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

\[ \mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f \]

\[ E = \frac{\hbar^2}{2m_i} (k_i^2 - k_f^2) \]

Bragg first condition: \(|\mathbf{k}_i| = |\mathbf{k}_m| = |\mathbf{k}_i - \mathbf{\tau}| \) (shown)

Bragg second condition: \(|\mathbf{k}_f| = |\mathbf{k}_f + \mathbf{\tau}| \) (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

\textit{i.} Parasitic scattering (depleted beam intensity)
  - Lines of weakened intensity streak across data
\textit{ii.} Elastic-elastic double scattering with instrument
  - Misplaced powder rings
\textit{iii.} Elastic-elastic double scattering within sample
  - Modified Bragg intensities
\textit{iv.} Elastic-inelastic double scattering
  - ‘Ghost’ excitations
**Example: Paraelectric (cubic)/ferroelectric crystal (pseudo-cubic)**

$E_i = 25 \text{ meV}$

Paraelectric state

- Ewald sphere for $k_i$ simultaneously intersects 16 Bragg reflections at one angle!

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

Ferroelectric state

- Angular acceptance of crystal is better match to beam divergence.

ARCS beam divergence ~$0.6^\circ$

Paraelectric crystal mosaic ~$0.02^\circ$

Angular acceptance of crystal much narrower than beam divergence.

ARCS beam divergence ~$0.6^\circ$

Ferroelectric crystal mosaic ~$0.2^\circ$

Angular acceptance of crystal is better match to beam divergence.

No Bragg

Paraelectric state

- Paraelectric crystal mosaic ~$0.02^\circ$

Angular acceptance of crystal much narrower than beam divergence.

Ferroelectric state

- Angular acceptance of crystal is better match to beam divergence.

Ewald sphere for $k_i$ simultaneously intersects 16 Bragg reflections at one angle!
Parasitic diffraction line cuts through TA phonon

Paraelectric

Ferroelectric

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

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

Minimize effect by reducing crystal size and/or mosaic (hard)
Explanation for slope of parasitic diffraction line (fixed $k_i$)

\[ Q = k_i - k_f \]

\[ E = \frac{\hbar^2}{2m_n} (k_i^2 - k_f^2) \]
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])

Allowing 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


Lower energy probe squeezes lines into a smaller energy range

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

Limiting case of high energy probe ($E_i$):

$$(E_i - E_f)/E_i \to 0 \text{ (inelastic x-ray scattering is in this limit)}$$

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.

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.

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Simultaneous reflections alters intensities – check for by rotating about Q
Elastic-inelastic scattering for single crystals (ghostons)

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

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Available online at www.sciencedirect.com


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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: \(|\mathbf{k}_i| = |\mathbf{k}_i - \mathbf{\tau}| \) (Bragg first)
2. Excitation at \( \mathbf{q} = \mathbf{Q} - \mathbf{\tau} \) (with energy \( E \))

Weak effect (<1-2%) but common because dispersion curves are everywhere
Example: Multiple scattering in liquid $^4$He


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

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"ghost" roton from multiple scattering

FIG. 5. As Fig. 3 at filling (4), $N_c = 43.5$ 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$_2$

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

Triplet-axis inelastic neutron scattering (BT7, NIST)  Inelastic x-ray scattering (HERIX-30, APS)


Finite Q resolution introduces transverse components in longitudinal scans
Useful software tools for triple-axis


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$_5$Mn$_{36.6}$In$_{13.4}$

Absorption 'shadows'

Flat plate geometry

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