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: H_2O, D_2O, 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 << N**

Russell et al, 1995 170 nm thick sample

Macromolecules, Vol. 28, No. 3, 1995

Rubinson et al, 2008
0.5% Protein in D_2O

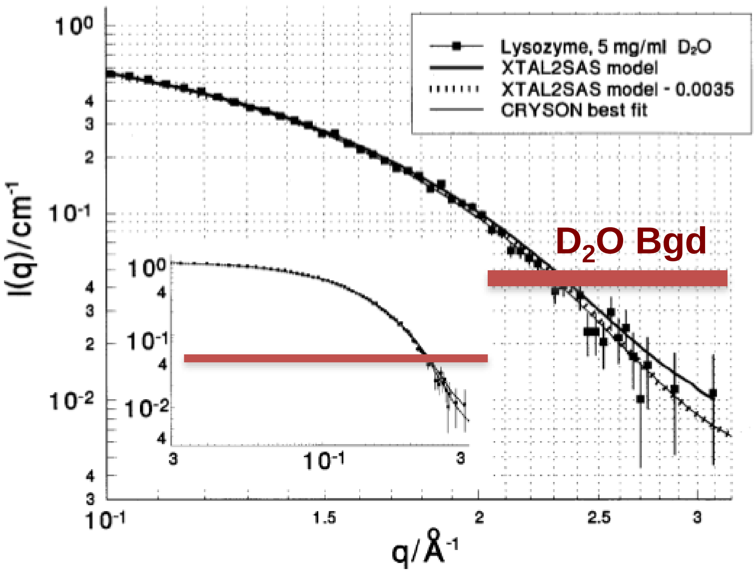


Figure 6
SANS data from 5 mg ml^{-1} lysozyme in D_2O buffer and the resulting model SANS curves from *CRYSON* and *XTAL2SAS* using the structure from the PDB-listed parameters of 6lyz. The *CRYSON* curve represents the best fit to the SANS data assuming a 3 Å bound D_2O layer (fit parameters shown in 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 $q < 0.2$ Å^{-1} are smaller than the data points. The inset shows the SANS curve in the full measured q range.

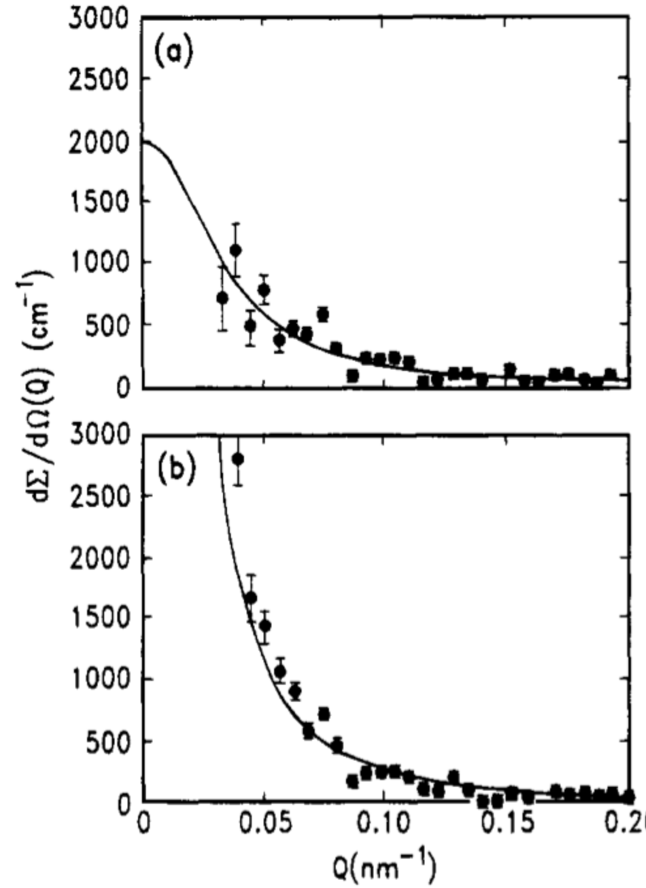


Figure 3. $d\Sigma/d\Omega$ as a function of Q for a mixture of PS-760K and dPS-690K (a) as-cast from a toluene solution and (b) heated to 130 °C for 15 h. The sample thickness is 170 nm. The solid lines are fits the scattering profiles using a Debye function.

