Practical Experimental Considerations

by

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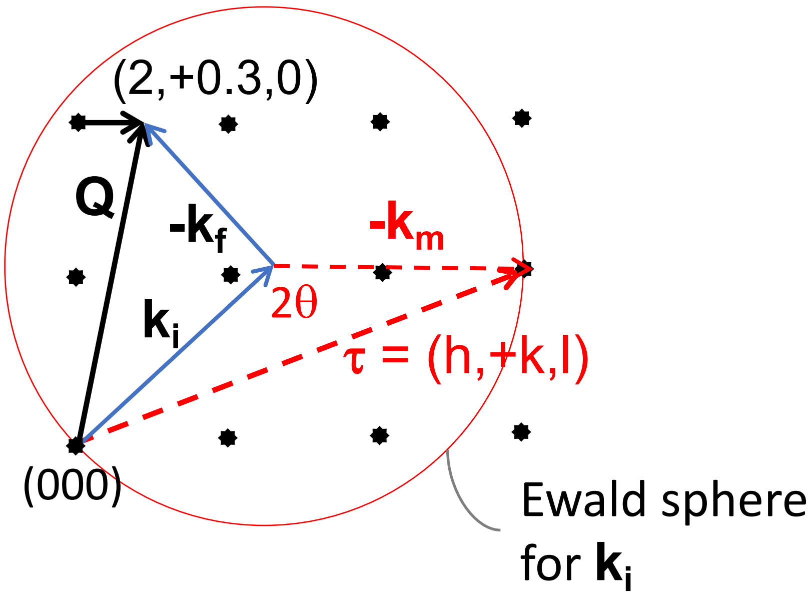
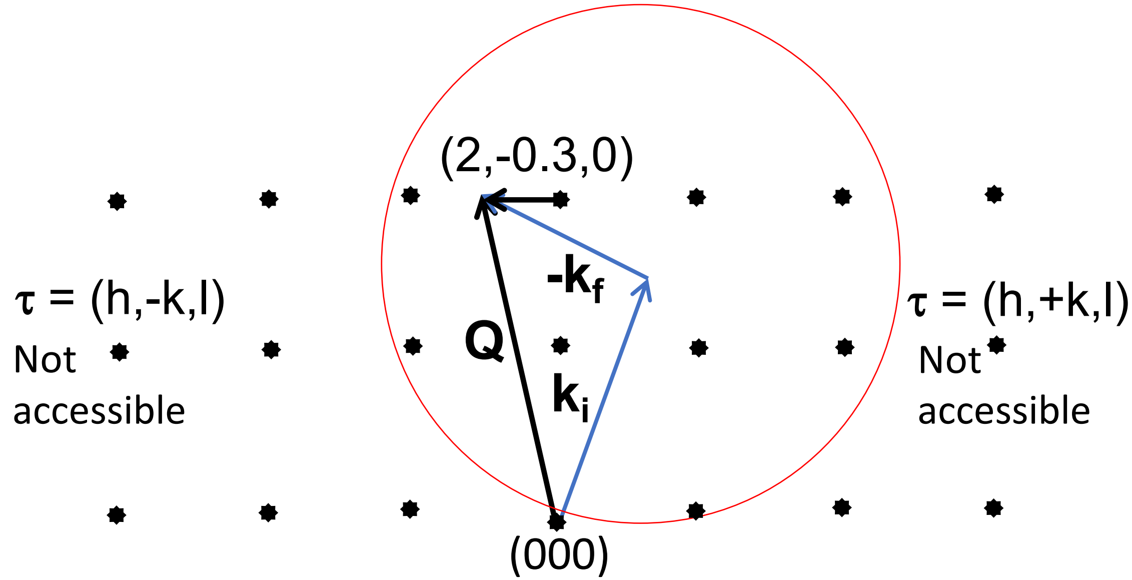
"Nothing looks so much like a new effect as a screw-up."

Dr. Thomas Noggle

Corollary: New effects often look like screw ups.

Simultaneous scattering for single crystals





$\mathbf{Q} = \mathbf{k_i} - \mathbf{k_f}$ $E = \frac{\hbar^2}{2m_n}\left(k_i^2 - k_f^2\right)$

Bragg first condition: $|\mathbf{k_i}|=|\mathbf{k_m}|=|\mathbf{k_i}-\tau|$ (shown)

Bragg second condition: $|\mathbf{k_f}|=|\mathbf{k_f}+\tau|$ (not shown)

Different conditions for symmetry equivalent points.
Important to check results in multiple zones.

Strong clue something is an artifact is that it breaks crystal symmetry.

Some effects of simultaneous scattering

i. Parasitic scattering (depleted beam intensity)

- Lines of weakened intensity streak across data

ii. Elastic-elastic double scattering with instrument

- Misplaced powder rings

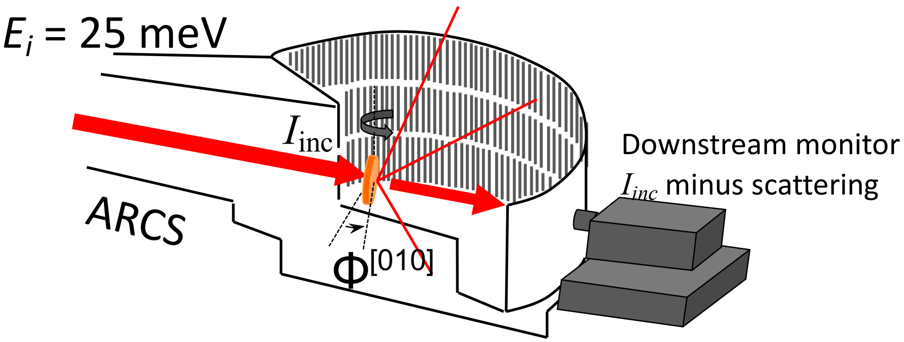
iii. Elastic-elastic double scattering within sample

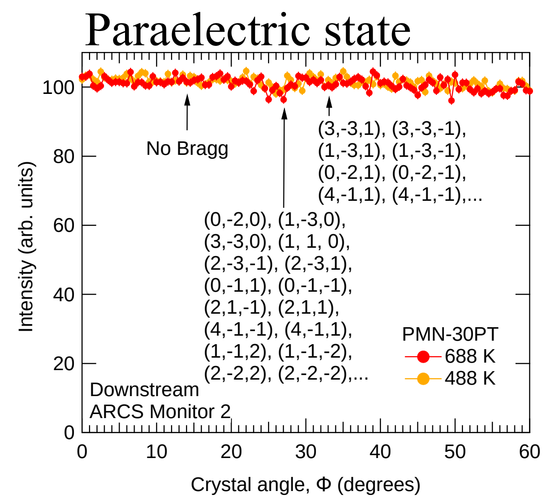
- Modified Bragg intensities

iv. Elastic-inelastic double scattering

- ‘Ghost’ excitations

Example: Paraelectric (cubic)/ferroelectric crystal (pseudo-cubic)

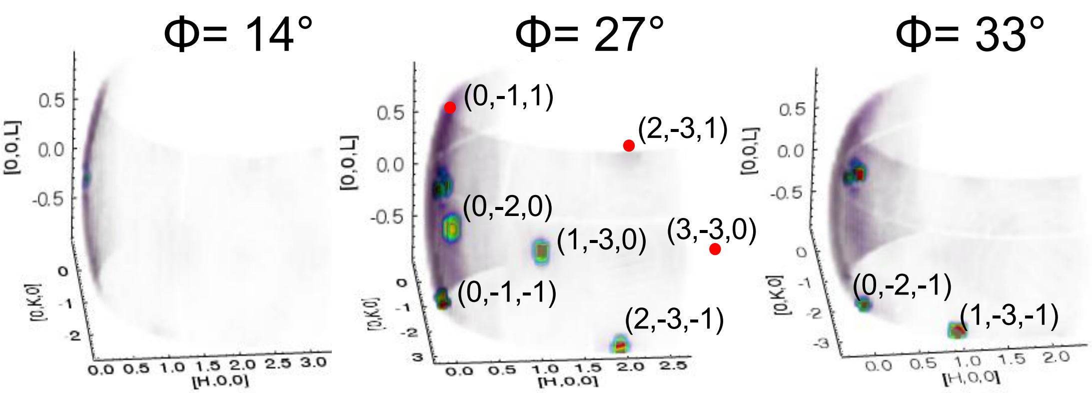


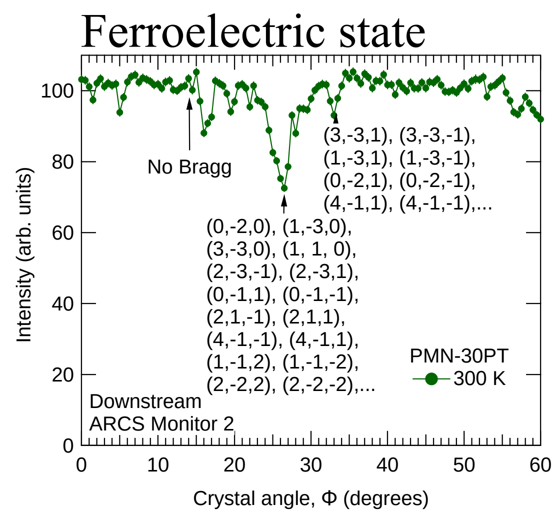


ARCS beam divergence ~0.6°

Paraelectric crystal mosaic ~0.02°

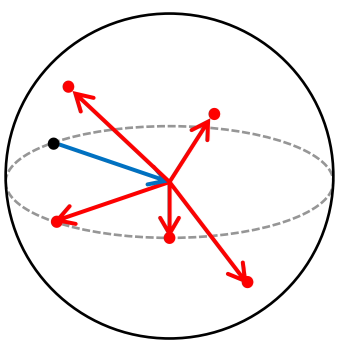
Angular acceptance of crystal much narrower than beam divergence.





