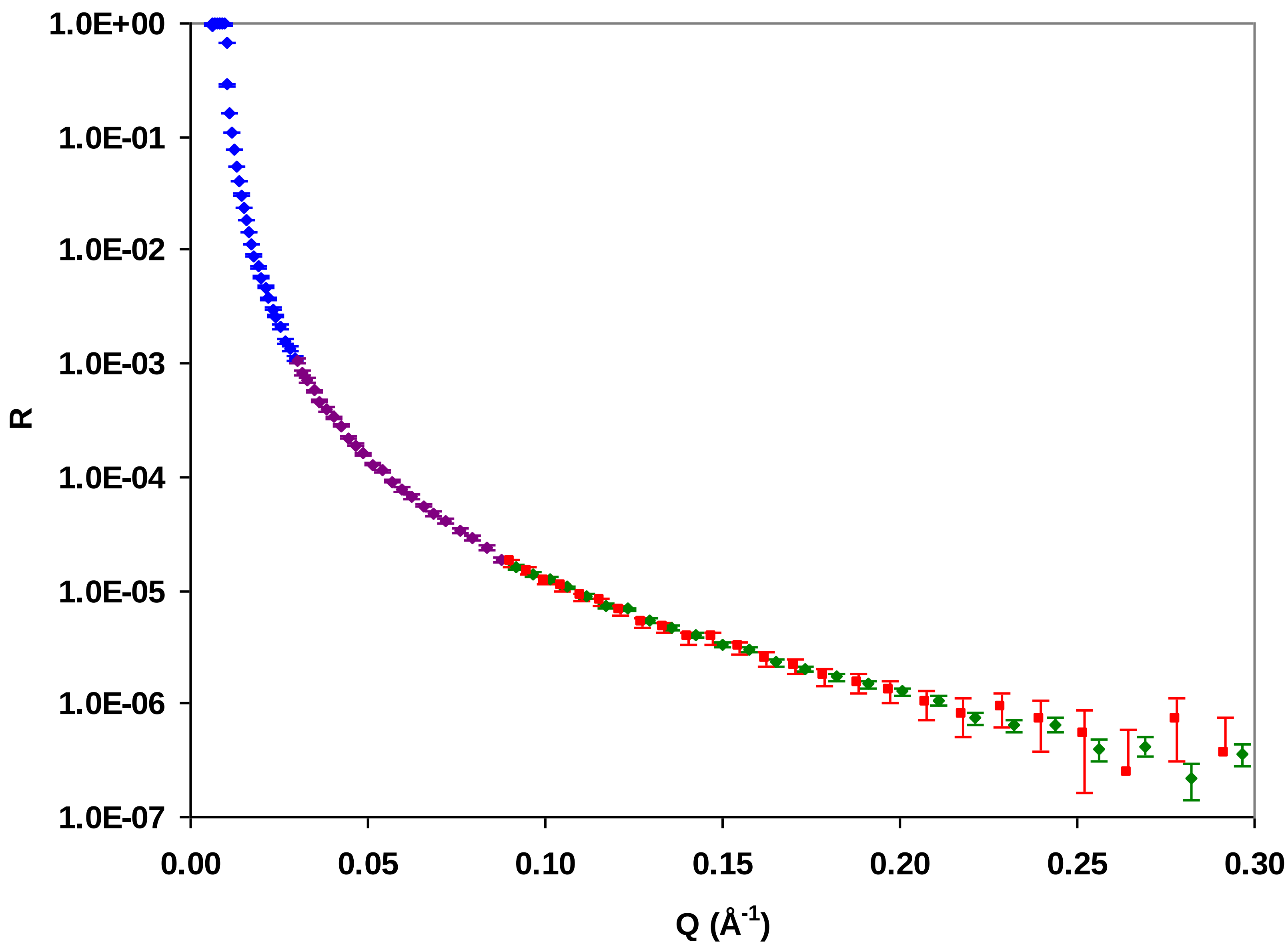


First Data: Silicon Wafer

Platypus



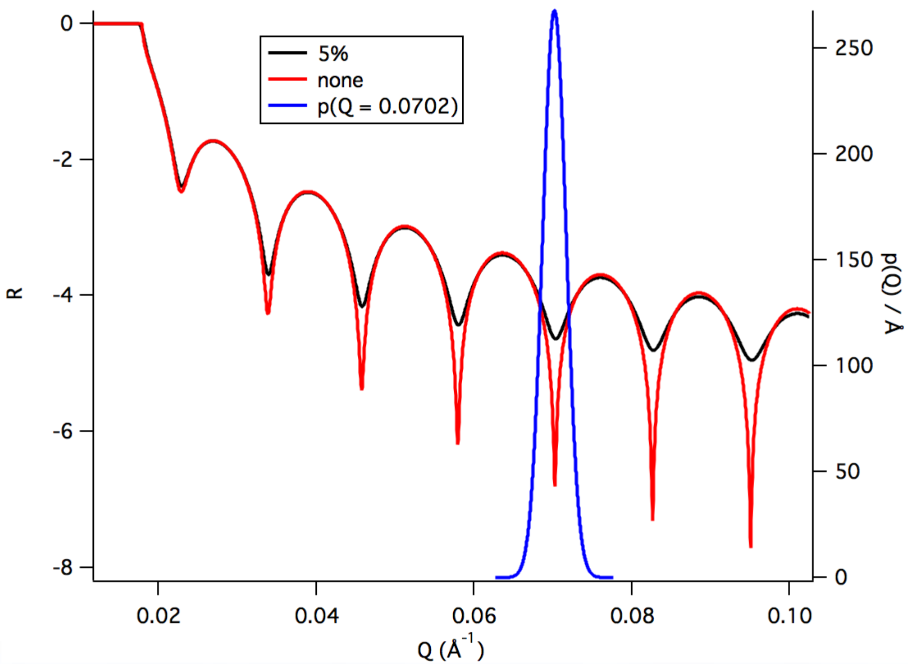
First Data: Silicon Wafer



Instrumental Resolution

$$R(Q_{z,0}) = \int_0^\infty dQ_z \, p(Q_z, Q_{z,0}) \, R(Q_z)$$

Smeared model reflectivity   Instrument resolution function   model reflectivity



$$\left(\frac{dQ_z}{Q_z}\right)^2 = \left(\frac{d\lambda}{\lambda}\right)^2 + \left(\frac{d\Omega}{\Omega}\right)^2$$

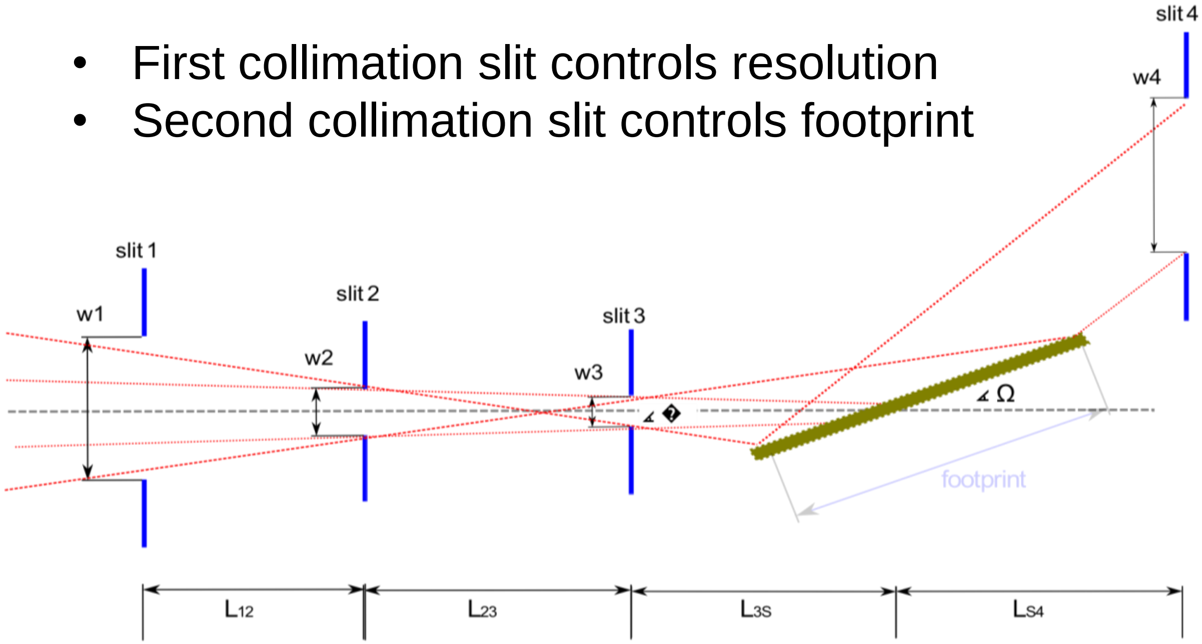
- Thin films (< 200 Å) require low resolution
  dQ/Q ~ 8%
- Thick films (> 1000 Å) and multilayer stacks require high resolution
  dQ/Q ~ 2%
- Split angular + wavelength equally
- Much harder to tune angular dispersive resolution

- Beam intensity ∝ resolution

van Well, A. A. & Fredrikze, H., *Physica B-Cond. Mat.*, **2005**, *357*, 204

Angular resolution considerations

- First collimation slit controls resolution
- Second collimation slit controls footprint



Typical Ω = 0.6 deg

\+

d Ω/ Ω = 3%

\+

Footprint = 25 mm

→

w2 = 1.5 mm

w3 = 0.15 mm

Goniometer requirements

- ~0.01 mm height precision
- 0.001 degrees in tilt

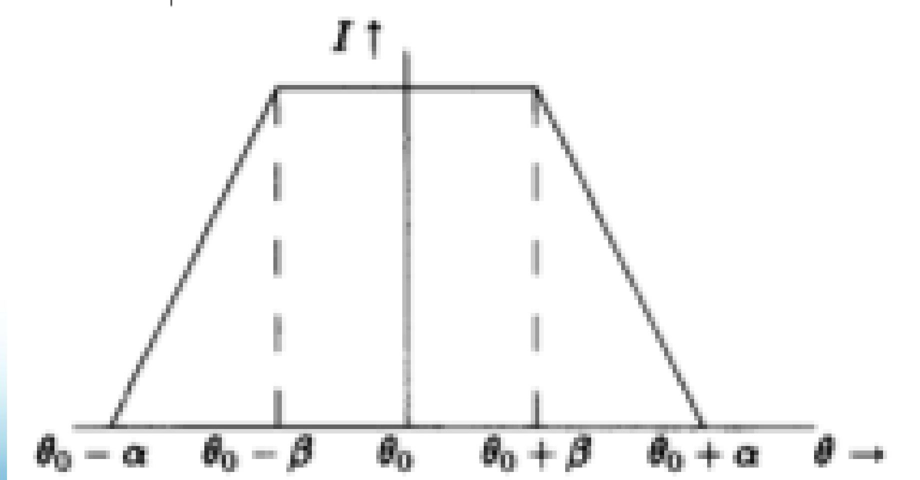


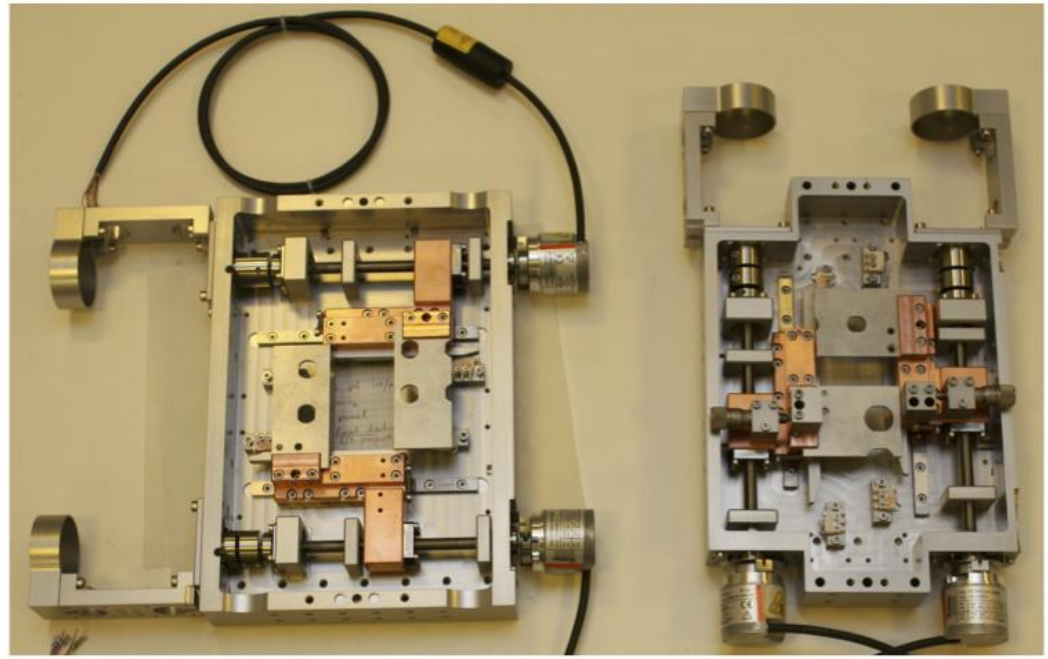
Fig. 6. Neutron intensity as a function of incident angle: $\alpha = (d_1 + d_2)/2L_{12}$ and $\beta = |(d_1 - d_2)/2L_{12}|$.