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

$$c_t \cong \frac{c_o}{1 - c_o t_d}$$

t_d = detector dead time
c = count rate, o → observed, t → true

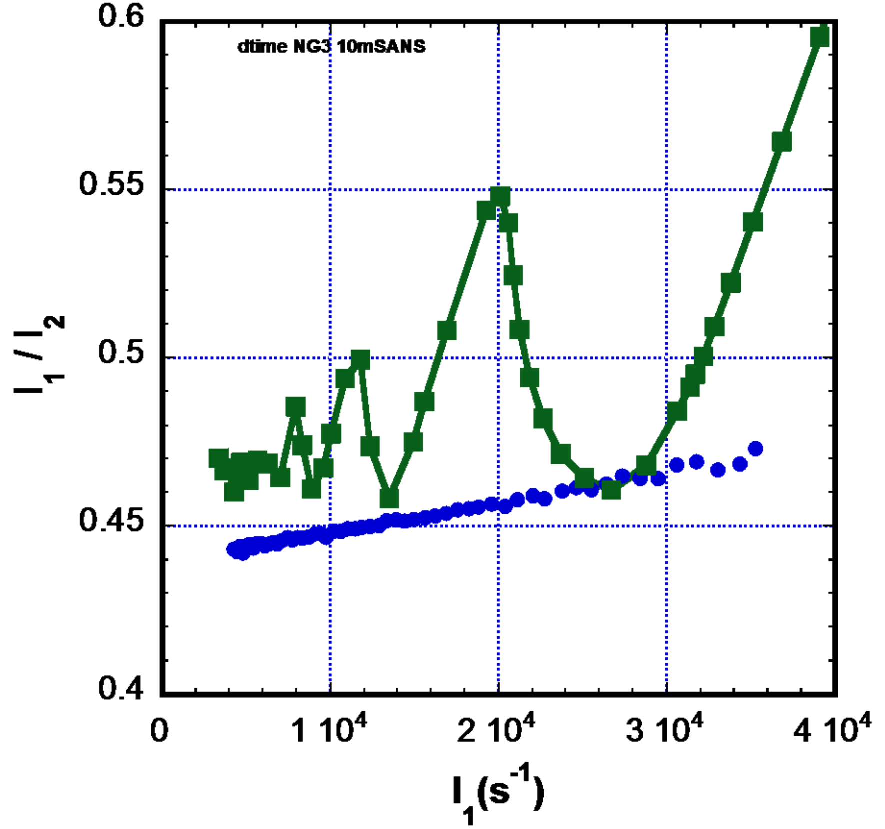
Correction incorporated in IGOR data reduction software.

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

$$R_o = R_t + t_d (1 - R_t) c_{so}$$

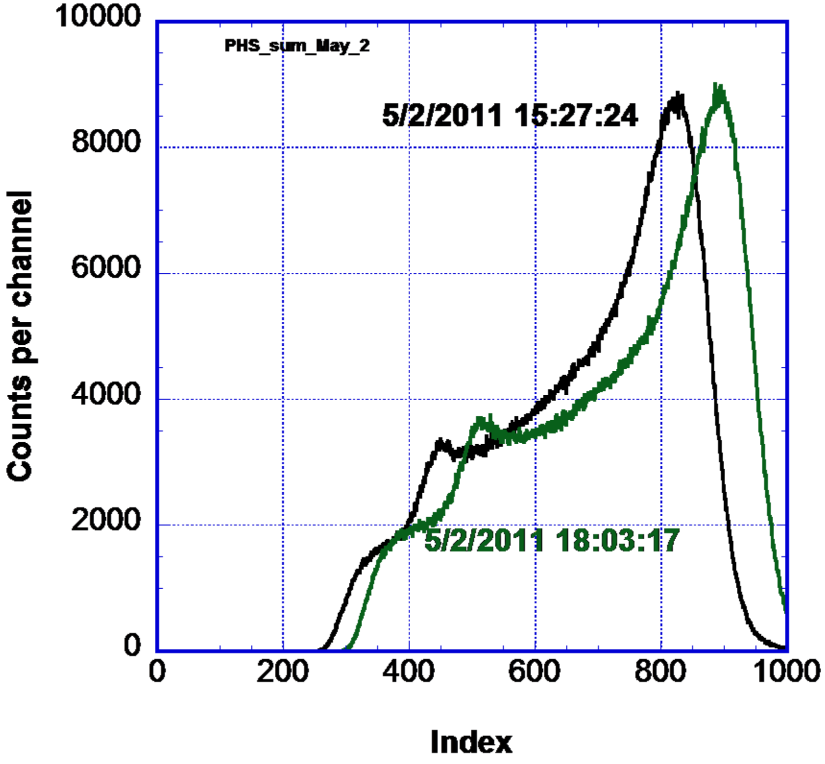
All three SANS Ordela detectors exhibit oscillating detector efficiency dependence on count rate, which is **NOT** corrected by data reduction.
→ Background is not completely subtracted: $f_E * I_E + f_B * I_B$