Ferroelectric crystal mosaic ~0.2°

Angular acceptance of crystal is better match to beam divergence.

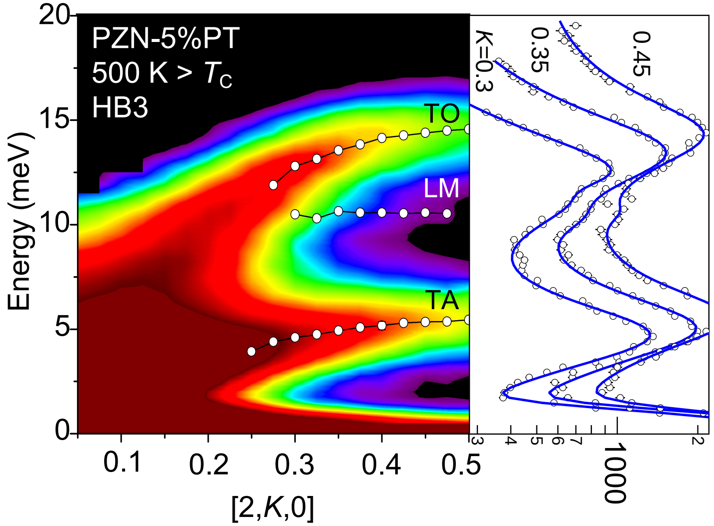


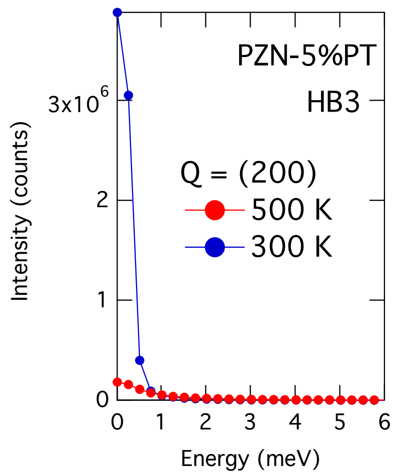
Ewald sphere for $\mathbf{k}_i$ simultaneously intersects 16 Bragg reflections at one angle!

Mosaic from ferroelectric domains increases flux of diffracted beams by an order of magnitude.

Parasitic diffraction line cuts through TA phonon

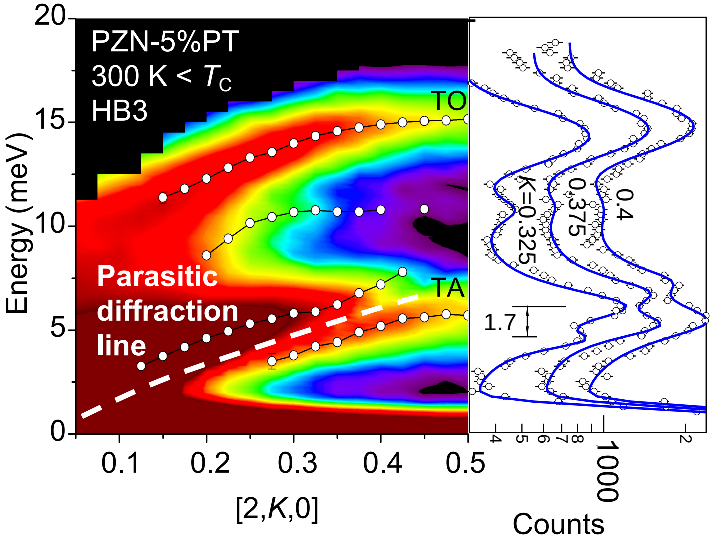
Paraelectric





$$\frac{I(200)_{300K}}{I(200)_{500K}} = 21.4$$

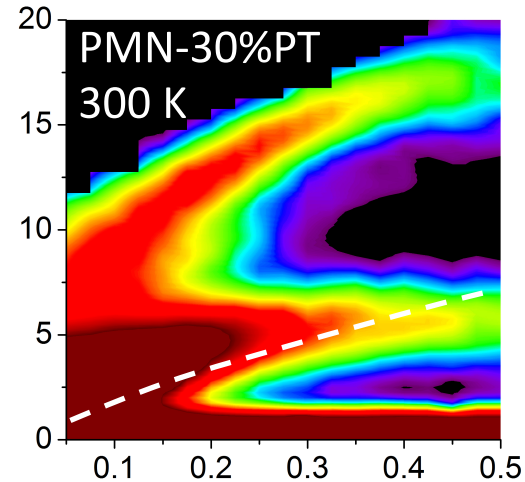
Ferroelectric



Thick crystal (~1.5 cm)

Parasitic diffraction condition along line extending out from strong (200) reflection

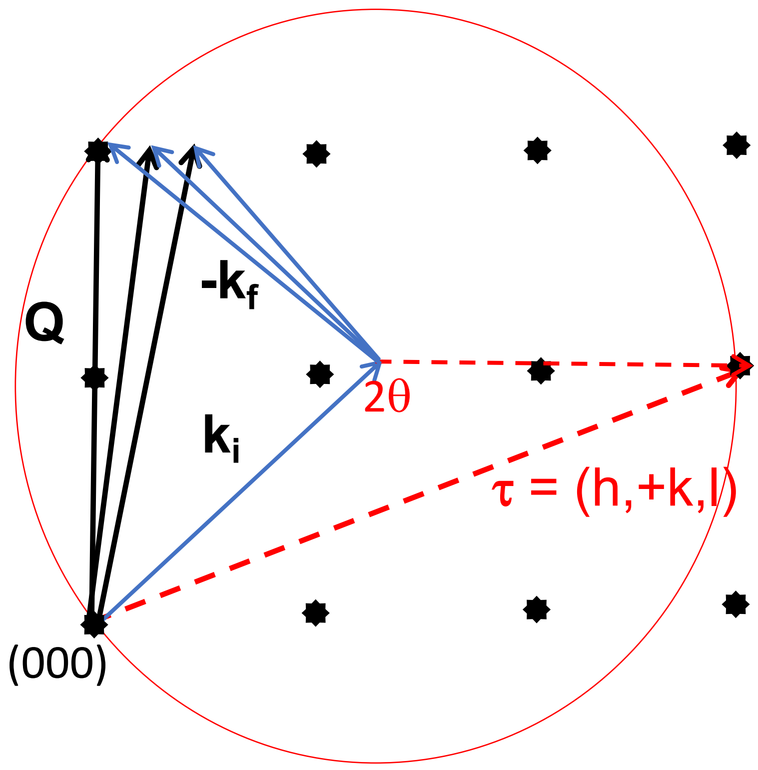
Ferroelectric

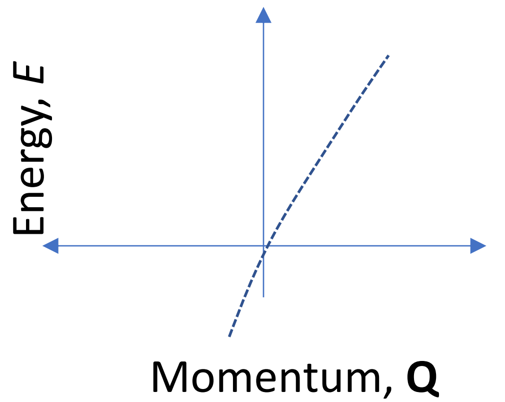


Thin crystal (~0.5 cm)

Minimize effect by reducing crystal size and/or mosaic (hard)

Explanation for slope of parasitic diffraction line (fixed k_i)





$$\mathbf{Q} = \mathbf{k_i} - \mathbf{k_f}$$

$$E = \frac{\hbar^2}{2m_n}\left(k_i^2 - k_f^2\right)$$

Useful command in HORACE for time-of-flight data

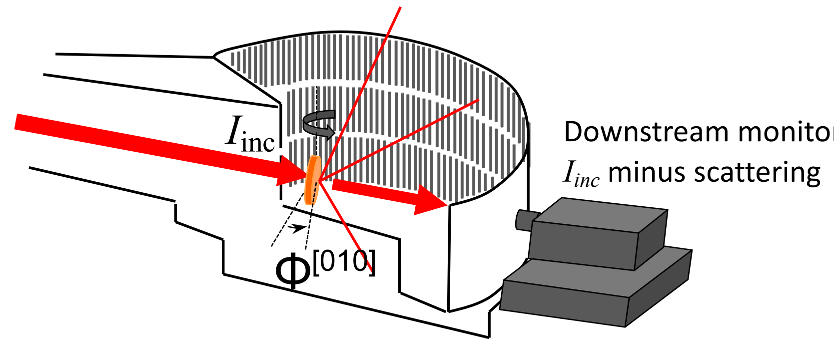
http://horace.isis.rl.ac.uk/Run_inspector