http://refcalc.appspot.com/slits

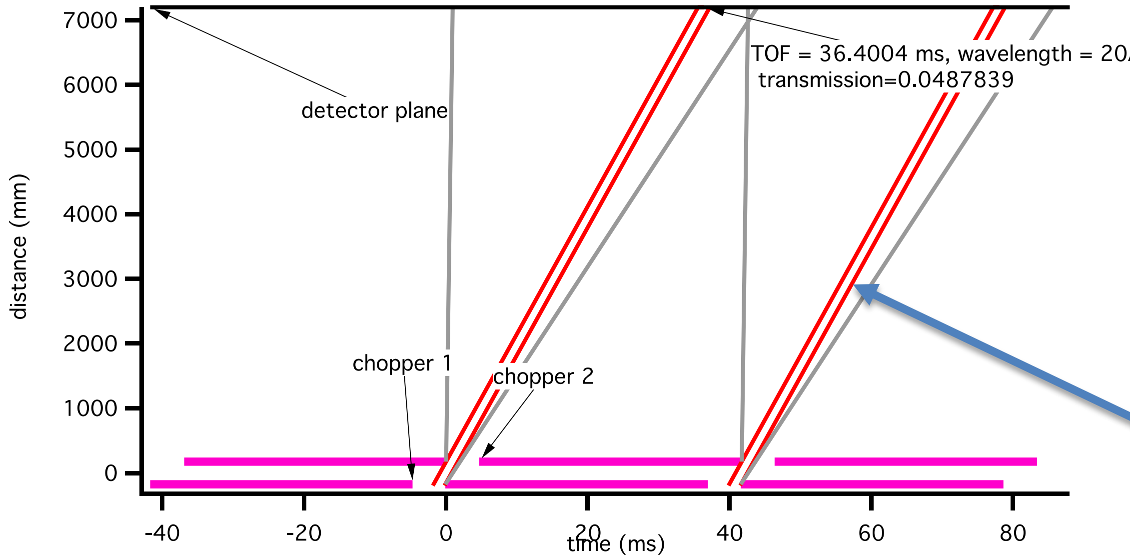
de Haan et al. *Nucl. Instr. Meth. A.*, **362**, 434

Collimation slit requirements

- Micron reproducibility
- Micron accuracy
- Optical encoding (w. tape)
- Absolute encoding
- Ball screws
- Low magnetic signature
- Hot pressed/sintered B_4C
- Chamfered edge (low albedo)



Wavelength resolution - TOF



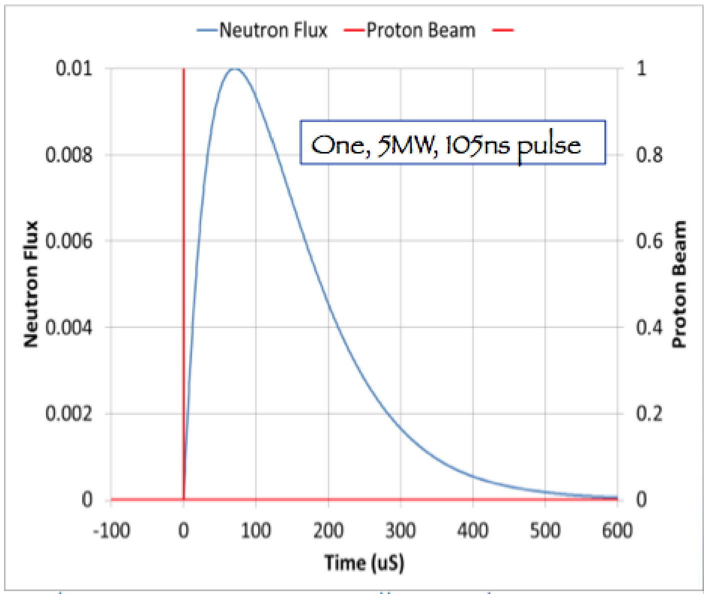
Continuous source w. choppers

- **Pulse is rectangular**

$$\frac{\Delta\lambda}{\lambda} = \frac{\Delta d}{d} = \frac{\Delta t}{t}$$

d = flight length
Δd = distance between choppers
Δt = "burst time" of pulse
t = time-of-flight

602 μs for 10Å neutrons at 3% res.



Spallation source

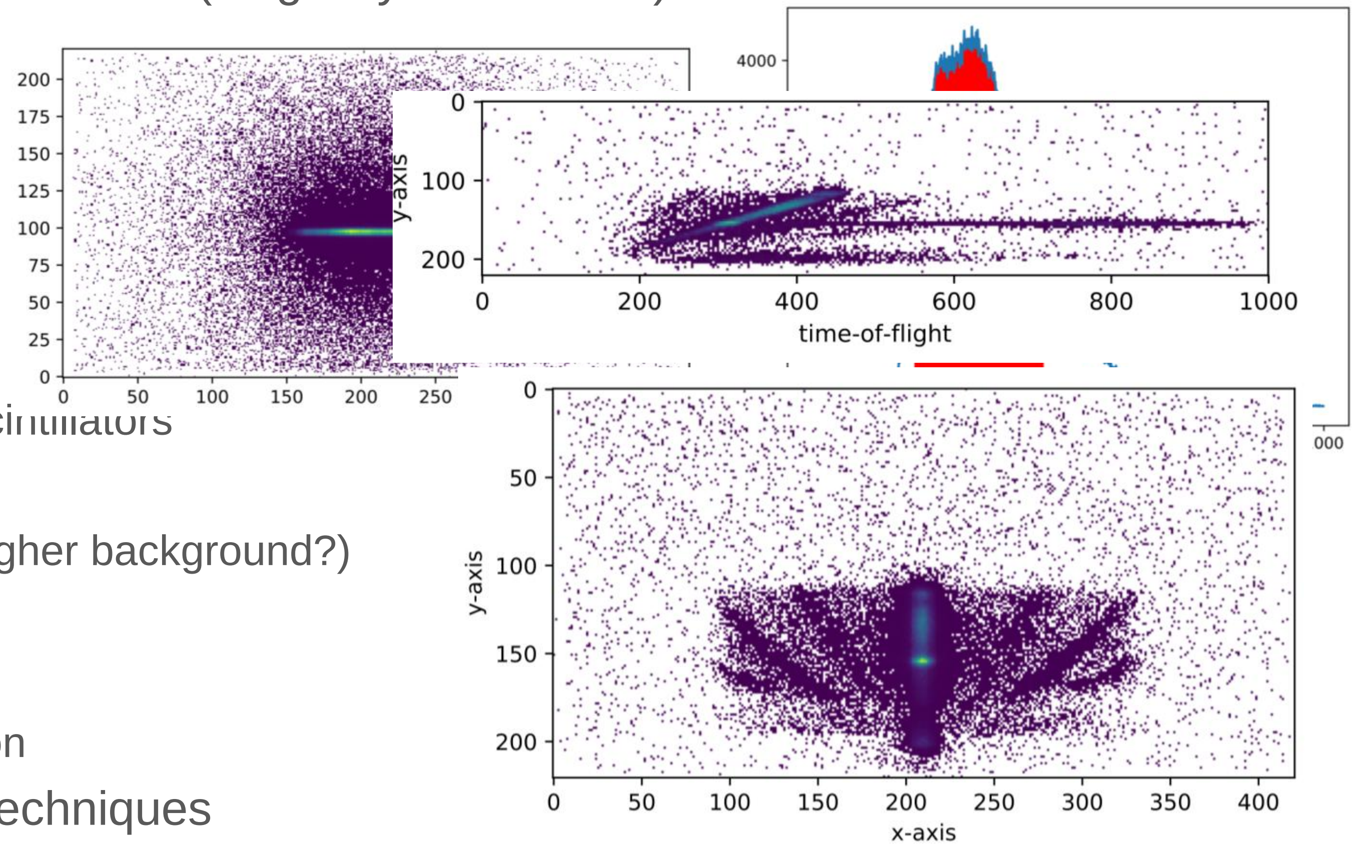
- FWHM of neutron pulse:
  - $22\lambda(\text{Å})\mu s$ (thermalised)
  - depends on moderator
- Pulse is fixed length (in general),
- Resolution dictated by instrument length

~220 μs for 10Å neutrons

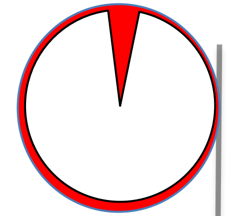
Andersen, K; Carlile, C., Journal of Physics: Conference Series **746** (2016) 012030

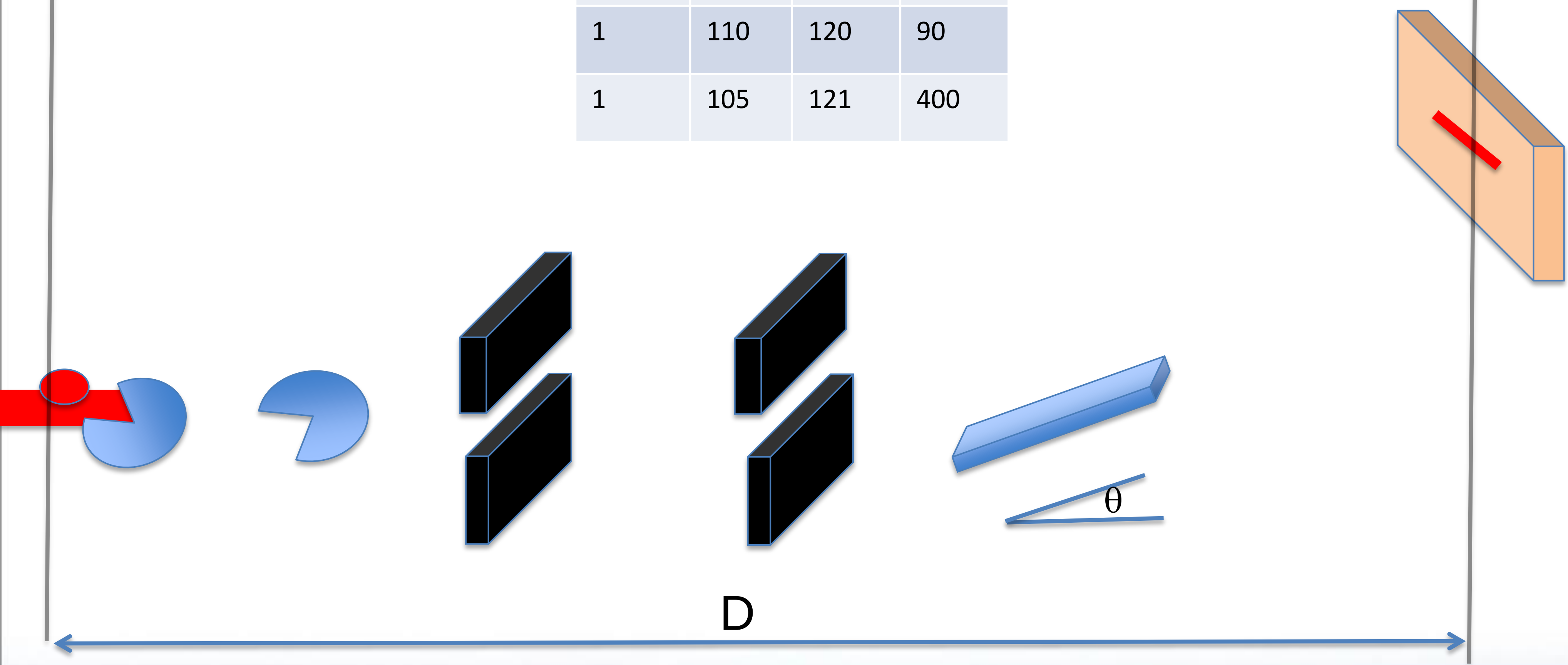
Detector considerations

- Detector copes with high count rates (longevity + deadtime):
  - Globally
  - Locally
  - Instantaneous
- Efficiency
  - 3He Gas pressure
  - Scintillators
  - Future: 10B / advanced scintillators
- 1D vs 2D
  - Offspecular + GISANS (higher background?)
  - Background subtraction
- Resolution:
  - better than 2 mm resolution
- Advanced data acquisition techniques
  - Event mode for neutrons / sample environment
  - Stroboscopic