Dead time measured on SANS Ordela 2D detectors



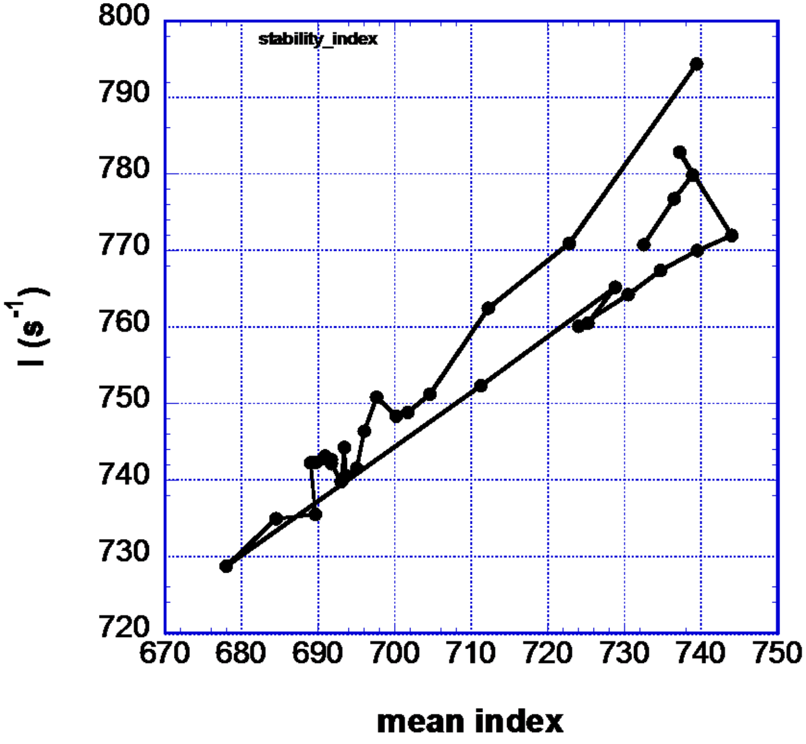
Detector Efficiency can also change with **time**:
Measured **VSANS tube detectors** efficiency once an hour over two days.