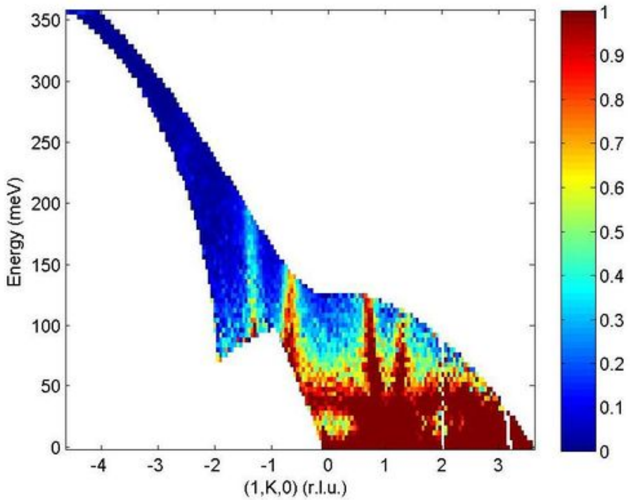
Run inspector

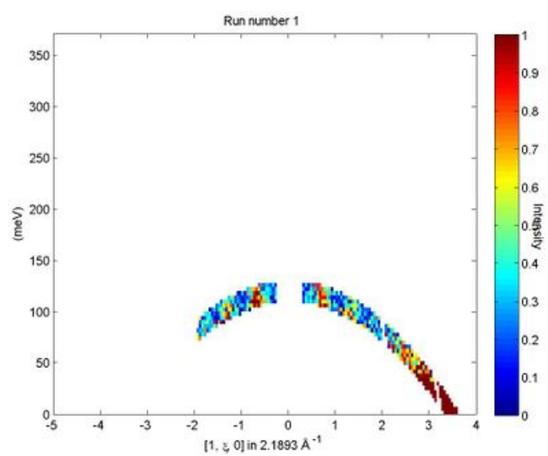
run_inspector

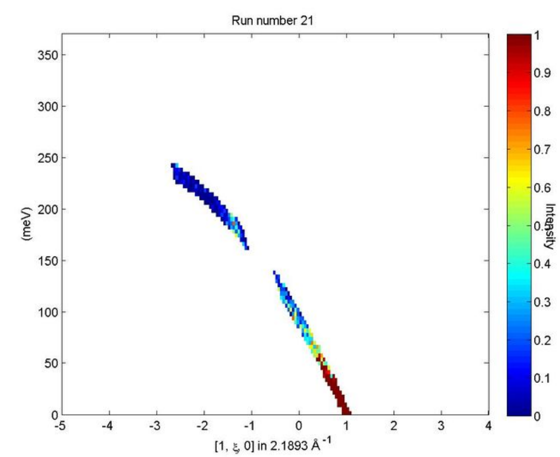
The `run_inspector` routine may be used on 1d or 2d sqw objects to plot the data from each individual run.

```
run_inspector(w)

run_inspector(w,'ax',[-5,4,0,370])

run_inspector(w,'ax',[-5,4,0,370],'col',[0,1])
```







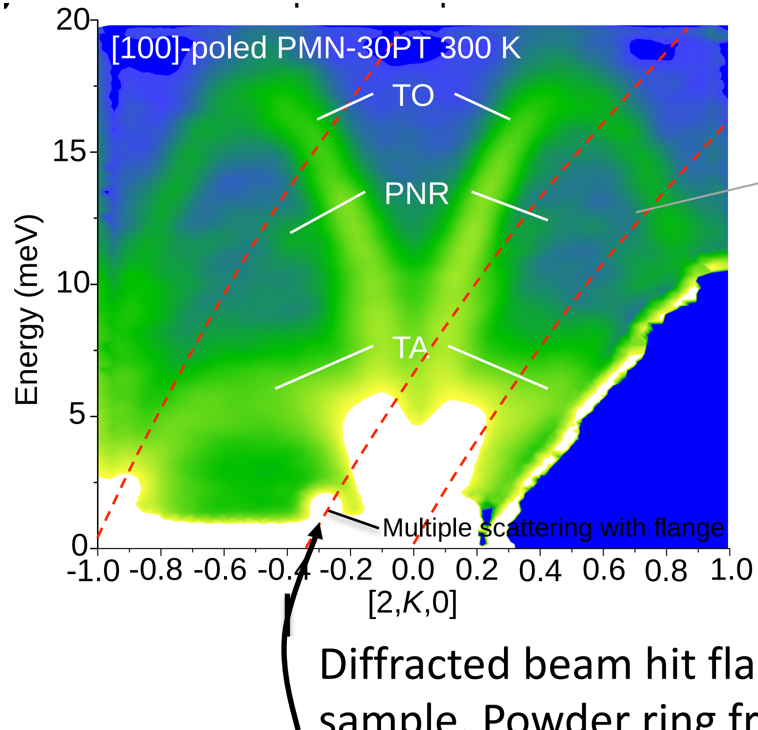


Allows you to find which run contributed to a particular part of the spectrum

Parasitic diffraction line depend on probe energy

ARCS fixed E_i = 25 meV

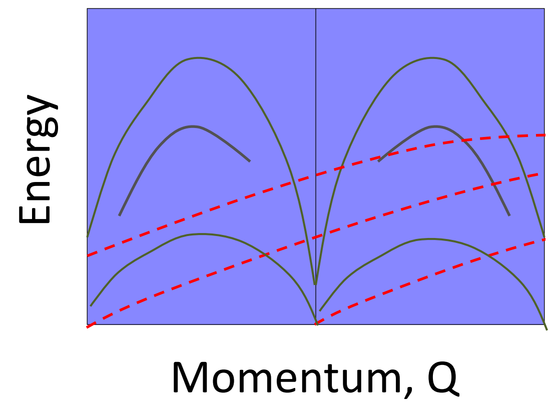
M. E. Manley, *et al.* *Phys. Rev. B* **94**, 104304 (2016).



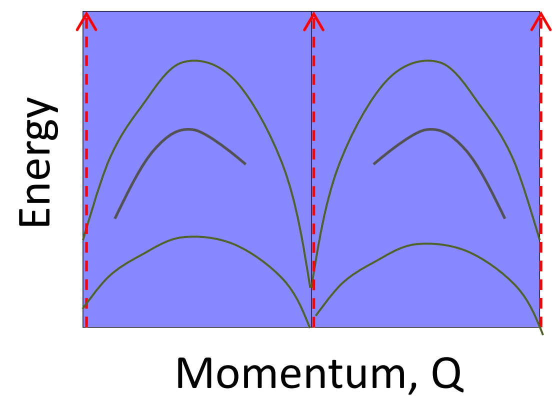
Diffraction by crystal along these lines (lines of constant crystal angle for fixed E_i).

Diffracted beam hit flange above sample. Powder ring from flange appears offset in energy because a path length increase on time-of-flight instrument mimics energy loss.

Lower energy probe squeezes lines into a smaller energy range

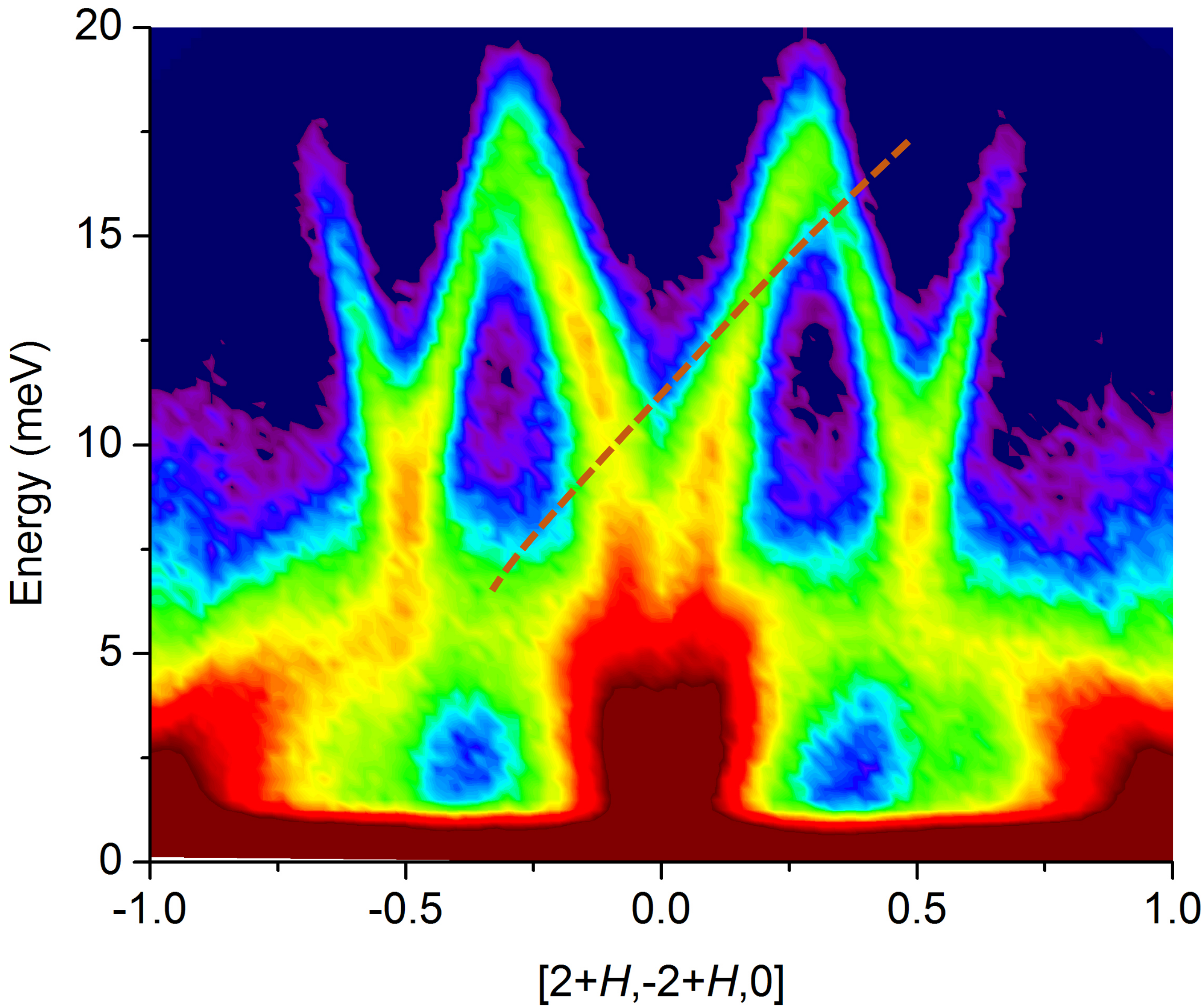


Limiting case of high energy probe (E_i):
$(E_i - E_f)/E_i \rightarrow 0$ (inelastic x-ray scattering is in this limit)



Vary the incident/final probe energy (easy) or instrument type (hard)

Parasitic diffraction lines break crystal symmetry



Can you identify the parasitic diffraction line in this data?