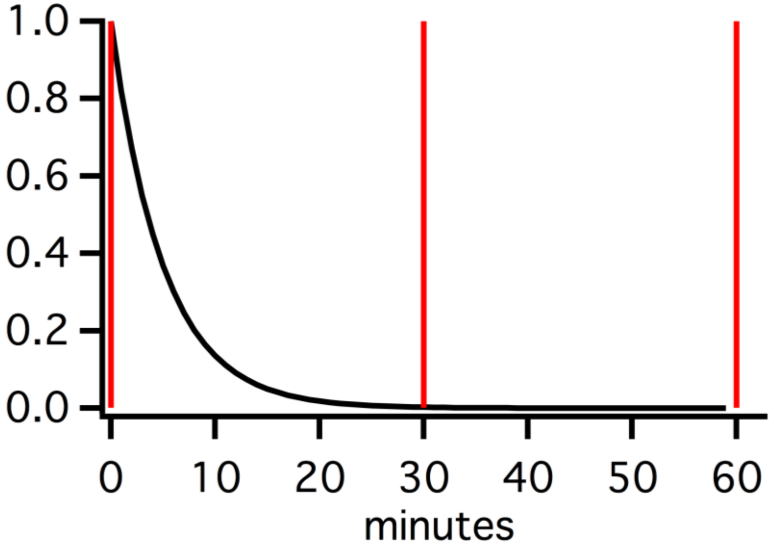
Frame	x	y	time
0	100	122	100
0	101	123	120
1	110	120	90
1	105	121	400



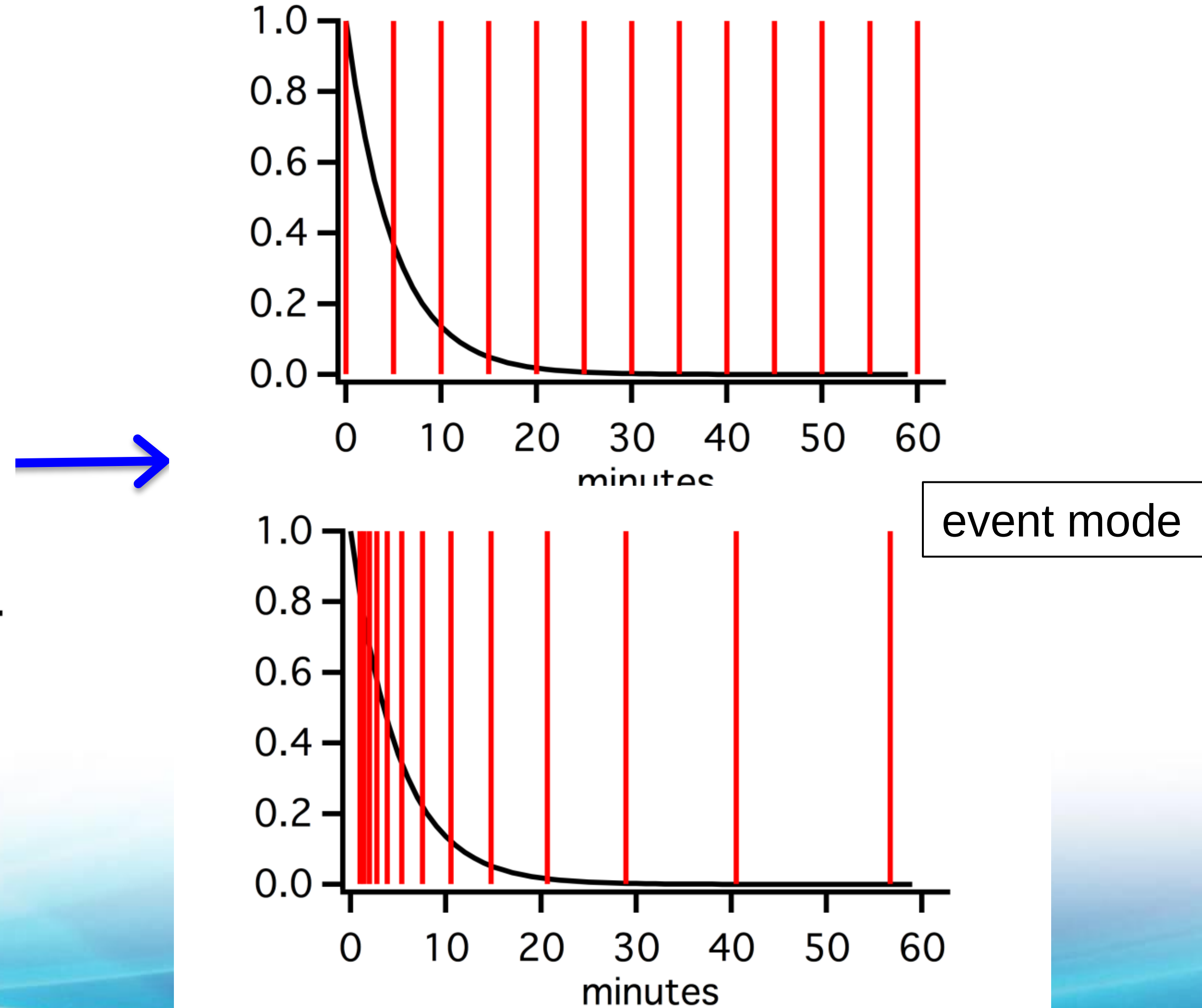


$$l = \frac{h}{p} = \frac{h}{mv} = \frac{ht}{mD}$$

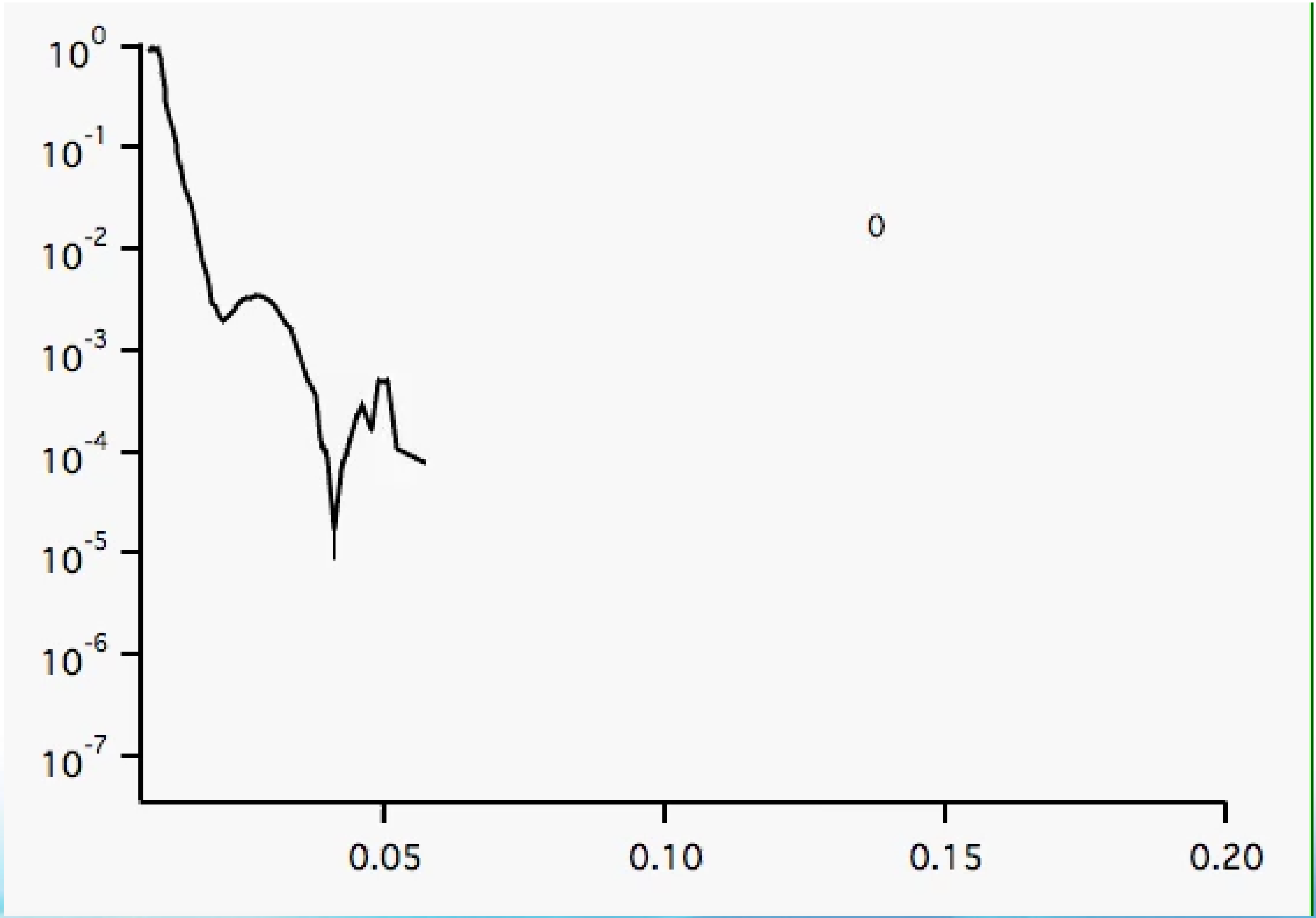
Dealing with the kinetics is easier afterwards



