Background Correction for SANS Measurements II

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Talk Overview

**Background signal**: Any signal collected by detector that is not produced by Small-angle scattering (SAS) from *desired* sample structure.

- Detector Efficiency Contribution to BGD
- Double Bragg Scattering / Precipitates / Slit Diffraction
- Integrating (adding) BGD along flight path
- Gas scattering: Ar, He and Air (Sachs & Teller theory)
- Liquid scattering: H₂O, D₂O, and PMMA (plexiglas)
- Phonon (inelastic) scattering in Solids (Debye model)
- How to improve S/N
- Conclusions
**Motivation**: Ability to reduce background to make more sensitive measurements of Weak scattering samples: $S \ll N$

Rubinson et al, 2008
0.5% Protein in $D_2O$

Russell et al, 1995 170 nm thick sample
*Macromolecules, Vol. 28, No. 3, 1995*

**Figure 6**
SANS data from 5 mg ml$^{-1}$ lysozyme in $D_2O$ buffer and the resulting model SANS curves from $CRYSON$ and $XTAL2SAS$ using the structure from the PDB-listed parameters of 6lyz. The $CRYSON$ curve represents the best fit to the SANS data assuming a 3 Å bound $D_2O$ layer (fit parameters shown in Table 4), whereas the $XTAL2SAS$ model curve assumes no hydration layer. The $XTAL2SAS$ curve with a baseline having a constant 0.0035 cm$^{-1}$ subtracted is also shown for comparison. Error bars in the data for $q < 0.2 \ \text{Å}^{-1}$ are smaller than the data points. The inset shows the SANS curve in the full measured $q$ range.

**Figure 3.** $d\Sigma/d\Omega$ as a function of $Q$ for a mixture of PS-760K and dPS-690K (a) as-cast from a toluene solution and (b) heated to 130 °C for 15 h. The sample thickness is 170 nm. The solid lines are fits the scattering profiles using a Debye function.
**Background Correction**

Three separate scattering measurements:
Sample, Empty and Beam Blocked

Corrected = Sample – T*Empty – (1-T)* Blocked Beam

\[ I_c(q_i) = I_s(q_i) - T_s I_E(q_i) - (1 - T_s) I_B(q_i) \]

**Assumption:** Detector efficiency is the same or is corrected between measurements.

Detector “dead time” correction: Detector efficiency decreases as the count rate increases

\[ c_t = \frac{c_o}{1 - c_o t_d} \quad t_d = \text{detector dead time} \]
\[ c = \text{count rate, } c_\text{observed, } t_\text{true} \]

Correction incorporated in IGOR data reduction software.

Dead time is measured as ratio (R) of count rate with the two different size apertures:

\[ R_o = R_t + t_d (1 - R_t) c_{so} \]

All three SANS Ordela detectors exhibit oscillating detector efficiency dependence on count rate, which is **NOT** corrected by data reduction.

\[ \rightarrow \text{Background is not completely subtracted: } f_E I_E + f_B I_B \]
Detector Efficiency can also change with **time**: Measured **VSANS tube detectors** efficiency once an hour over two days.

Observed 7% variation caused by amplifier gain changing with temperature.

**Ordela** 2D Detectors are stable to 0.1% day to day.

Pulse Height Spectrum for one-hour runs made 2.5 hours apart. Observed count rates are 729 s⁻¹ for first and 752 s⁻¹ for second run.
**Double Bragg Scattering:**
Produces scattering which mimics structural SANS
Eliminated if wavelength chosen larger than $2d_{\text{max}}$

**Structural Metal Alloys:**
Typically contain small precipitates that scatter strongly!!!
Some alloys have high strength with lower BGD
{ fine grain size + substitional + martensitic }
Example: Ti – 6Al – 4V

Scattering from 99.99% pure Al and Al alloy 6061

Scattering from Al alloy 6061 having preferred texture
Aligning disks along (111) orientation.
{ Guinier Preston Zones – copper precipitates.}
Slit Diffraction: Fraunhofer diffraction of the beam occurs as it passes through the circular sample aperture

\[
\frac{dS_F}{dq}(q) = 4 \frac{R_2^2}{qR_2} J_1(qR_2)
\]

\[
\frac{d}{dq}(q) = \frac{4}{R_2d_s} q^3
\]

R_2 = aperture radius
d_s = sample thickness

Parasitic BGD around beam stop
Is typically 4x prediction.
... Another source ???