Large data sets collected on modern instruments make identification easier

Simultaneous reflections for single-crystal diffraction

Acta Cryst. (1964). **17**, 805

The Effects of Simultaneous Reflections on Single-Crystal Neutron Diffraction Intensities*

By R. M. Moon† and C. G. Shull

Department of Physics, Massachusetts Institute of Technology, Cambridge, Mass., U.S.A.

(*Received 8 July* 1963)

The intensity changes produced in single-crystal diffraction reflections when one or more secondary reflections occur simultaneously are discussed both theoretically and experimentally. The theory is an extension of the usual treatment of secondary extinction, based on the mosaic crystal model. An approximate solution, valid in the thin crystal limit, is in good agreement with neutron diffraction experiments on single crystals of iron. Both theory and experiment demonstrate the importance of sample geometry on the magnitude and sign of the simultaneous reflection effects. The effects may be minimized by controlling the sample geometry in addition to the usual precautions taken to reduce secondary extinction.

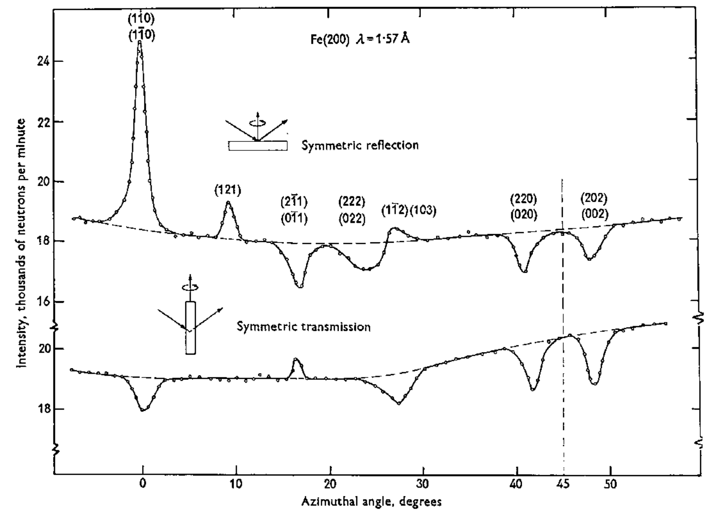
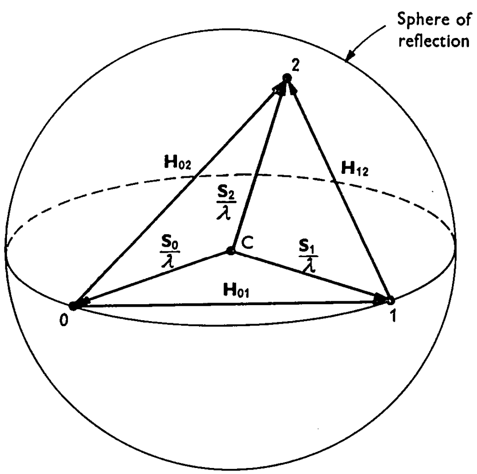
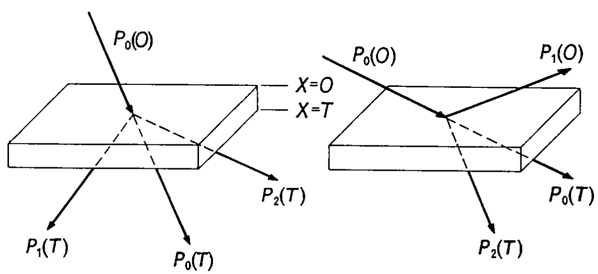
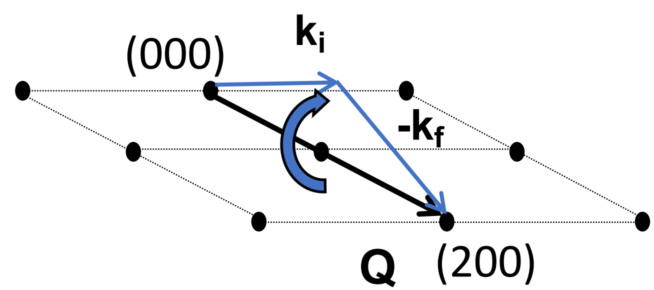


Fig. 5. Simultaneous reflection effects observed in the 200 reflection from iron as the crystal is rotated around the scattering vector. Pronounced differences are to be noted for the reflecting and transmitting crystals.







Simultaneous reflections alters intensities – check for by rotating about Q

Elastic-inelastic scattering for single crystals (ghostons)

ELSEVIER

Available online at www.sciencedirect.com

SCIENCE DIRECT

Physica B 350 (2004) 11–16

PHYSICA B

www.elsevier.com/locate/physb

Chasing ghosts in reciprocal space—a novel inelastic neutron multiple scattering process

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[b] *MDN/SPSMS/DRFMC, CEA-Grenoble, 38054 Grenoble, France*
[c] *Laboratoire de Cristallographie, CNRS, 38042 Grenoble, France*

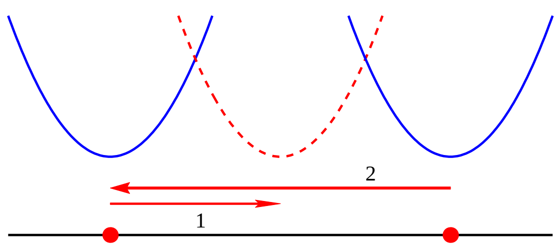
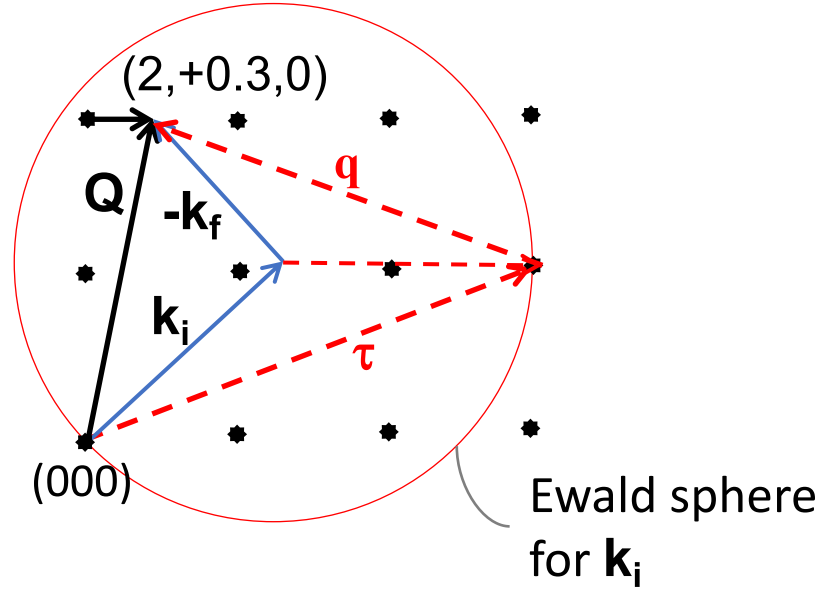


Fig. 4. Illustration of how multiple scattering shifts inelastic intensity in reciprocal space. (1) New 'ghoston' modes occur when a dispersion is shifted to an empty zone in reciprocal space. (2) Back-folding of intensity from the same excitation in a different zone.