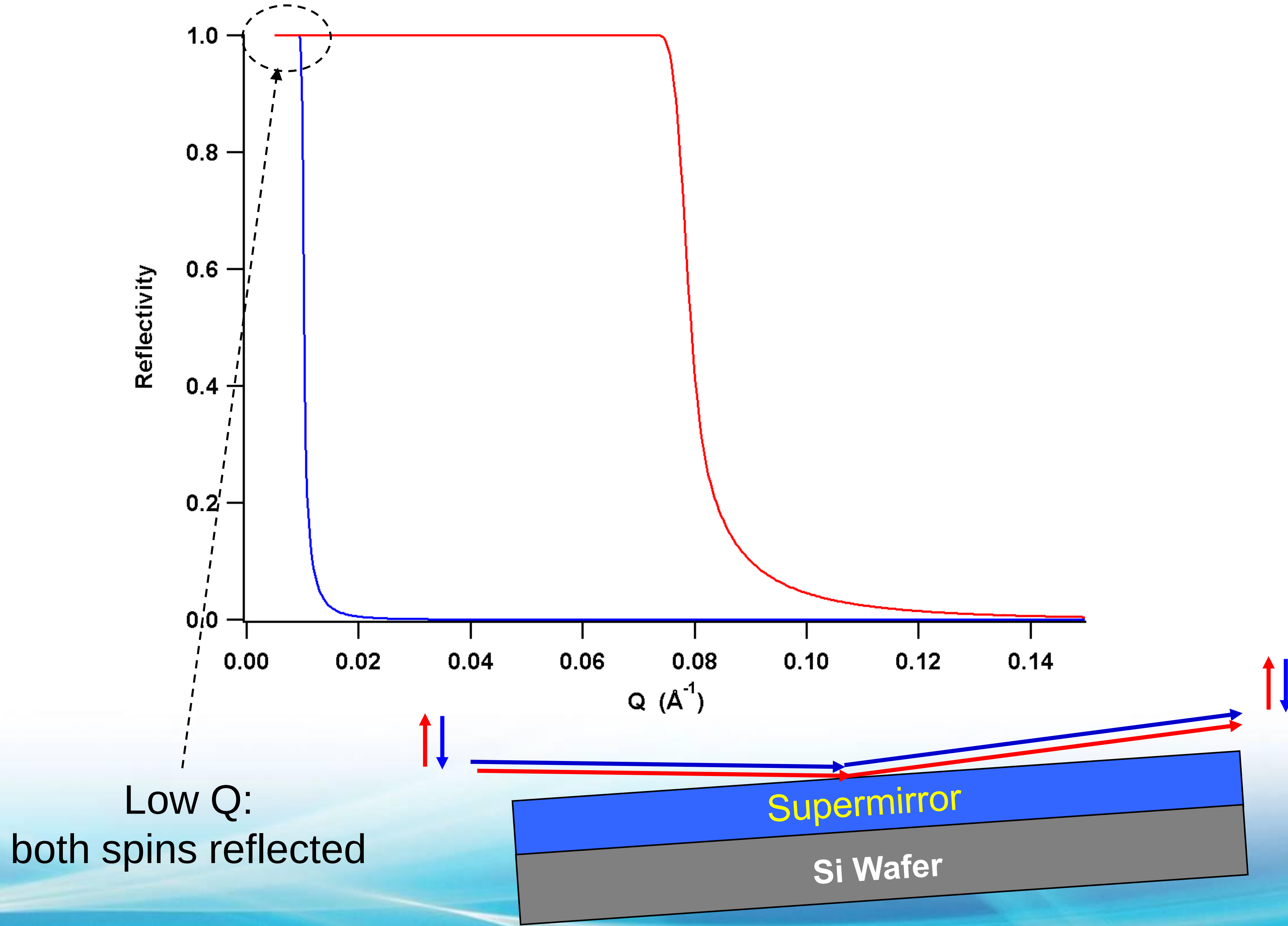
Normal acquisition



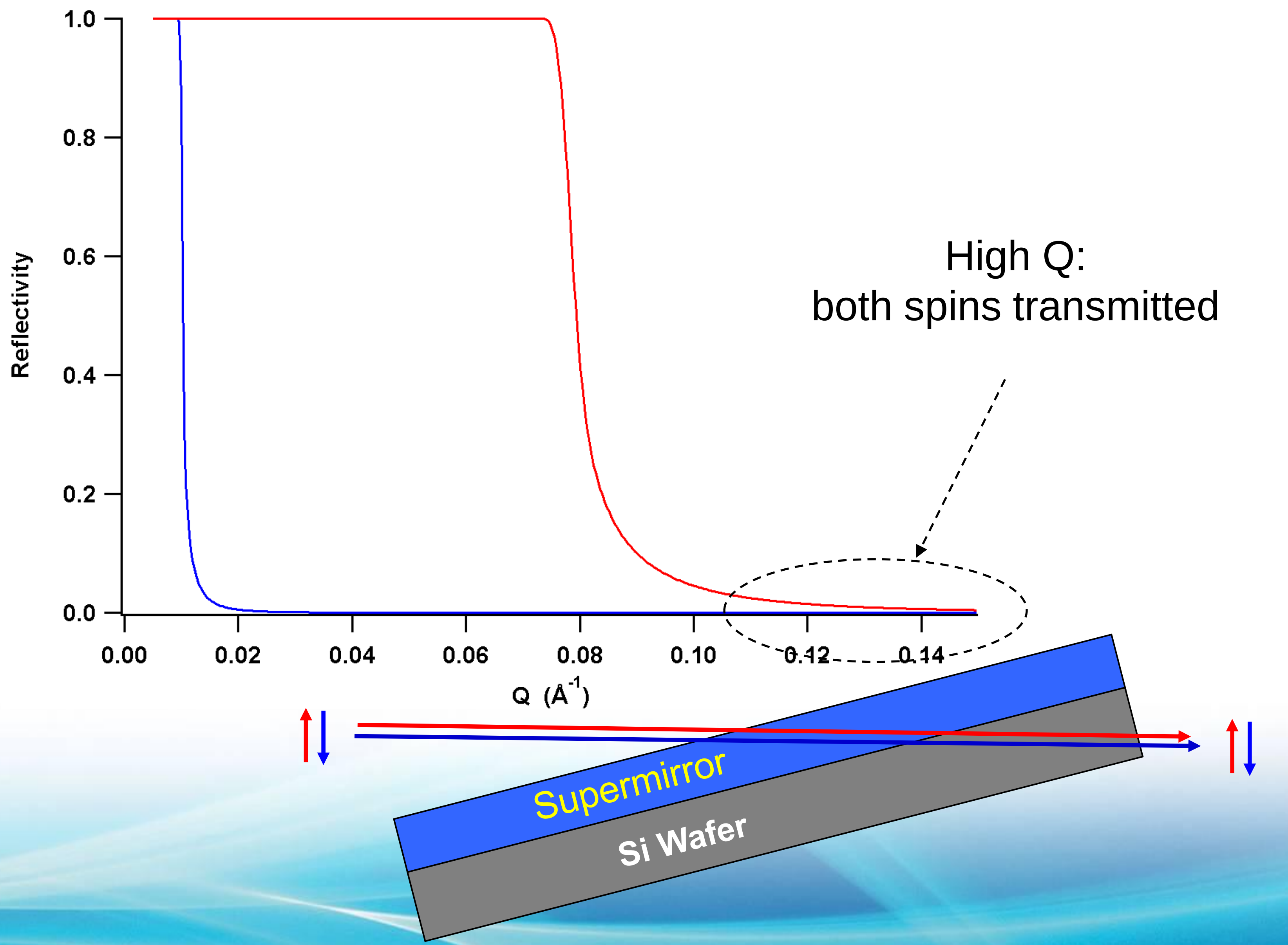
Event mode – kinetic enabler



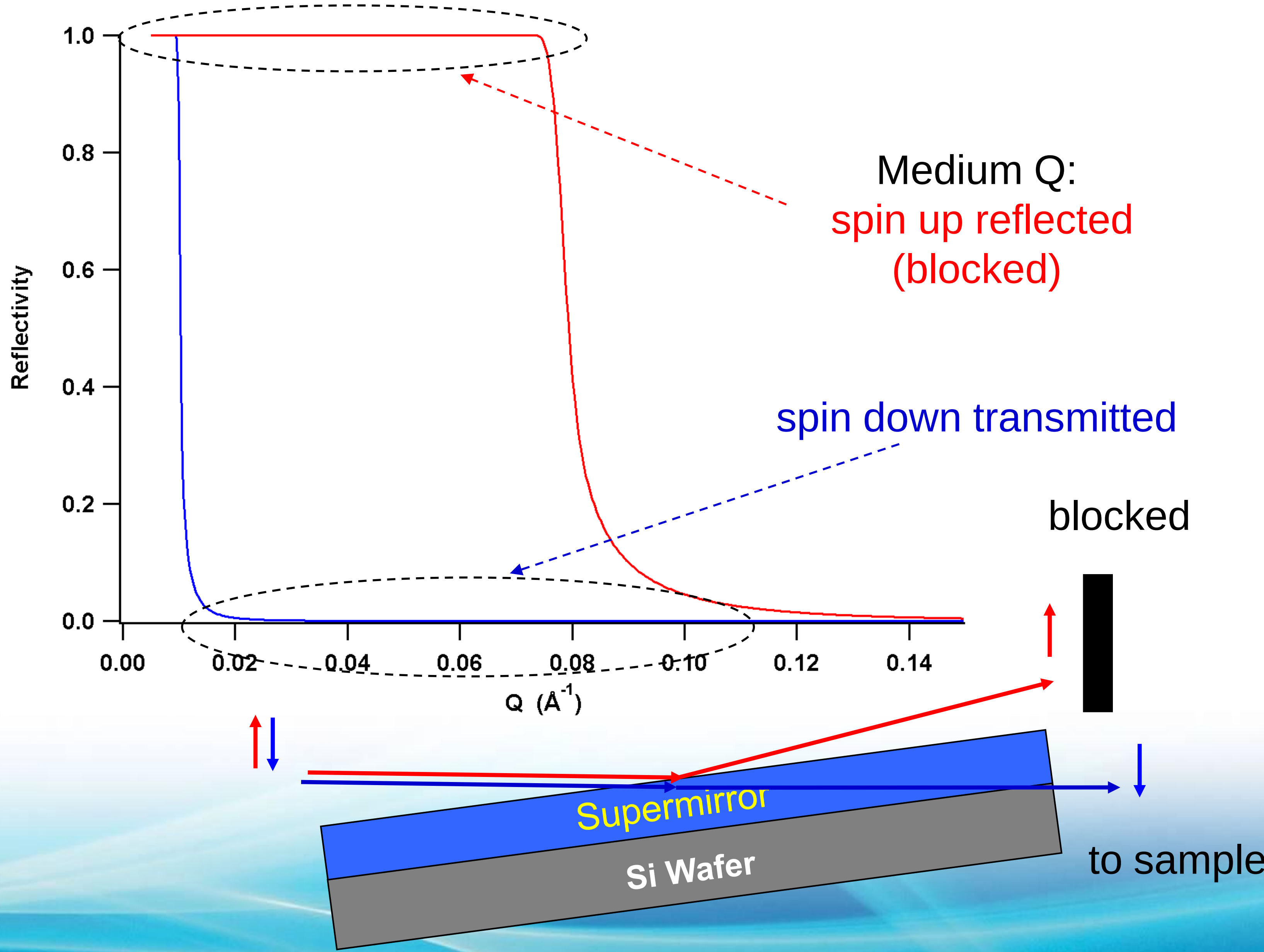
Generating Polarized Neutrons



Generating Polarized Neutrons

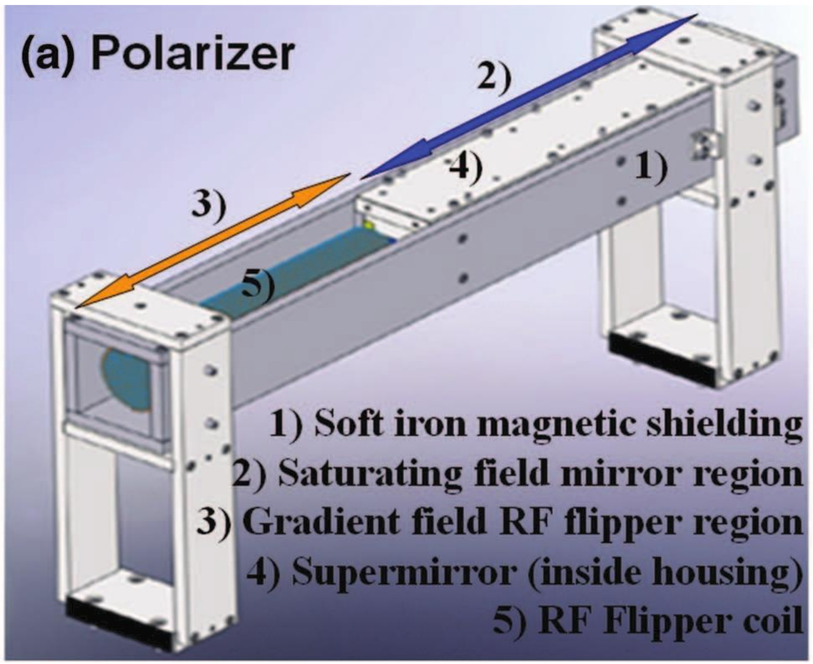


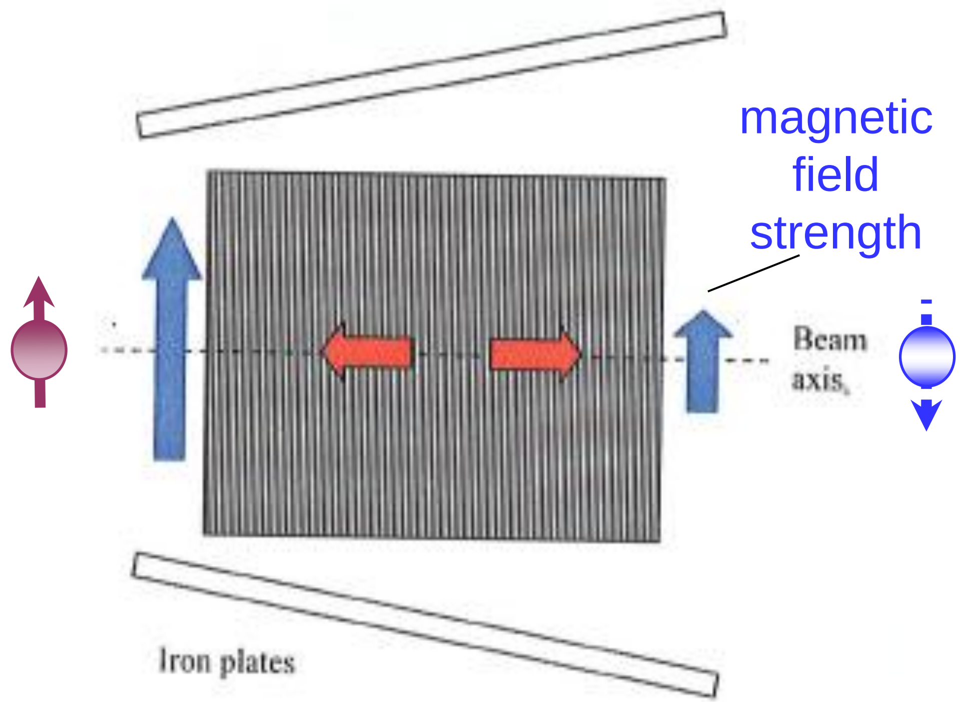
Generating Polarized Neutrons



RF Spin Flippers

Used to flip the spin state of variable wavelength neutrons





Induces Larmor precession similar to that used for NMR

Diverging iron plates create a field gradient

Rotating field produced by a RF signal in coils around the beam axis

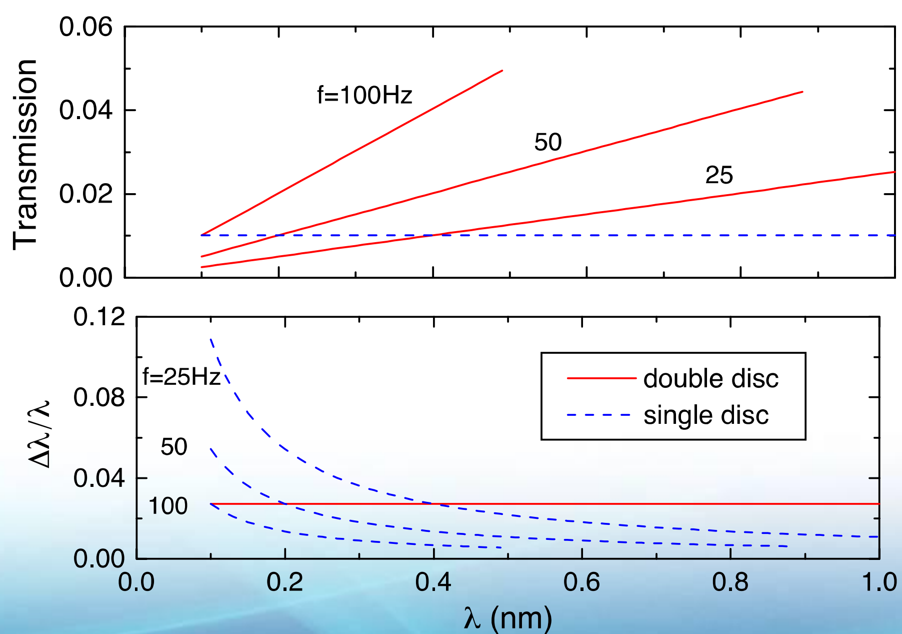
Chopped TOF at Continuous Sources

Pros

- **Good for kinetic processes** (large dynamic Q range)
  - Stroboscopic
  - Single shot
- Only 2-3 angles required
- Easy to under-illuminate (larger angles)
- Easier liquid interfaces
- Constant dQ/Q
- Vary resolution
- Area detector
  - simultaneous background measurement
  - Offspecular
  - GISANS
  - Hi-res = act as a slit