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



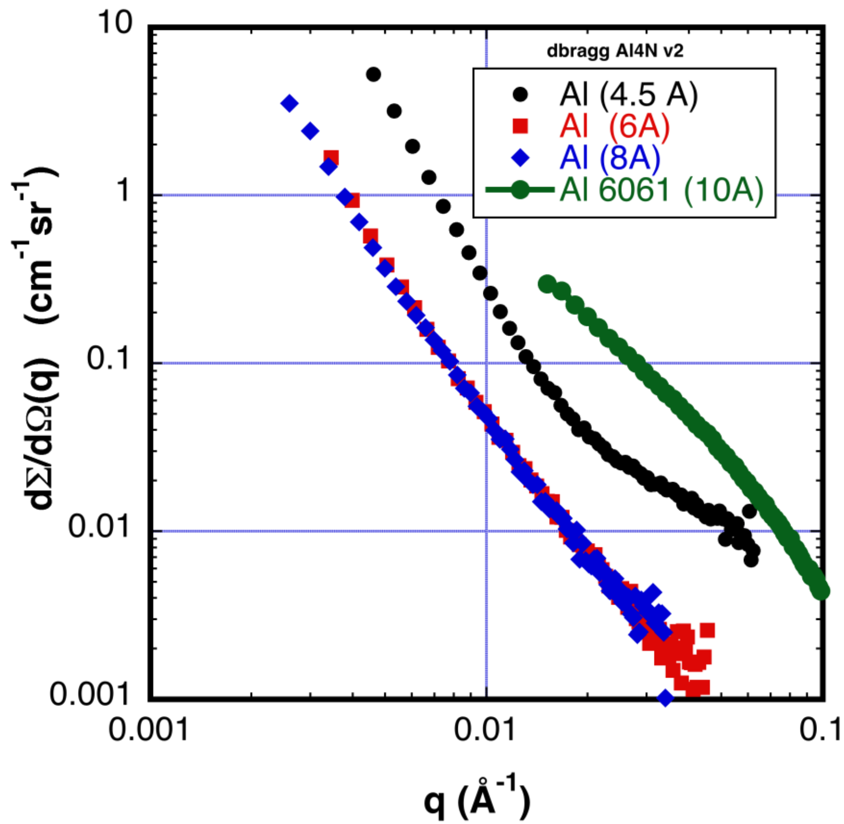
Pulse Height Spectrum for one-hour runs made 2.5 hours apart.
Observed count rates are 729 s^{-1} for first and 752 s^{-1} for second run.

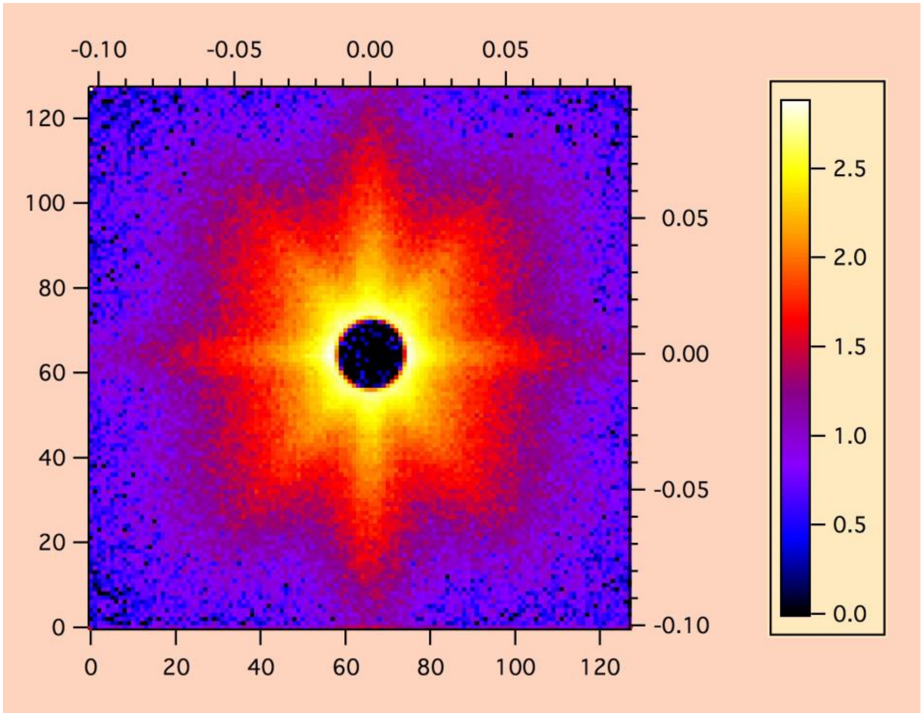
Ordela 2D Detectors are stable to 0.1% day to day.



Double Bragg Scattering:
Produces scattering which mimics structural SANS
Eliminated if wavelength chosen larger than $2d_{max}$

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

$$\frac{d\Sigma_F}{d\Omega}(q) = \frac{4\pi R_2^2}{\lambda_0^2}\left[\frac{J_1(qR_2)}{qR_2}\right]^2 \qquad \frac{d\Sigma}{d\Omega}(q) \cong \frac{4}{R_2 d_s \lambda_0^2} q^{-3}$$