To Minimize background:
• Use larger sample aperture
• Use longer wavelength
To put raw corrected intensity into **absolute units**:

\[
\frac{d}{d} I_c(q_i) = \frac{I_c(q_i)}{J_B d_s T_s} = k_{sp} I_c(q_i)
\]

\( J_B \) = Beam current, \( d_s \) = sample thickness, \( T_s \) = transmission
\( \varepsilon_D \) = detector efficiency, \( \Delta \Omega \) = solid angle

**“Flat” Scattering BGD sources:**

\[
\frac{d}{d} G_D(q) G(q) \frac{T_s}{4d_s T_s}
\]

{ Inelastic scattering + Multiple Scattering Corrections }

G_D Correction depends upon the **detector**:

Three SANS detectors:
- BF_3 \rightarrow ILL (1970’s – 2000), Saclay
- \(^3\)He \rightarrow NIST, etc..  
- Scintillator \rightarrow FRM2 (KSW2), ISIS

**Inelastic** scattered neutrons have short
Wavelength: 1 Å to 2 Å
Quasi-Isotropic scattering: scattering is equally probable over all $4\pi$ solid angle. Valid for incoherent scattering.

1) Calculate for infinite slab, no absorption using Monte Carlo

2) Analytical Solution from Astronomy: {Chandrasekhar, 1950}

Transmitted:
\[
G (\theta) = \frac{A_G}{T(\tau)} \left[ Y(\tau)X(\theta) - X(\tau)Y(\theta) \right]
\]

Reflected:
\[
G (\theta) = \frac{A_G}{R(\tau)} \left[ X(\tau)X(\theta) - Y(\tau)Y(\theta) \right]
\]

Large Angle Transmission correction:
\[
T(\theta) = T_0 \frac{T_0^{a(\theta)}}{a(\theta) \ln(T_0)}
\]
\[
a(\theta) = \frac{1}{\cos(\theta)}
\]

\{ Incorporated into Igor data reduction \}
**Multiple Scattering** increases the measured cross section for 1 mm H$_2$O by 50% assuming quasi-isotropic scattering.

Inelastic scattering events are less likely to be detected:

$$G_D(\tau) = \frac{1}{D(\tau)} \int P_F(\tau_F) D(\tau_F) d\tau_F$$

$$P_F(\tau_F) = 2 f_i \frac{4}{\tau_F} \exp \left[ \frac{2}{\tau_F} \right] + (1 - f_i) P_{QE}$$
Gas Scattering
The total scattering cross-section from gasses can be estimated according to the formalism originally developed by Sachs & Teller (1941),

\[ s_{i,s} = P N_{av} \frac{N}{n} R T \sum_{i=1}^{N} s_i, s_i = s_i, b \frac{A e^{-x^2}}{A e^{-x^2} + 1} + \frac{1}{2} x e^{-x^2} \]

Ideal Gas law:
\[ s = \frac{P N_{av}}{R T} \prod_{i=1}^{N} X_i, s_{i,s} \]

Temperature & Wavelength Dependence:
\[ 2 = \frac{A e E_N}{k_b T} = \frac{A e k_E}{k_b T} \]

![Graph showing scattering cross-sections vs wavelength]

Lines assume quasi-isotropic scattering & \( G_D = G_0 = 1 \)
Scattering from Air: Is not flat at large wavelengths !!!!
For window scattering, thickness $d_{s,b}$, Detector distance $L_{2,b}$

$$\frac{d^e}{d} (q) = C_f \frac{d}{d} (q_e)$$

$$C_f = d_{s,b} \frac{\left( \cos \left( \frac{b}{d} \right) \right)^3}{\cos \left( \frac{b}{d} \right)} \approx d_{s,b} \left( \frac{L_2}{L_{2,b}} \right)^2$$

$$q_e = q \frac{\sin \left( \frac{b}{d} \right)}{\sin \left( \frac{b}{d} \right)} \approx q \frac{L_2}{L_{2,b}}$$

Background scales as:
1) Ratio of window to sample thickness: $d_{s,b} / d_s$
2) Ratio of sample-to-detector distances: $(L_2 / L_{2,b})^2$
3) Ignores transmission corrections, see:

Residual air in Detector vacuum tank:
Air scattering near detector is enhanced via larger solid angle

$$C_F = \frac{L_2^2}{d_s} \frac{L_2}{\tan^1 \left( \frac{R}{x} \right)} \frac{\cos \tan^{-1} \left( \frac{R}{x} \right)^3}{x^2} dx$$