Cons

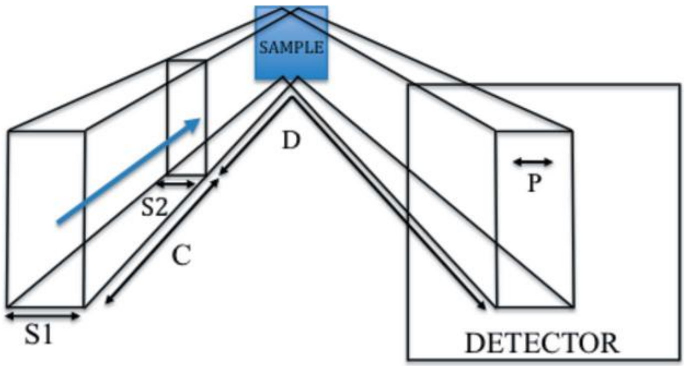
- Transmission typically <10%
- A little bit harder to operate
- Gravity effects (vertical scattering plane)
- End guide position
- Polarisation more difficult



from: van Well

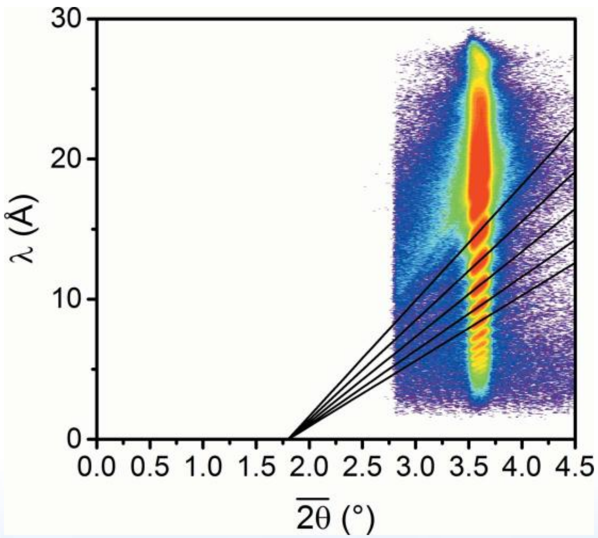
- de Haan, V., et al., *Nucl. Inst. Meth. A.* ***362*** (1995) 434-453
- van Well, A., et al., *Physica B* **357** (2005) 204–207
- James, M., et al., *Nucl. Inst. Meth. A.* **632** (2011) 112-123
- Campbell, R., et al., *Eur. Phys. J. Plus* **126** (2011) 107

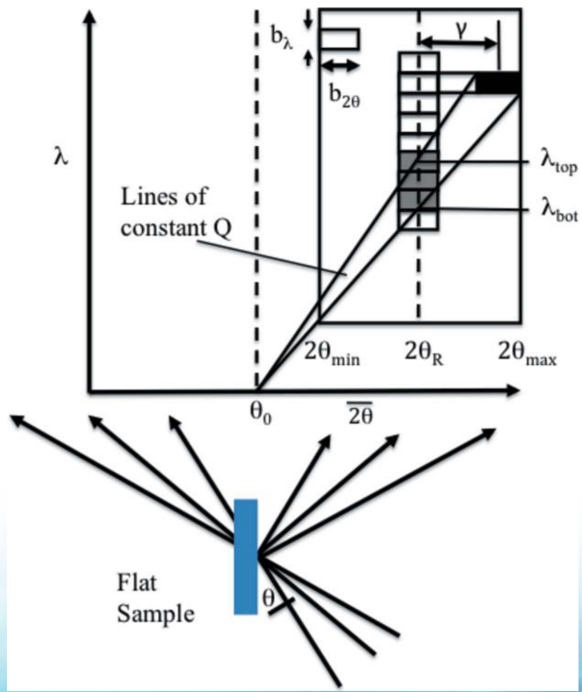
NR using divergent beams/non-flat samples



$$(\delta\alpha)^2 = (s1/C)^2 + (s2/C)^2$$

$$(\delta\theta)^2 = \begin{cases} (\delta\alpha)^2 & \text{if } (\delta\alpha)^2 < (s2/D)^2 + (P/D)^2 \\ (s2/D)^2 + (P/D)^2 & \text{if } (\delta\alpha)^2 > (s2/D)^2 + (P/D)^2 \end{cases}$$



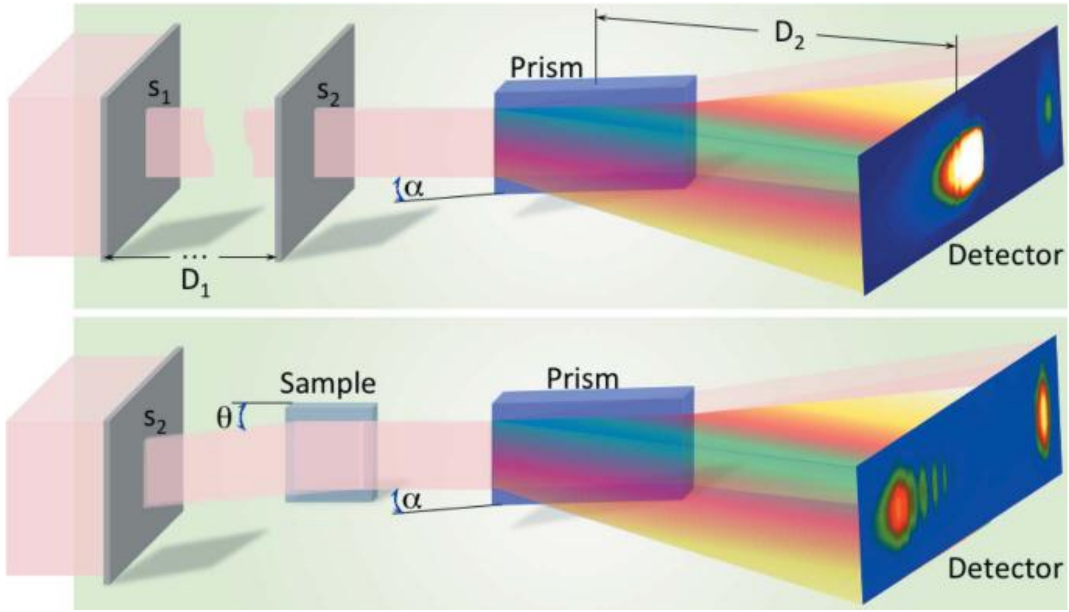


- Improve resolution if incoming divergence > angular resolution of detector
- $I \propto s1{\times}s2$, use relaxed collimation without resolution penalty
- non-flat samples can be measured

Cubitt, R., et al. *J. Appl. Cryst*. **48** (2015), 2006-2011

RAINBOWS – refractive analysis of reflected beam

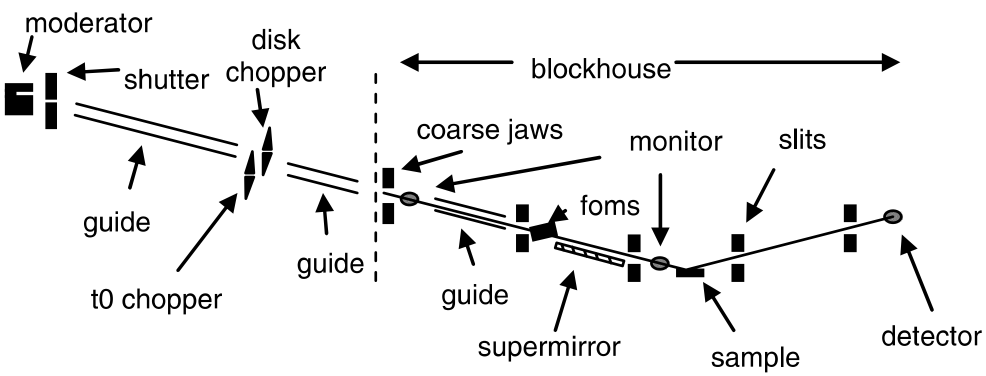
- TOF at continuous sources have low transmission, ~ 0.02
- Monochromatic have high transmission, but only use single wavelength (not suited to kinetic processes)

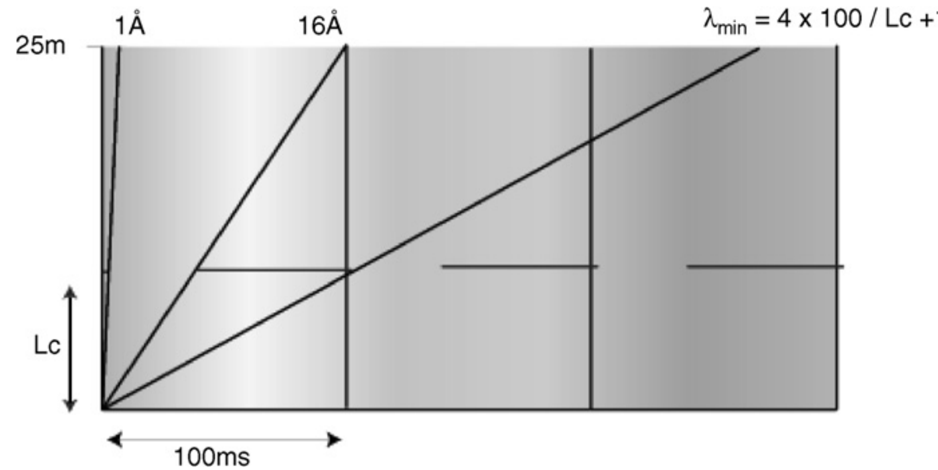


- MgF_2 refracts reflected beam
- Refraction angle is wavelength dependent
- High resolution detector is required
- Potential for large gains

Cubitt, R., et al., *J. Appl.Cryst.* **51** (2018) 257-263

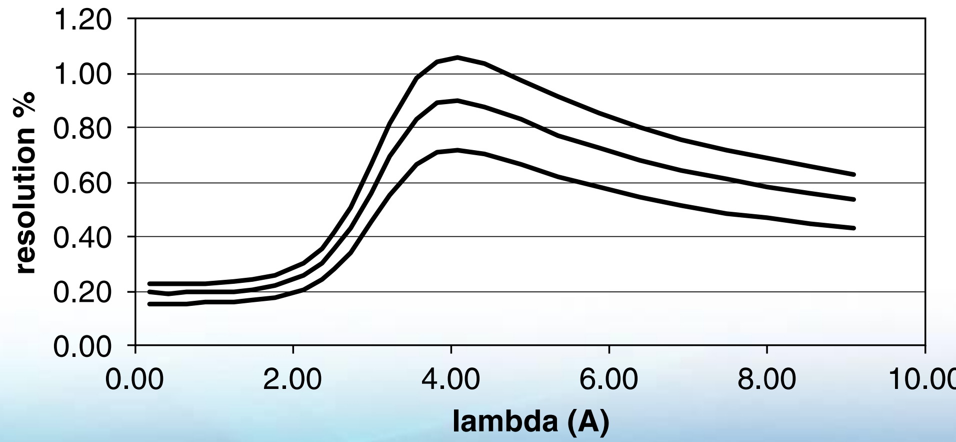
Spallation reflectometry - INTER





$$\Delta\lambda = \frac{h}{m_n L f}$$

- Moderator design important (brilliance)
- T_0 stops the prompt pulse of fast neutrons (some instruments use bender to reduce background, poss. limits λ_{min})
- Disc choppers control wavelength band
- 10 Hz
- L = 25m
- Inclined at 2.3 degrees
- Wavelength range [1.5, 16]
- High pressure detector (12-15 bar ^{3}He)