Requires two conditions (Bragg first case):

1. Bragg condition: $|\mathbf{k_i}|=|\mathbf{k_i}-\boldsymbol{\tau}|$ (Bragg first)
2. Excitation at $\mathbf{q} = \mathbf{Q} - \boldsymbol{\tau}$ (with energy E)

Weak effect (<1-2%) but common because dispersion curves are everywhere

Example: Multiple scattering in liquid ^{4}He

PHYSICAL REVIEW B **99**, 165425 (2019)

Phonons, rotons, and localized Bose-Einstein condensation in liquid ^{4}He confined in nanoporous FSM-16

Jacques Bossy,[1] Jacques Ollivier,[2] and H. R. Glyde[3]
[1]*Institut Néel, CNRS, Université Grenoble Alpes, BP 166, 38042 Grenoble Cedex 9, France*
[2]*Institut Laue-Langevin, BP 156, 38042 Grenoble, France*
[3]*Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716-2593, USA*

"ghost" roton from multiple scattering

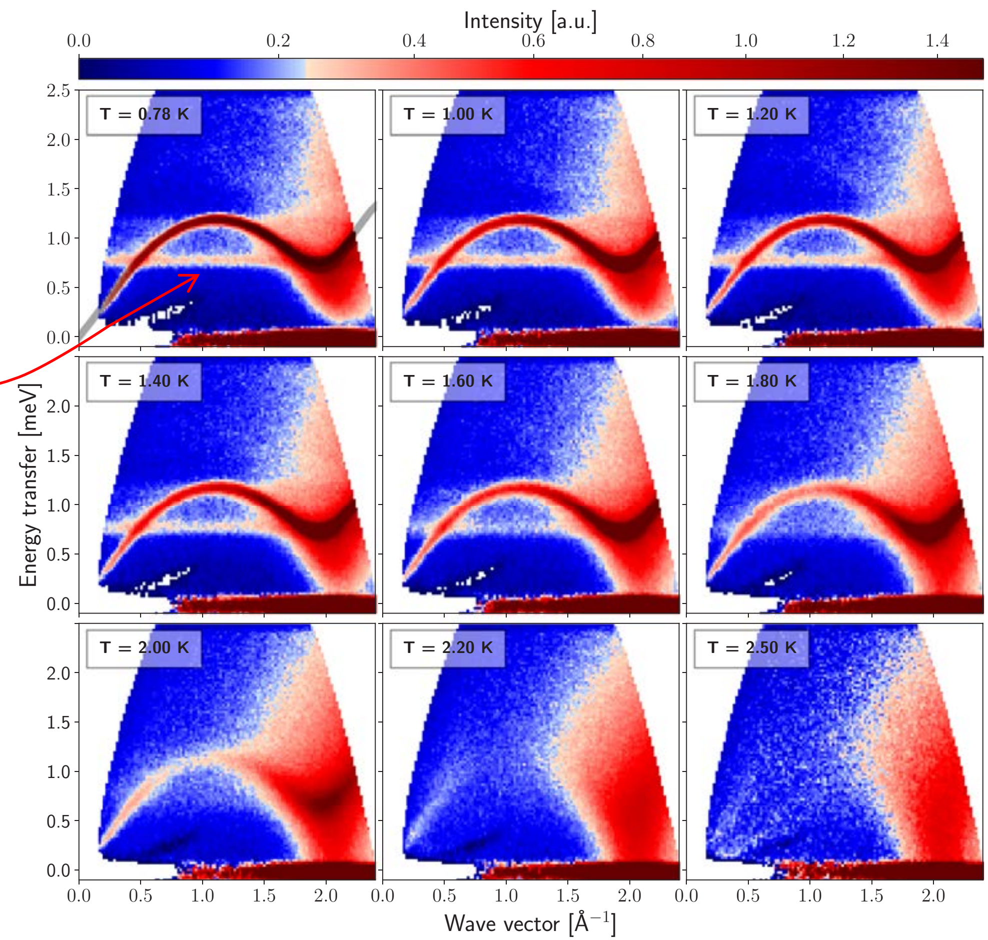
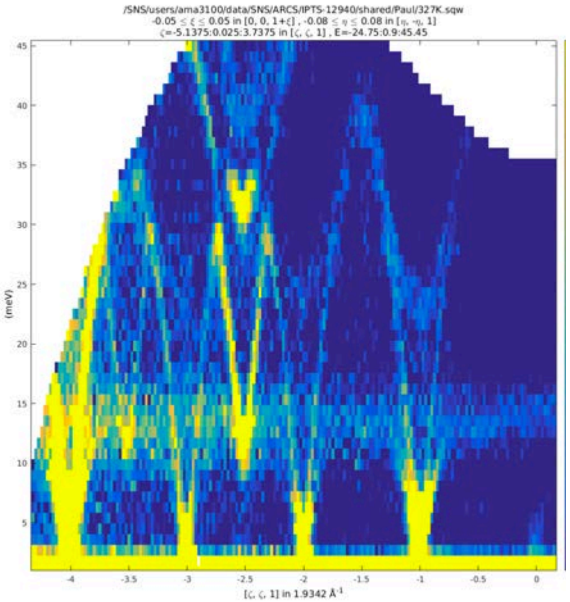
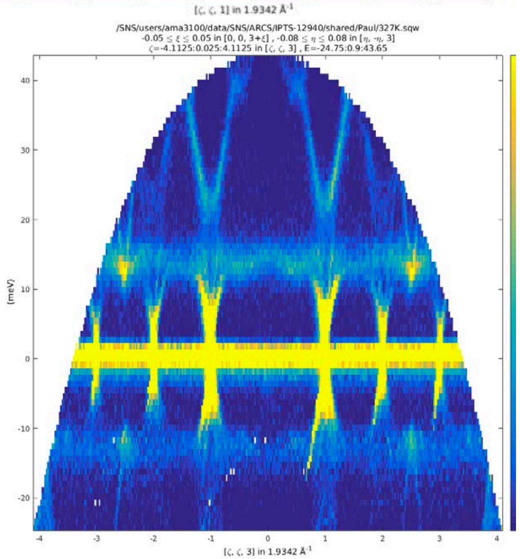


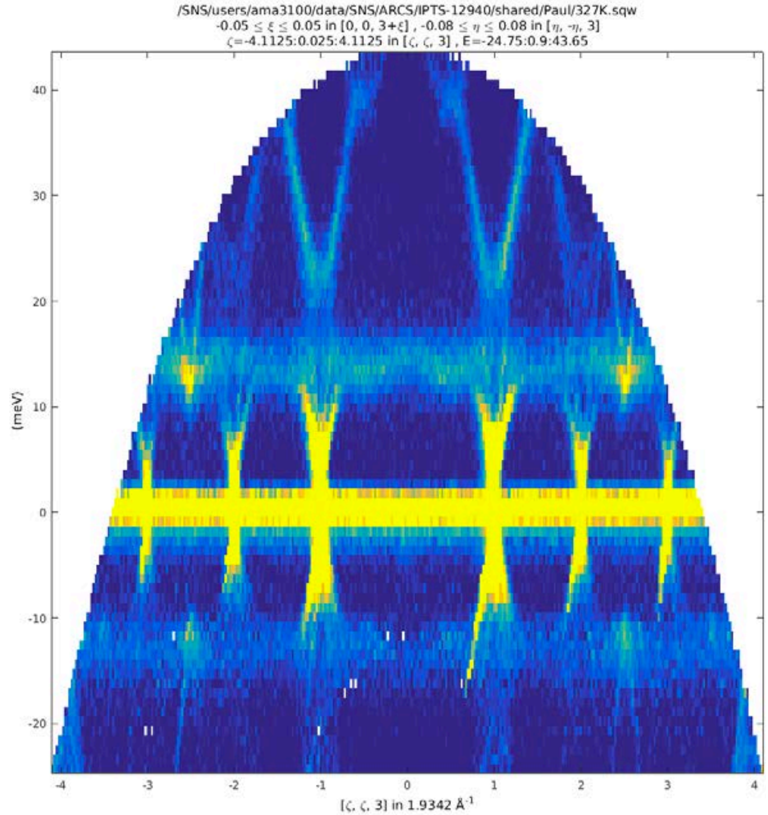
FIG. 5. As Fig. 3 at filling (4), $N_4 = 43.5$ mmol/g, overfilled nanopores. An intense, well-defined P-R mode of bulk liquid ^{4}He and multiple scattering from the roton at temperatures up to $T = 2.0$ K is observed.

For liquids/glasses and powders multiple scattering appears incoherent

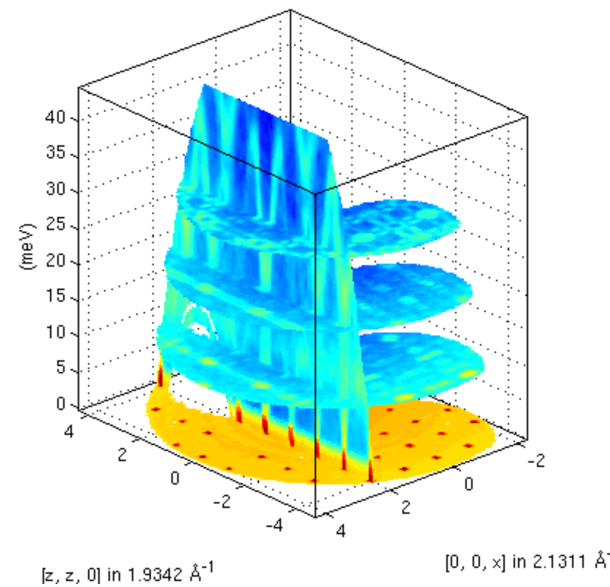
Example: Incoherent scattering in TiO_2







Neutron scattering lengths and cross sections							
Isotope	conc	Coh b	Inc b	Coh xs	Inc xs	Scatt xs	Abs xs
Ti	---	-3.438	---	1.485	2.87	4.35	6.09
46Ti	8.2	4.93	0	3.05	0	3.05	0.59
47Ti	7.4	3.63	-3.5	1.66	1.5	3.2	1.7
48Ti	73.8	-6.08	0	4.65	0	4.65	7.84
49Ti	5.4	1.04	5.1	0.14	3.3	3.4	2.2
50Ti	5.2	6.18	0	4.8	0	4.8	0.179