R_2 = aperture radius

d_s = sample thickness

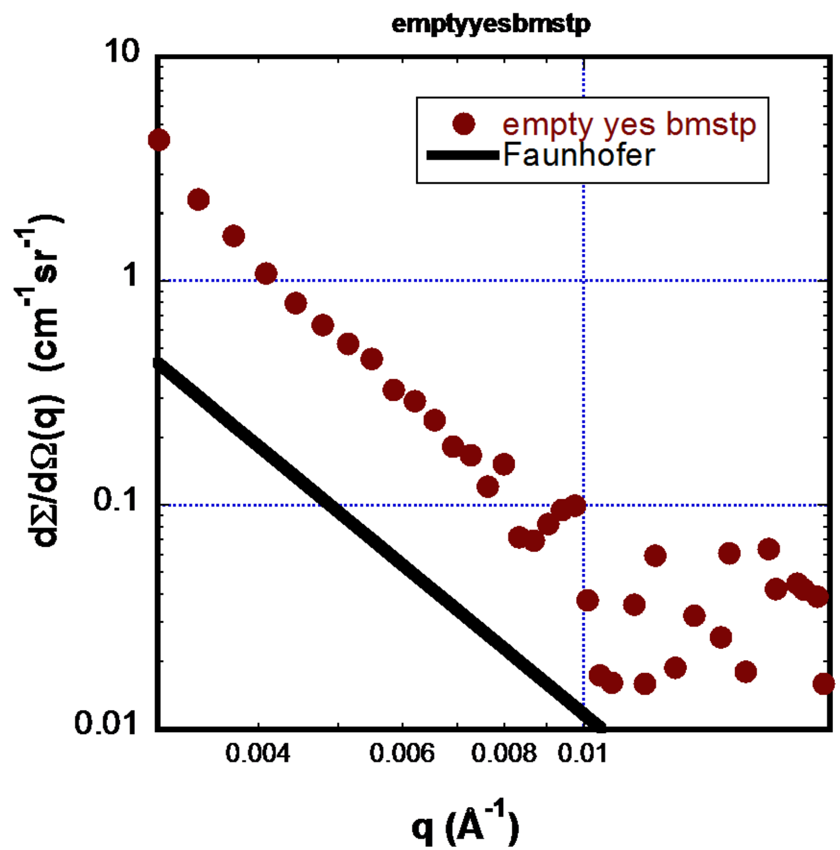
Parasitic BGD around beam stop

Is typically 4x prediction.

… Another source ???

To Minimize background:

- **Use larger sample aperture**
- **Use longer wavelength**



To put raw corrected intensity into **absolute units**:

$$\frac{d\Sigma}{d\Omega}(q_i) = \frac{I_c(q_i)}{\varepsilon_D(\lambda) J_B d_s T_s \Delta\Omega_i} = k_{sp} I_c(q_i)$$

J_B = Beam current, d_S = sample thickness, T_S = transmission

ε_D = detector efficiency, $\Delta\Omega$ = solid angle

G_D Correction depends upon the **detector**:

Three SANS detectors:

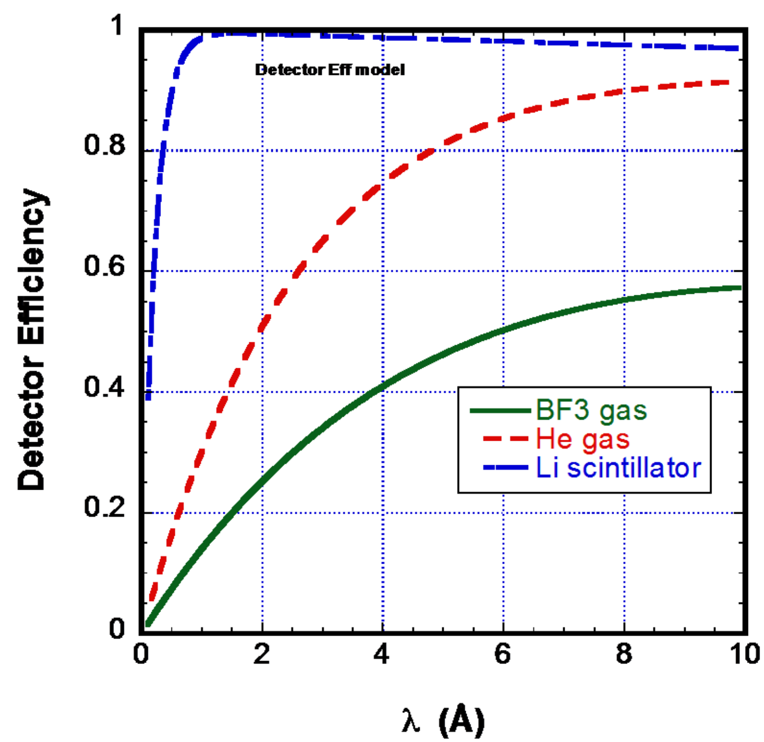
BF_3 → ILL (1970's – 2000), Saclay

^3He → NIST, etc..