Note that enhancement factor $C_F$ can be very large when integrated over the entire Length of detector vessel. $C_F > 10^4$

Dashed lines: beamstop located at detection plane:
$X_{BS} = 0$
Solid line: beamstop in front of dome: (shadowed)
$X_{BS} > 26$ cm
Detector dome scattering:
20 cm of Helium, P = 2 bar
0.48 cm of aluminum

Figure 3.9. Measured scattering (No beam stop) from dome of the detector, and calculated contributions from helium and aluminum window.
Porod Scatterer: Spherical glass beads

\[ I(q) = Aq^{-4} \]

\[ d_s = 2 \text{ mm}, 35 \mu \text{m diameter} \]

Scattering should be horizontal line:
Scattering from PMMA (Plexiglas) 1.35 mm thick

Total cross-section → Transmission

Forward cross section (5° to 20°)

- Increase with wavelength dominated by inelastic scattering
- Weak temperature dependence
- Forward scattering much stronger than isotropic approximation
Scattering from H$_2$O 1 mm thick

Total cross-section $\rightarrow$ Transmission

Forward cross section (5° to 20°)

- Increase with wavelength dominated by inelastic scattering
- Strong temperature dependence
- Forward scattering much stronger than isotropic approximation and higher than at other facilities (higher efficiency detector ??)
Scattering from D$_2$O 4 mm thick
Total cross-section $\rightarrow$ Transmission
Forward cross section (5° to 20°)

- Increase with wavelength dominated by inelastic scattering
- Strong temperature dependence
- Forward scattering much stronger than isotropic approximation and higher than at other facilities (higher efficiency detector ??)
Steyerl (1977) has given an expression for the calculation of the temperature-dependent single phonon scattering as a function of temperature \( T \) as

\[
\phi_{ph}(l, T) = \frac{C_{ph}}{M_e} \frac{1/2}{D} R(x) \\
\text{bat} = m N_A \frac{1}{N} x_i \frac{1}{M_i}
\]

\[
\sigma_{tot}(l, T) = \sigma_{abs}(l) + f_{sp} \phi_{ph}(l, T)
\]

Fit \( f_{sp} \) depends upon choice for Debye Temperature \( \Theta_D \)
Assume Phonon scattering is quasi-isotropic:

\[
\frac{d}{d} (0) = g_{sph} \left( \frac{sph + inc}{4} \right)
\]
Measured “Flat Background from 5° to 20° for all materials: $T = 298$ K, $\lambda = 10$ Å

Scattering by thickness:
- Strongest $\rightarrow$ H₂O, D₂O
- Weakest $\rightarrow$ Ar, He gas (1 bar)

Scattering per atom:
- Strongest $\rightarrow$ H₂O, Air
- Weakest $\rightarrow$ Sapphire, Silicon
TOF measurements with disk chopper to separate inelastic from quasi-elastic scattering:
Flat Background from 1 mm thick D$_2$O Sample:

<table>
<thead>
<tr>
<th>Material</th>
<th>Thickness</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air</td>
<td>150 mm</td>
</tr>
<tr>
<td>aQuartz</td>
<td>3.2 mm</td>
</tr>
<tr>
<td>Sapphire</td>
<td>3.2 mm</td>
</tr>
<tr>
<td>Silicon</td>
<td>6.4 mm</td>
</tr>
<tr>
<td>Vacuum (0.1 Torr)</td>
<td>4000 mm</td>
</tr>
</tbody>
</table>

For **low signal** Experiments, S/N can be increased by factor of three if:
1) Use TOF to remove inelastic scattering
2) Use vacuum, He or Ar in sample chamber
3) Replace aQuartz with Sapphire windows
Conclusions

- **Detector performance** critical for subtracting weak signal from strong Bgd.
- **Metal alloy** windows should not be used due to strong Bgd from precipitates.
- **Wavelength** should be chosen long enough to eliminate Double Bragg Scattering.
- **Wavelength** should be chosen short enough to minimize inelastic scattering.
- TOF can be used to eliminate inelastic Bgd.
- Single crystal windows preferred over amorphous quartz over metal alloy.
- Any air in path produces significant Bgd:
  - minimize path length or replace with Ar gas or evacuate
- Vacuum in Detector tank must be $P < 0.1$ Torr
- Parasitic halo around beamstop has slit diffraction component: $I \sim q^{-3}$
  { Major concern for VSANS instrument }
- Scattering in Detector Dome can produce an overlap problem {Lens config.}

Future Work:

Resolve discrepancy in Bgd measured at different facilities