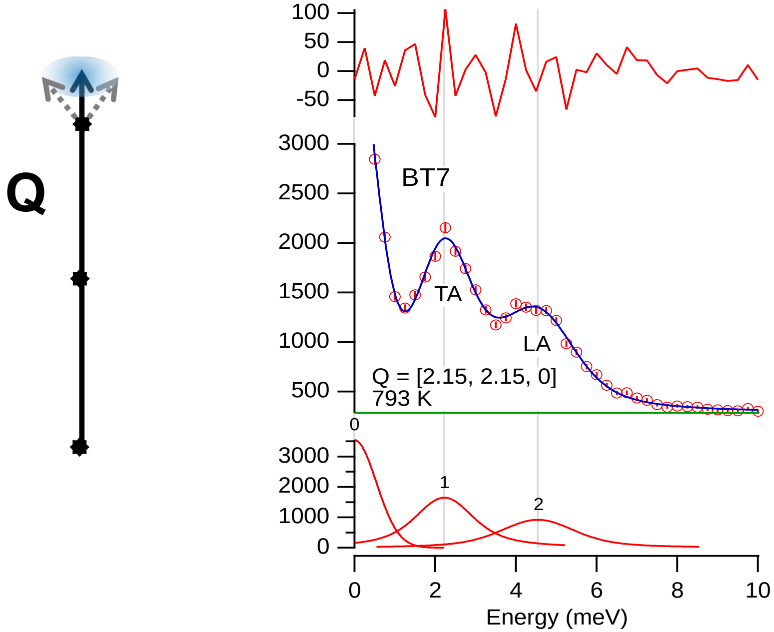


Incoherent scattering cross section projects the transform of the Ti self-correlation function – related to Ti partial phonon DOS.

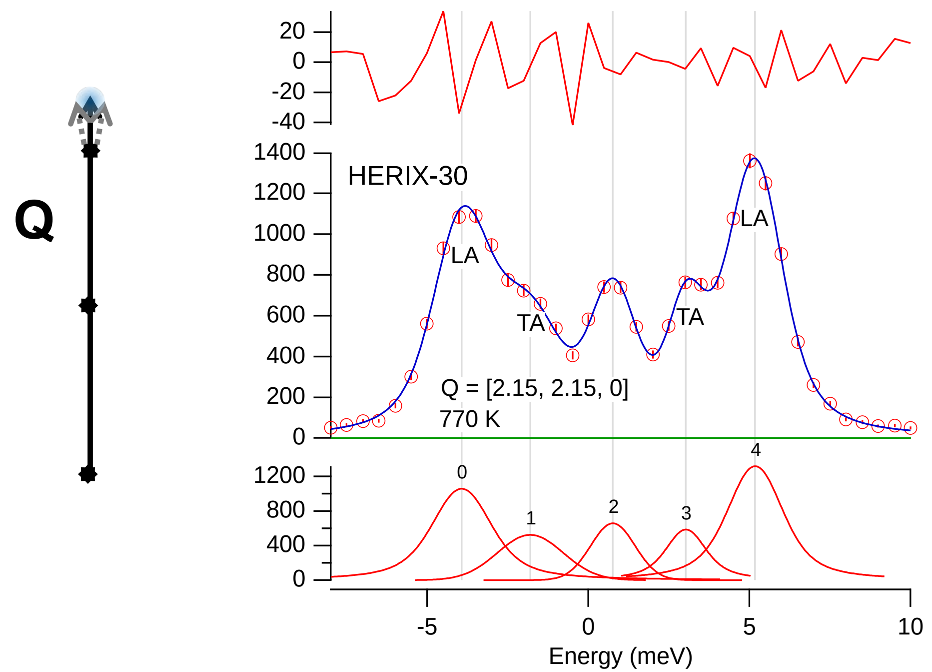
"Forbidden" modes (resolution effect)

Example: Transverse acoustic phonon in longitudinal scan in PbSe

Triple-axis inelastic neutron scattering (BT7, NIST)



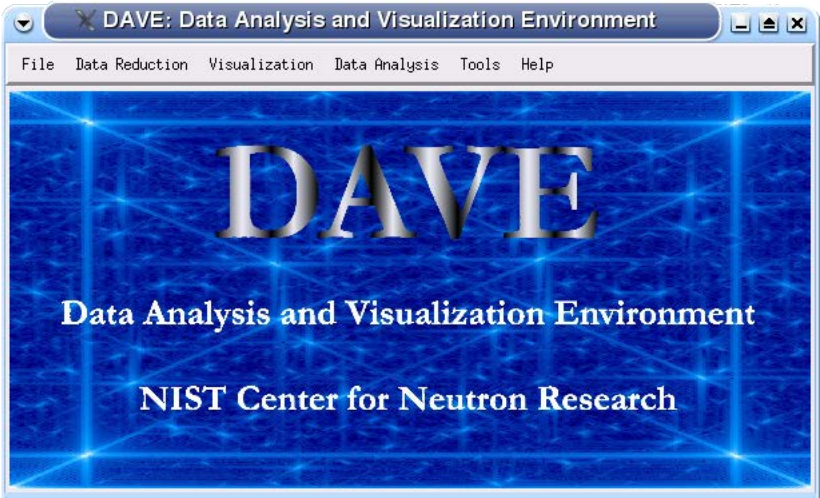
Inelastic x-ray scattering (HERIX-30, APS)

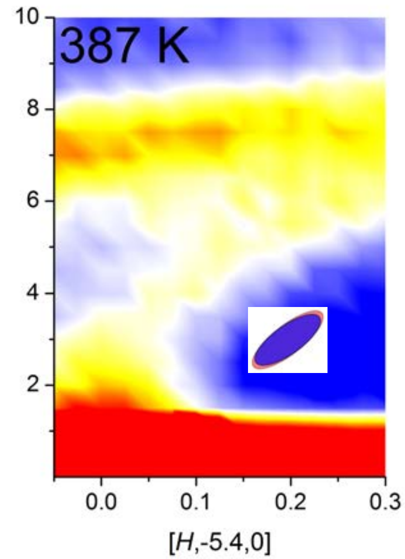


M. E. Manley, *et al.* *Nature Commun.* **10**, 1928 (2019).

Finite Q resolution introduces transverse components in longitudinal scans

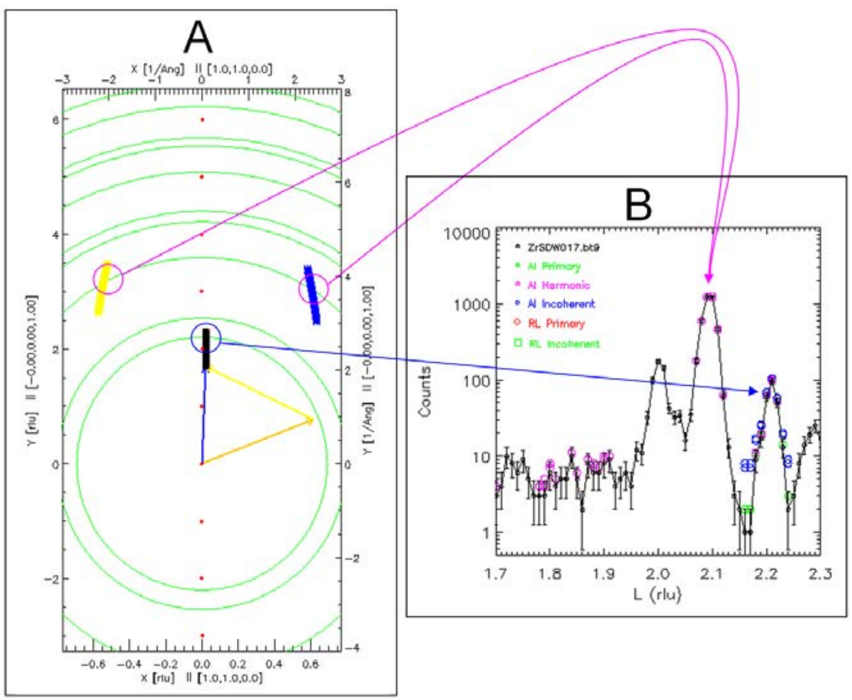
Useful software tools for triple-axis



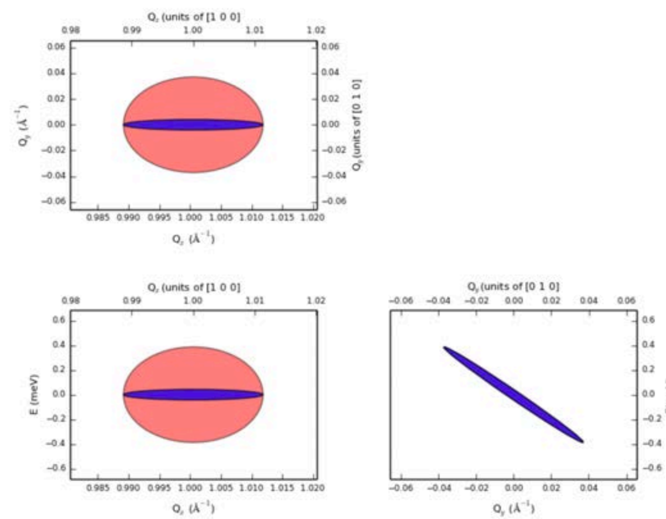


M. E. Manley *et al.* “Supersonic propagation of lattice energy by phasons in fresnoite,” [*Nature Commun.* **9**, 1823 (2018).]()