Scintillator → FRM2 (KSW2), ISIS

Inelastic scattered neutrons have short

Wavelength: 1 Å to 2 Å



"Flat" Scattering BGD sources:

$$\frac{d\Sigma}{d\Omega}(q) \cong G_D(\lambda) G_q(q) \frac{(1 - T_s)}{4\pi d_s T_s}$$

{ Inelastic scattering + Multiple Scattering Corrections }

Quasi-Isotropic scattering: scattering is equally probable over all 4π solid angle. Valid for incoherent scattering.

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

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

Transmitted:

$$G_q(q) = \frac{A_G m_0}{m_T(m_T - m_0)}\left[Y(m_T)X(m_0) - X(m_T)Y(m_0)\right]$$

Reflected:

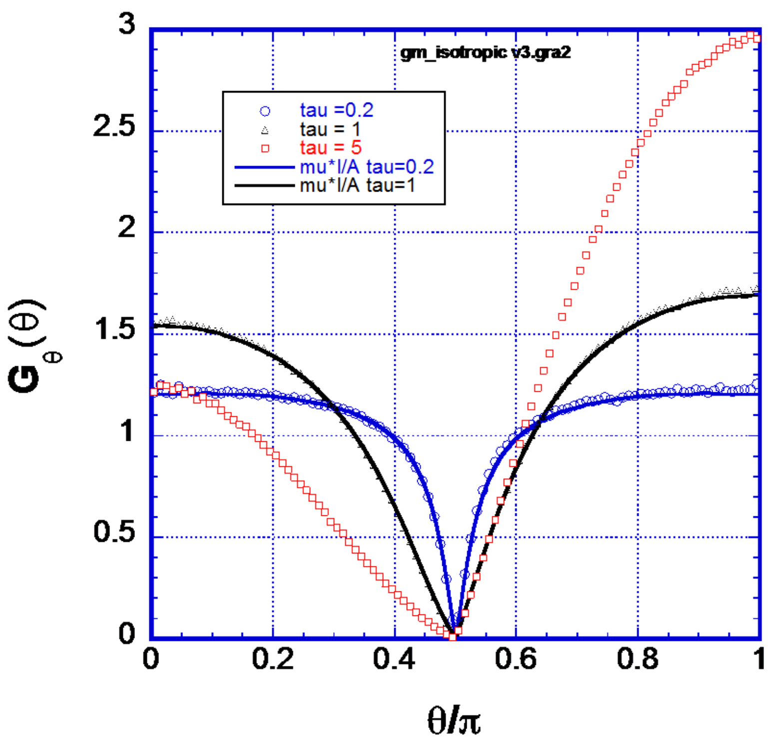
$$G_q(q) = \frac{A_G m_0}{m_R(m_R + m_0)}\left[X(m_R)X(m_0) - Y(m_R)Y(m_0)\right]$$