Webster, J., et al. *Physica B* **385–386** (2006) 1164–1166

Spallation reflectometry

Pros

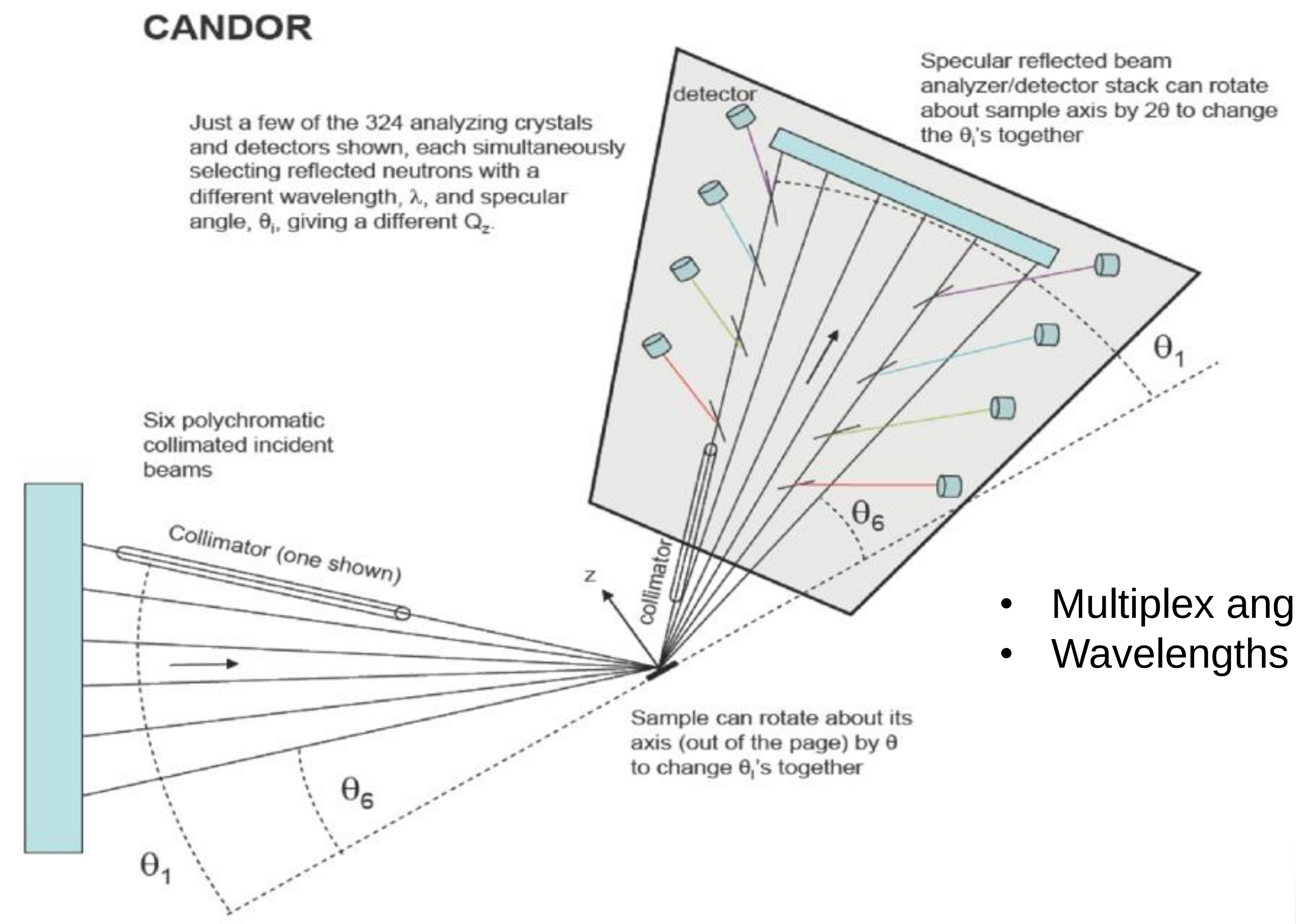
- **Good for kinetic processes** (large dynamic Q range)
  - Stroboscopic
  - Single shot
- Only 2-3 angles required (depends on bandwidth)
- Easy to under-illuminate (larger angles)
- Easy to measure free liquid interfaces

Cons

- wavelength resolution fixed by instrument length + pulse characteristics
- Bandwidth dictated by source frequency:
  - For a fast source
    f = 60 Hz, L=15 m → $\Delta\lambda = 4.4Å$
  - Might take 9 angles for full Q range
  - Can extend by frame skipping, if there's no problem with contamination from missed pulses

- CRISP/SURF/OFFSPEC/INTER/POLREF (ISIS)
- Liquid/Magnetism Reflectometers (SNS)
- SOFIA/SHARAKU (JPARC)
- Reflectometer (CSNS)
- SPEAR (LANSCE)

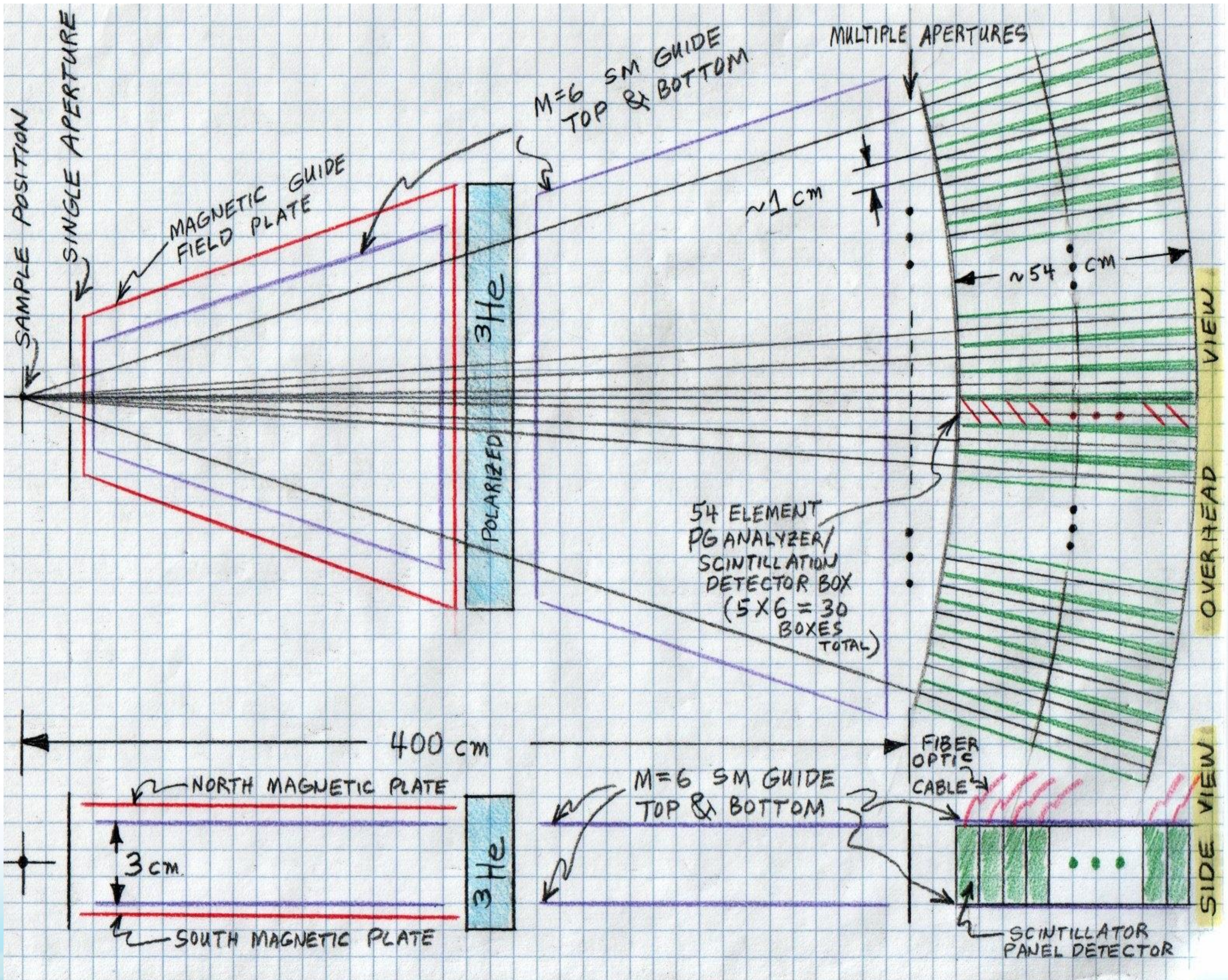
CANDOR



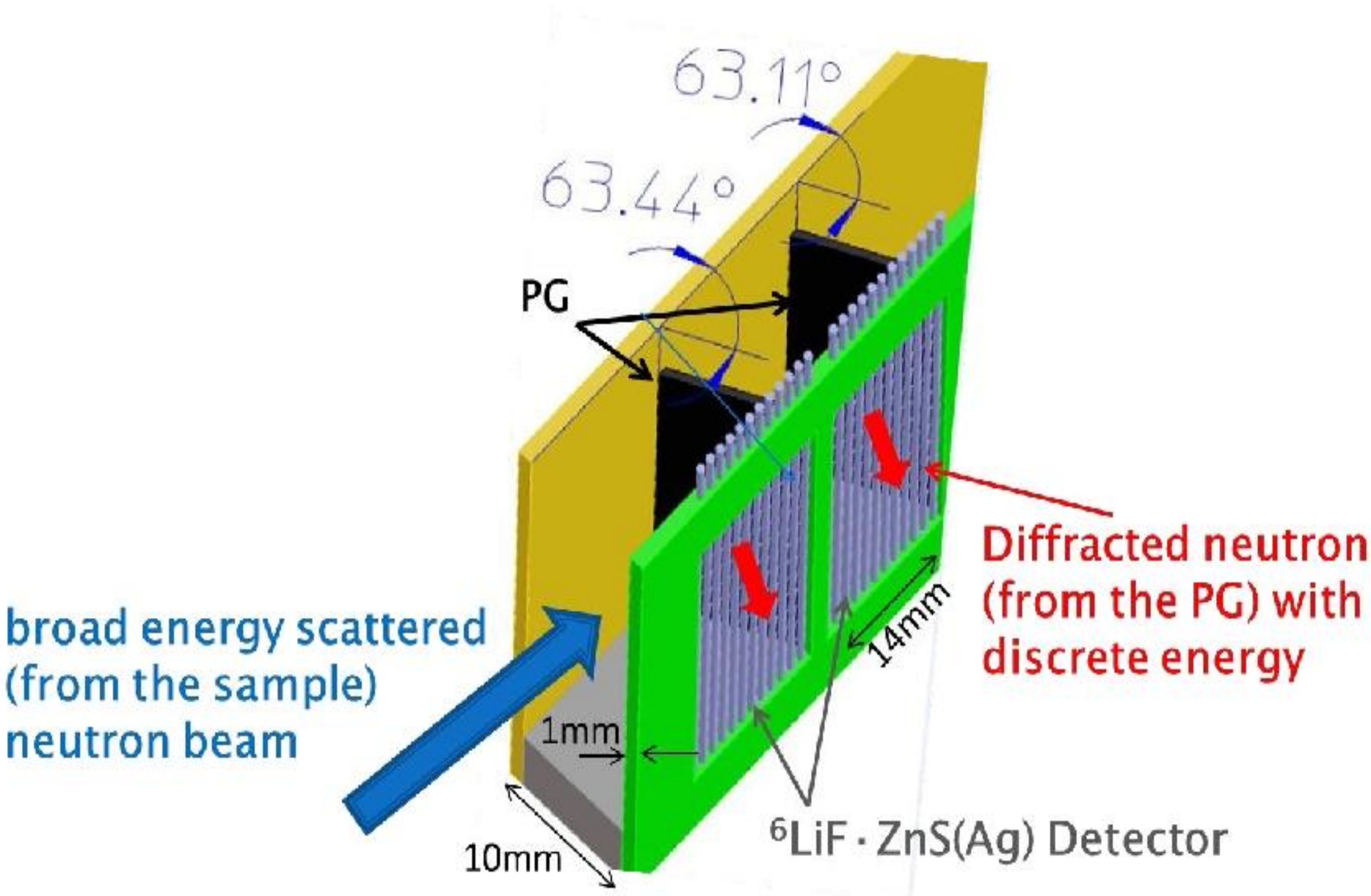
- Multiplex angles AND wavelengths
- Wavelengths [4, 6] Å

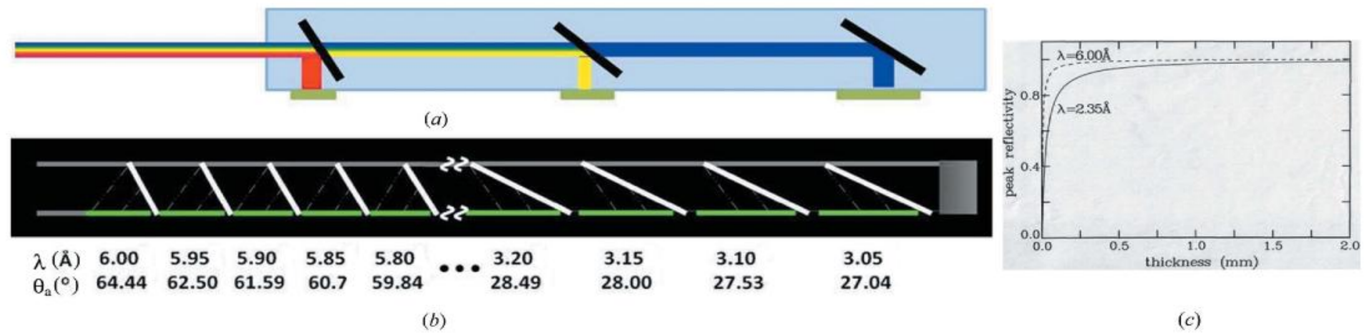
All CANDOR descriptions sourced from NIST website:
https://ncnr.nist.gov/equipment/msnew/ncnr/candor.html