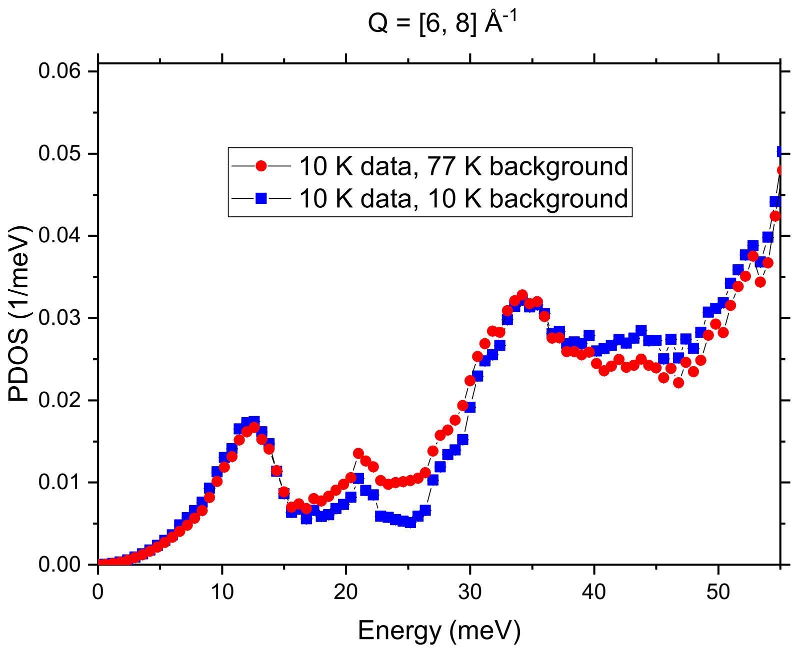
Spurion calculator for triple-axis

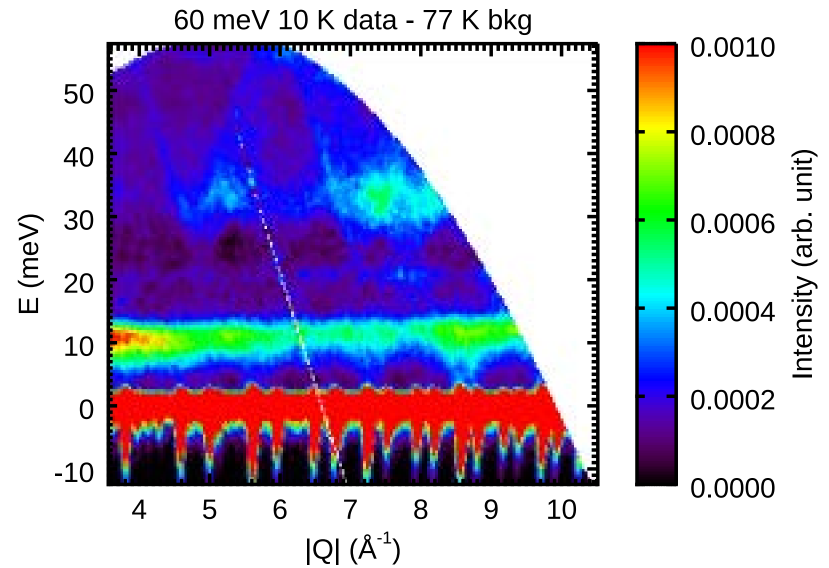


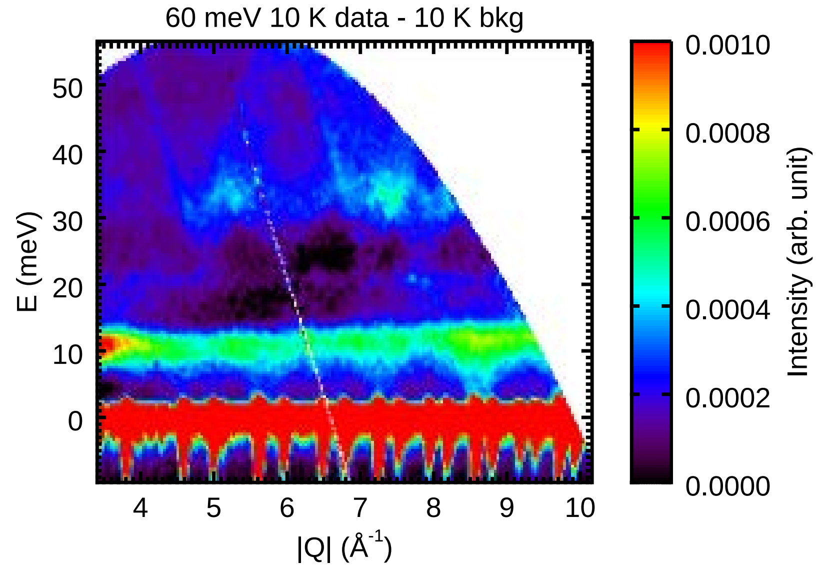
Resolution calculator for triple-axis
http://reflectometry.org/tas/res/