Detection arm



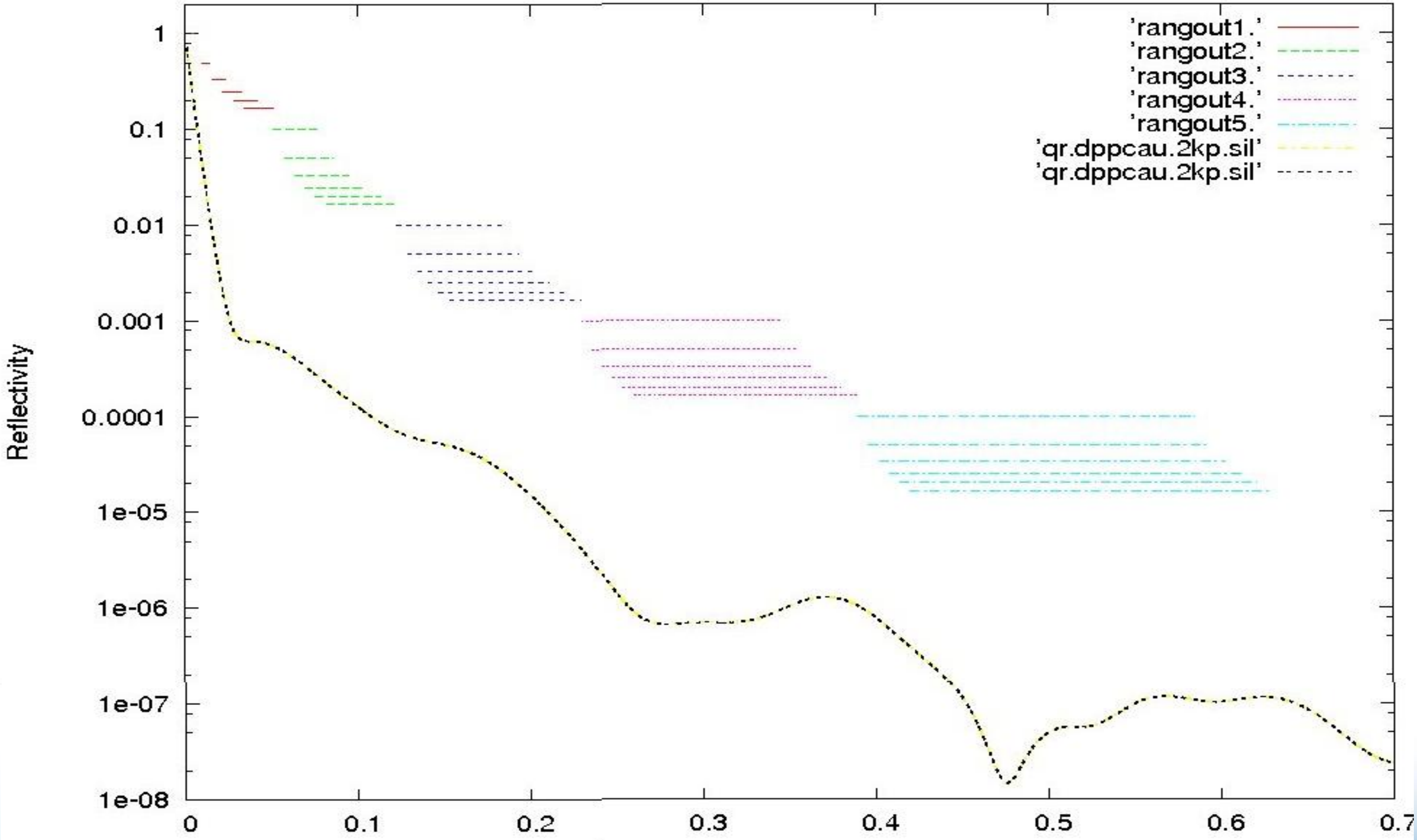
https://ncnr.nist.gov/equipment/msnew/ncnr/candor.html



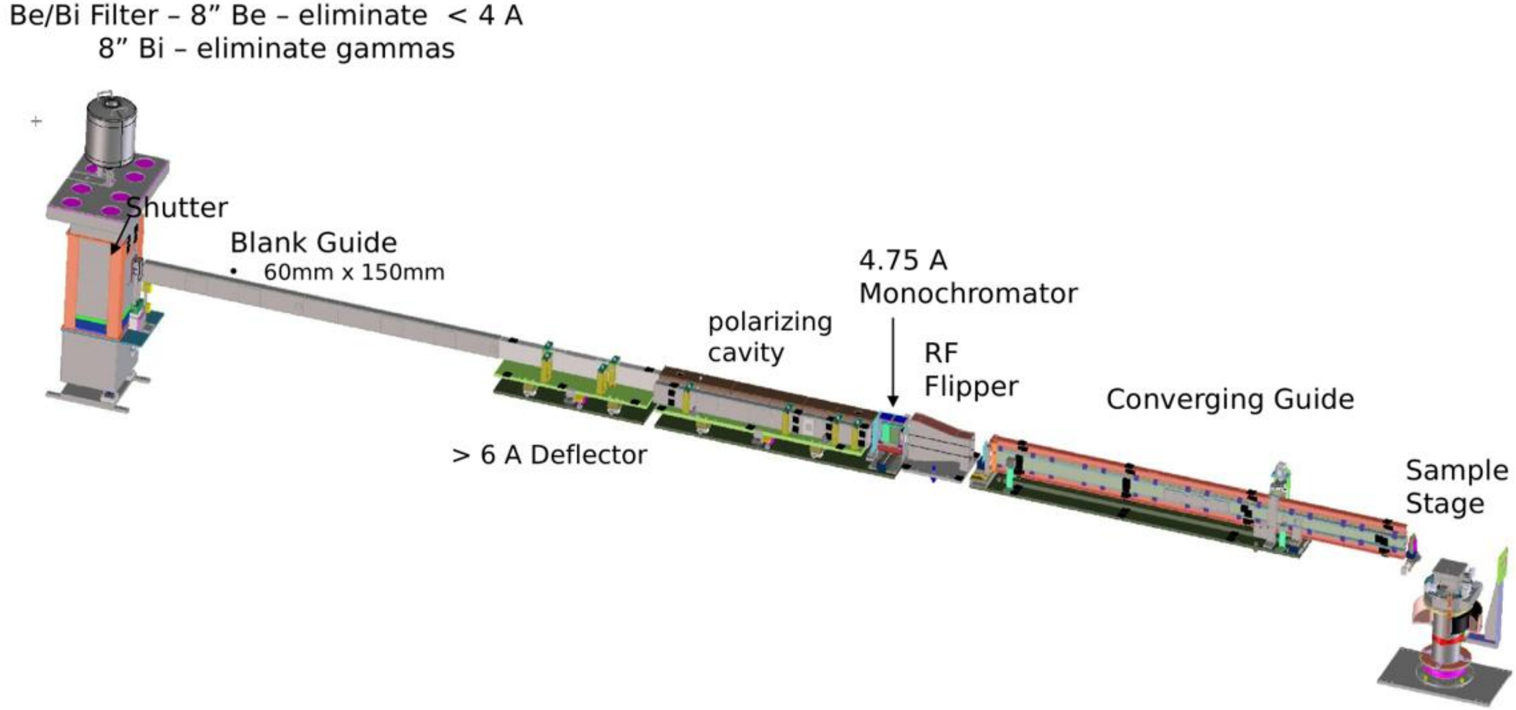


https://ncnr.nist.gov/equipment/msnew/ncnr/candor.html

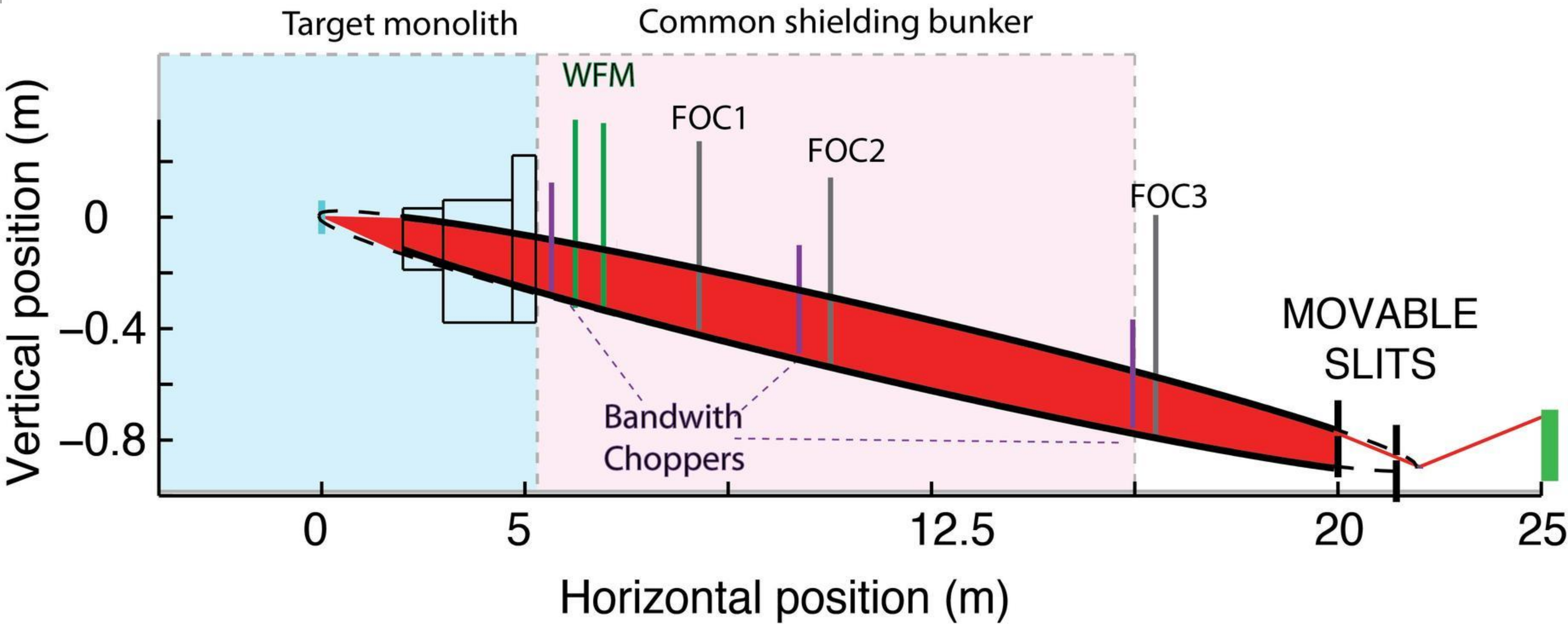
Q range covered by CANDOR



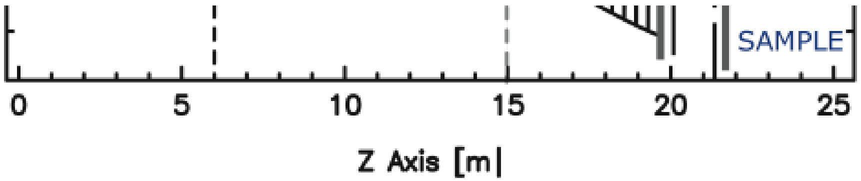
Incident beam optics

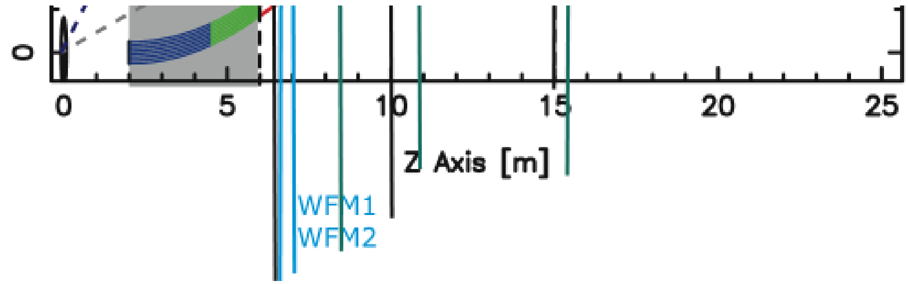


FREIA



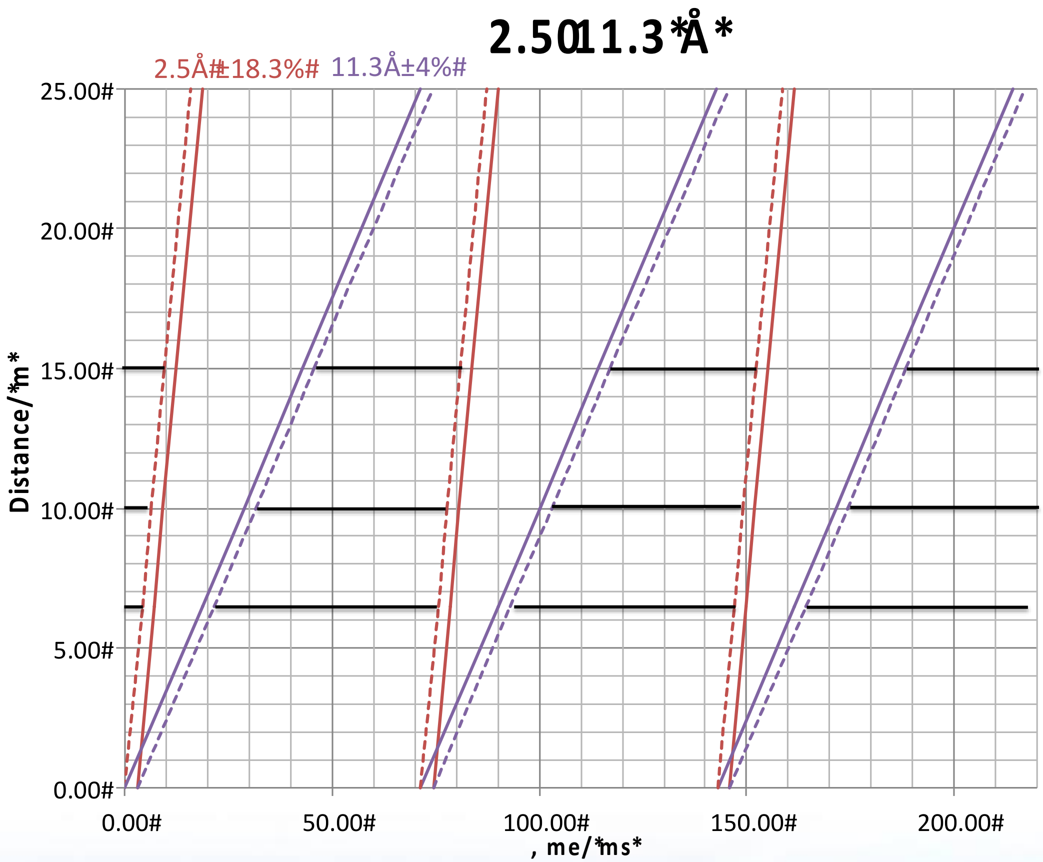
https://europeanspallationsource.se/instruments/freia#instrument-description

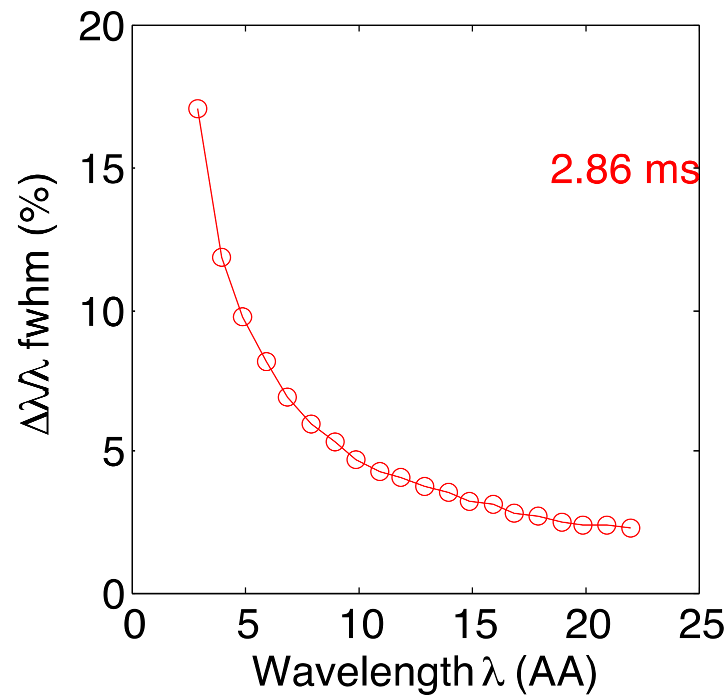




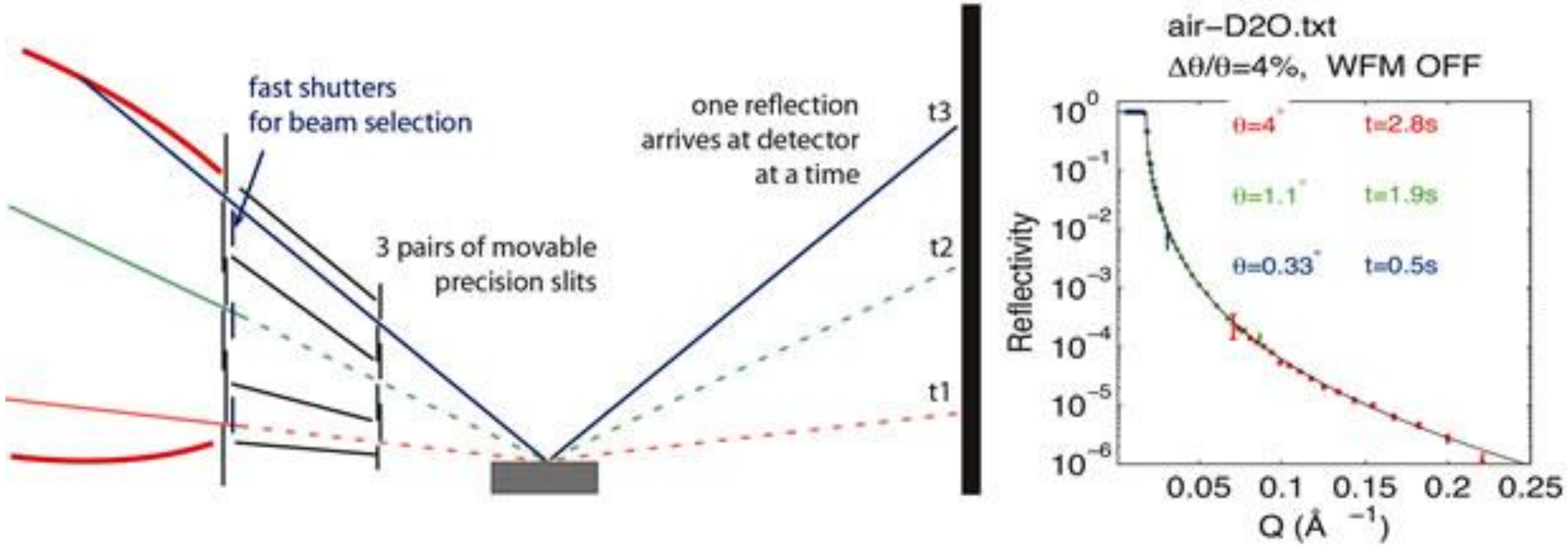
Freia proposal 2013: https://ess-public-legacy.esss.se/sites/default/files/freia_proposal.pdf

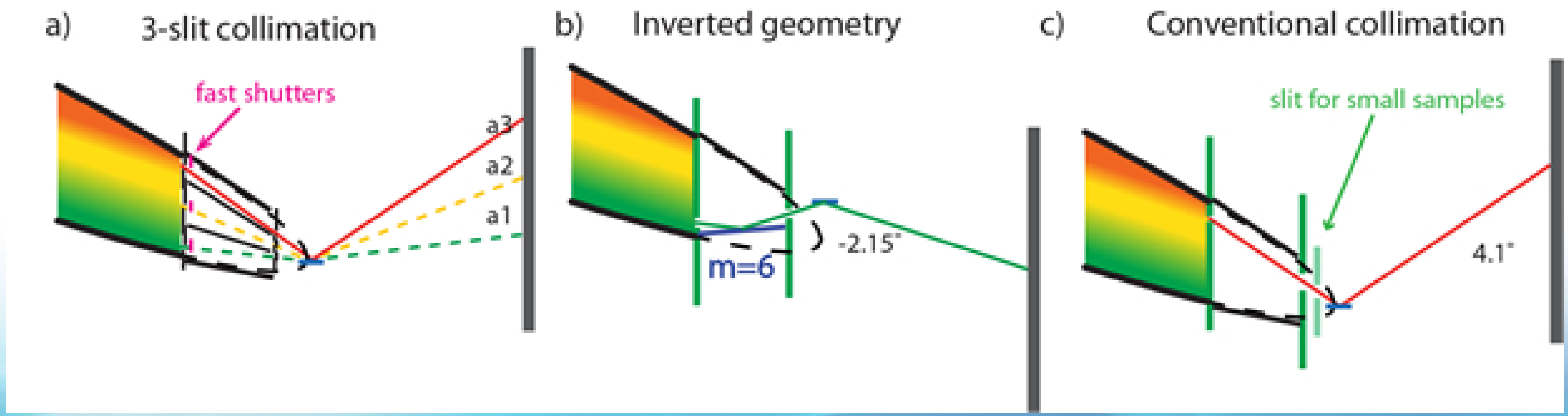
Intrinsic operation

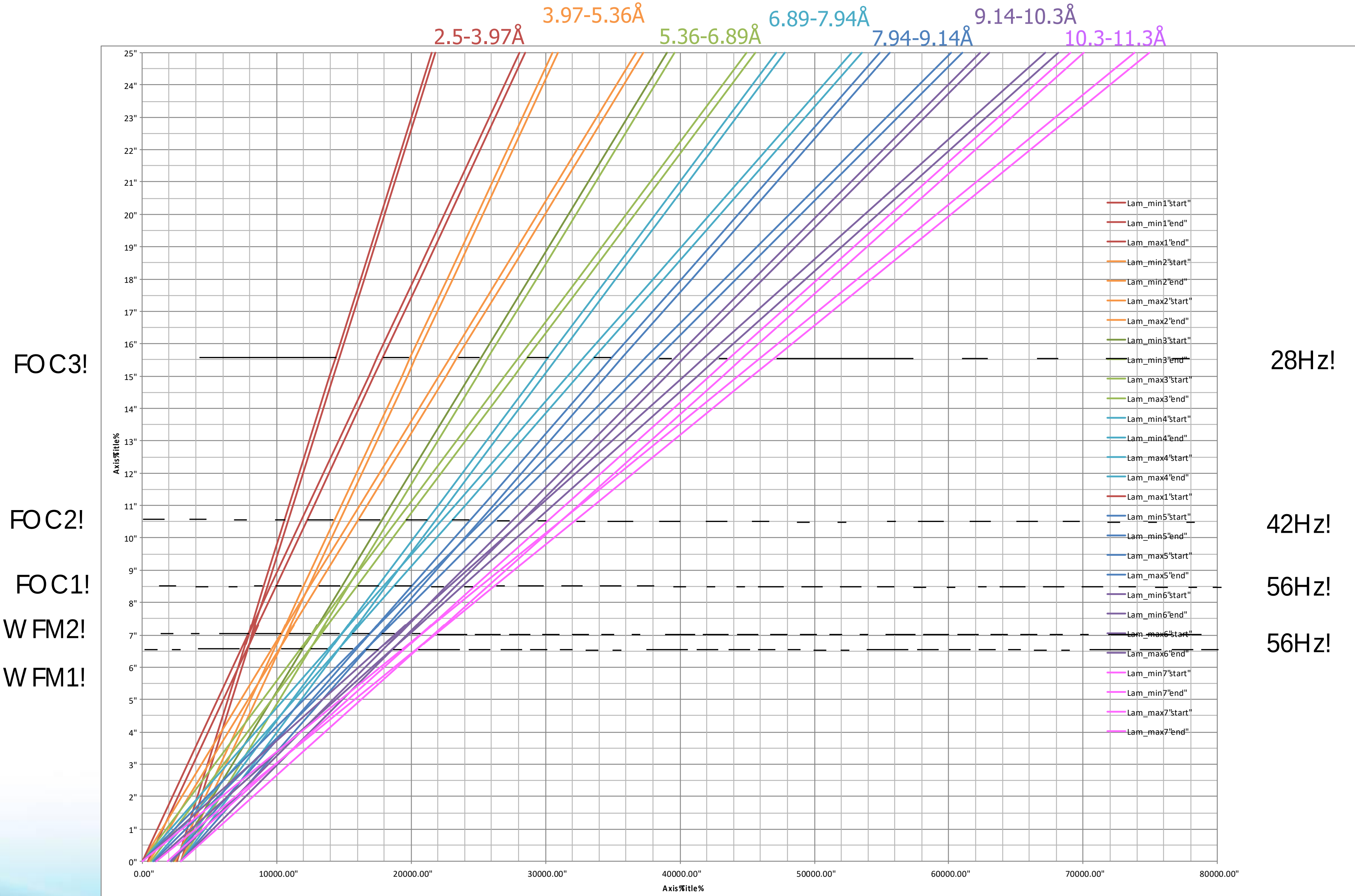




No need to move the sample!






2.5-3.97Å
3.97-5.36Å
5.36-6.89Å
6.89-7.94Å
7.94-9.14Å
9.14-10.3Å
10.3-11.3Å
FO C3!
FO C2!
FO C1!
W FM2!
W FM1!
28Hz!
42Hz!
56Hz!
56Hz!
Axis%itle%
Axis%itle%

Australian Government

ansto