Example: Background in UO_2 phonon DOS measurement

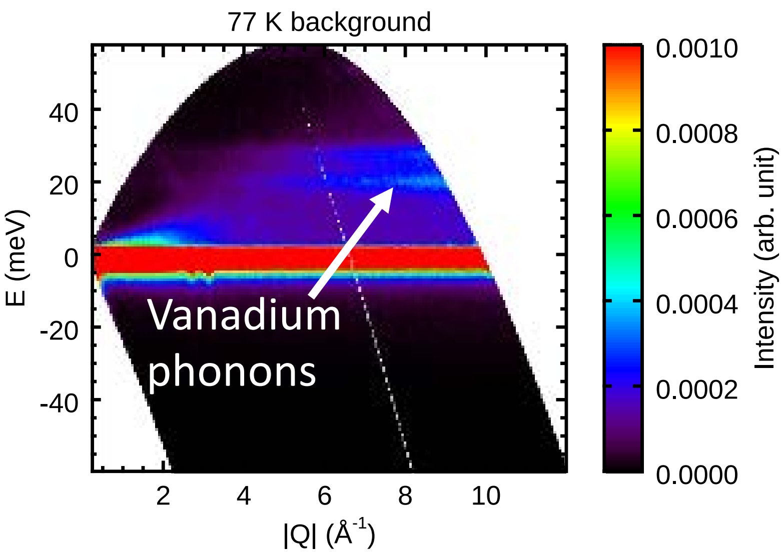


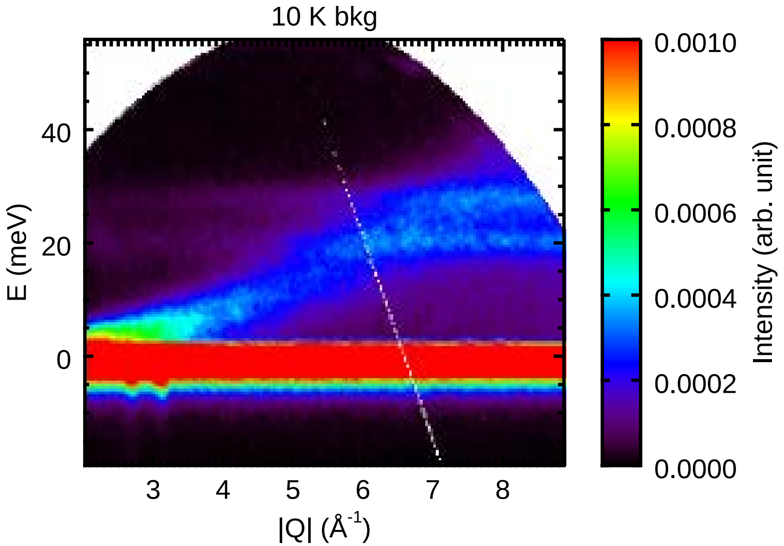


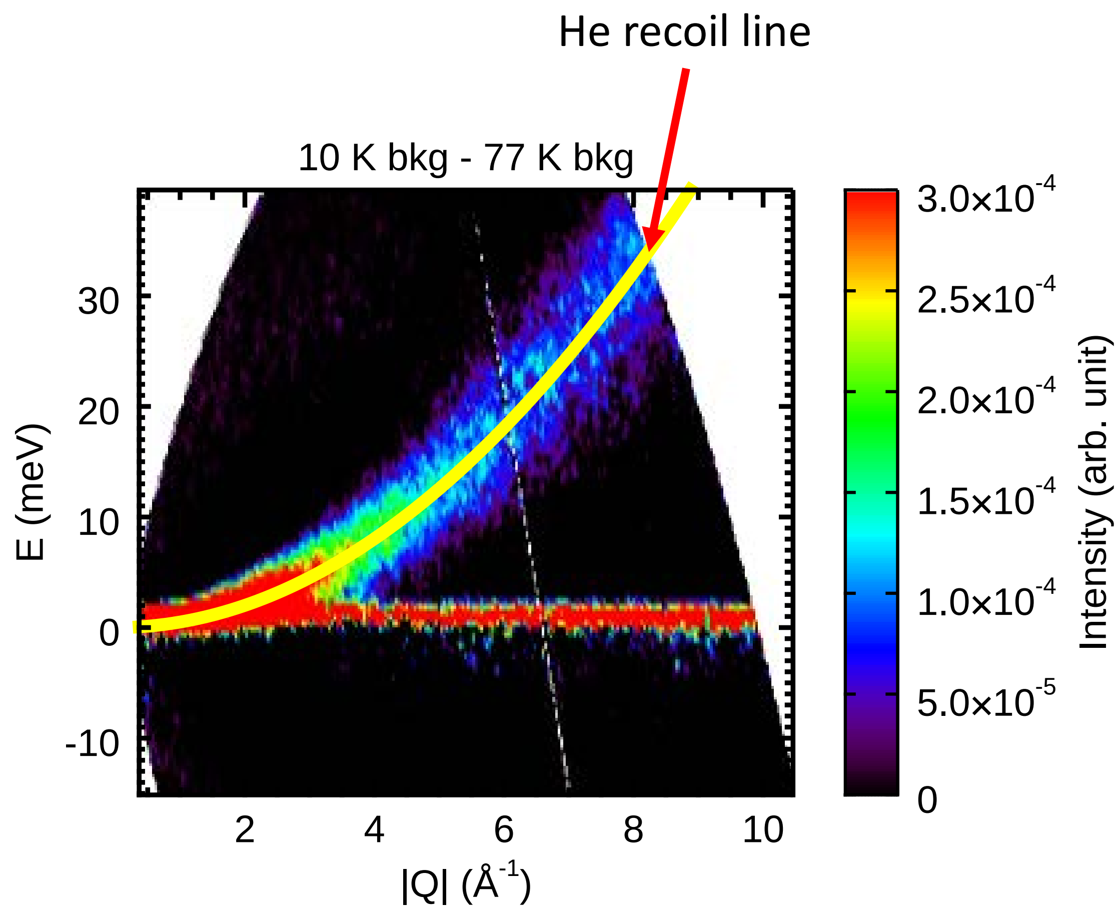


Courtesy of Matt Bryan (ORNL)

Recoil from Helium exchange gas







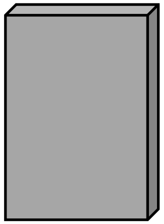
Over subtraction occurred because there was more exchange gas in the 'empty' sample can than in the loaded can (obviously).

Courtesy of Matt Bryan (ORNL)

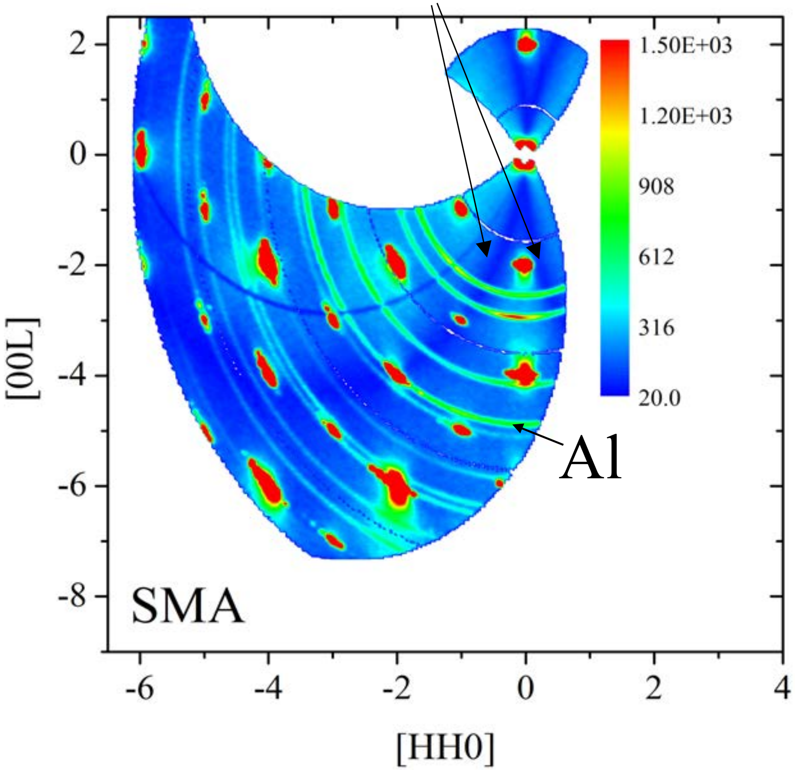
Absorption correction

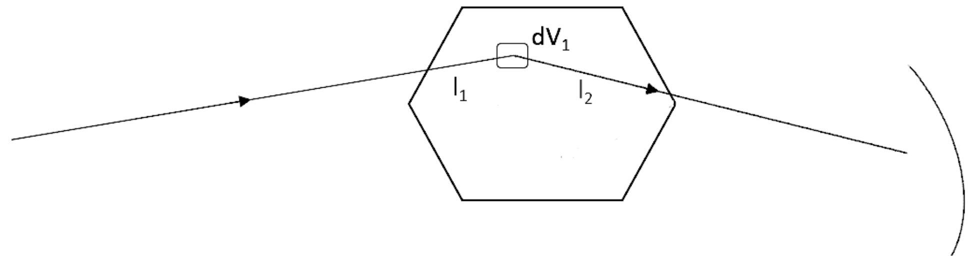
Magnetic Heusler alloy $Ni_{45}Co_5Mn_{36.6}In_{13.4}$

Flat plate geometry



Absorption 'shadows'





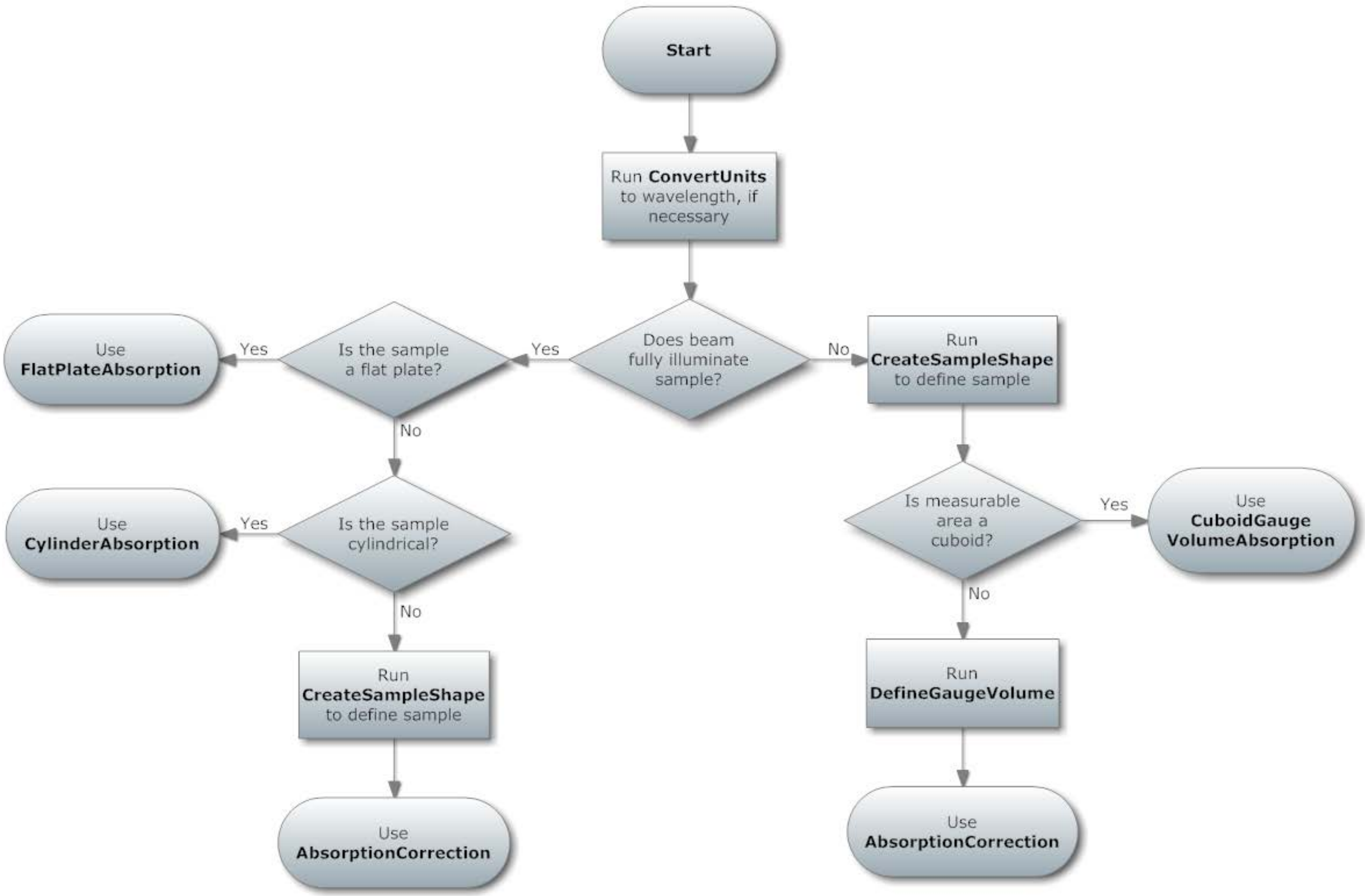
$$dI_1(\theta) = J_0 \rho \frac{d\sigma}{d\Omega}(\theta)\, exp\left[-\mu(\lambda_1)l_1 + -\mu(\lambda_2)l_2\right] dV$$

$$I_1(\theta) = \int_V dI_1$$

Detailed absorption correction calculations are always possible – but you cannot correct for no signal!

Absorption correction algorithms available in MANTID

https://docs.mantidproject.org/nightly/algorithms/AbsorptionCorrection-v1.html



“When you have eliminated all which is impossible, then whatever remains, however improbable, must be the truth.”

— Arthur Conan Doyle, The Case-Book of Sherlock Holmes