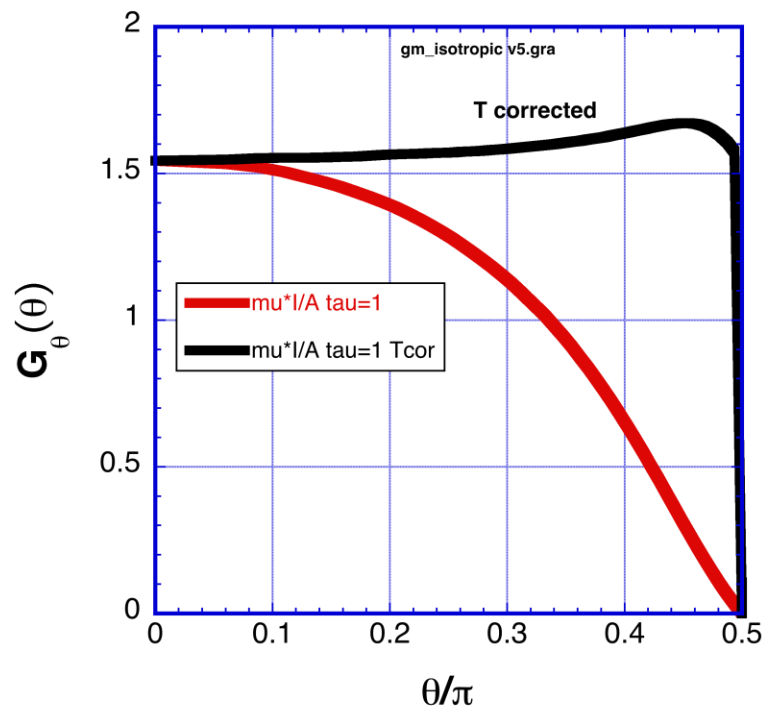
Large Angle Transmission correction:

$$T(q) = T_0 \frac{T_0^{a(q)} - 1}{a(q)\ln(T_0)}$$

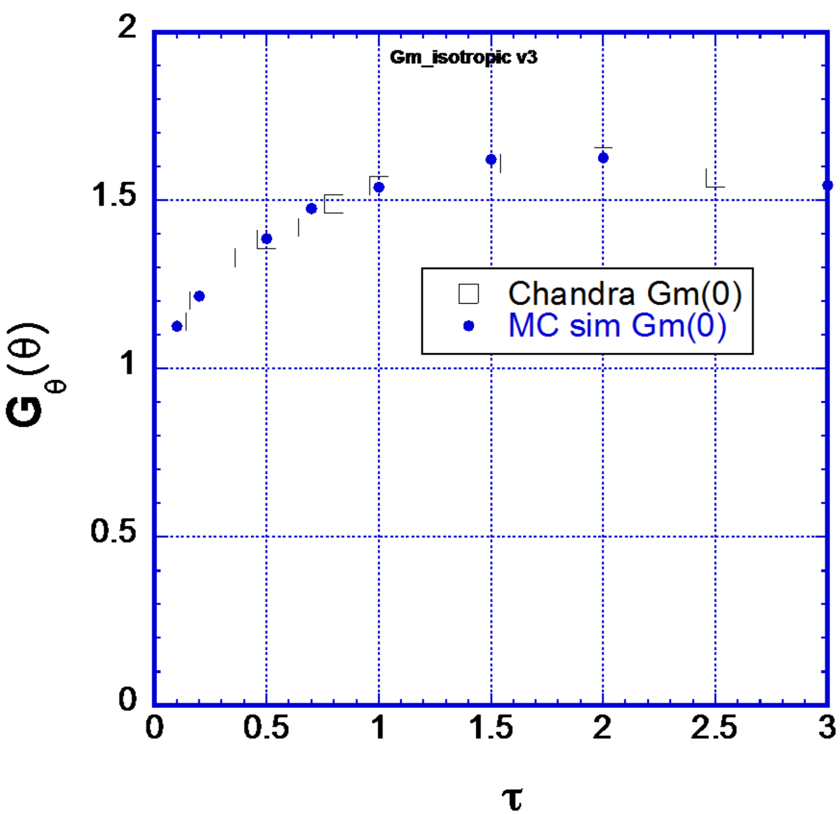
$$a(q) = \frac{1}{\cos(q)} - 1$$

{ Incorporated into Igor data reduction }





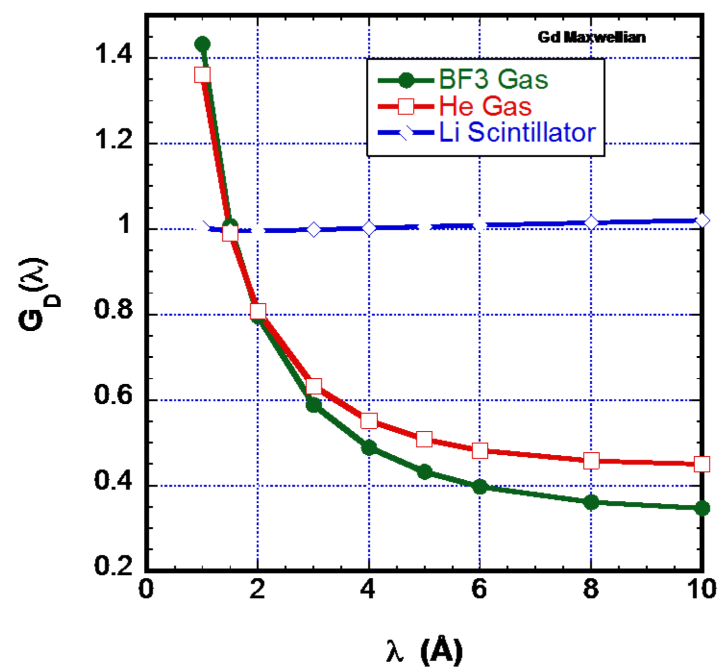
Multiple Scattering increases the measured Cross section for 1 mm H_2O by 50 % assuming Quasi-isotropic scattering.



Inelastic scattering events are less likely to be detected:

$$G_D(\lambda) = \frac{1}{\varepsilon_D(\lambda)} \int P_{\lambda_F}(\lambda_F)\,\varepsilon_D(\lambda_F)\,d\lambda_F$$

$$P_{\lambda_F}(\lambda_F) = 2 f_i \frac{\lambda_T^4}{\lambda_F^5} \exp\left(-\lambda_T^2 / \lambda_F^2\right) + (1 - f_i) P_{QE}$$



Gas Scattering

The total scattering cross-section from gasses can be estimated according to the formalism originally developed by Sachs & Teller (1941),

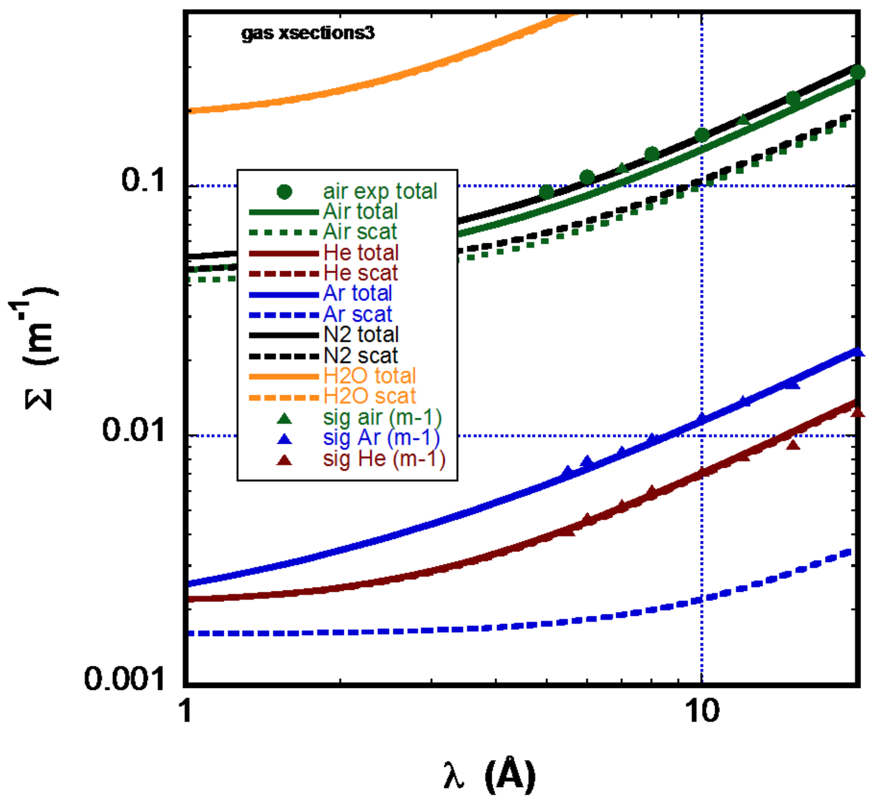
$$\sigma_{i,s} = \sigma_{i,b}\left(\frac{A_e}{A_e+1}\right)^2\left[\left(1+\frac{1}{2x^2}\right)erf(x)+\frac{1}{x\sqrt{\pi}}e^{-x^2}\right]$$

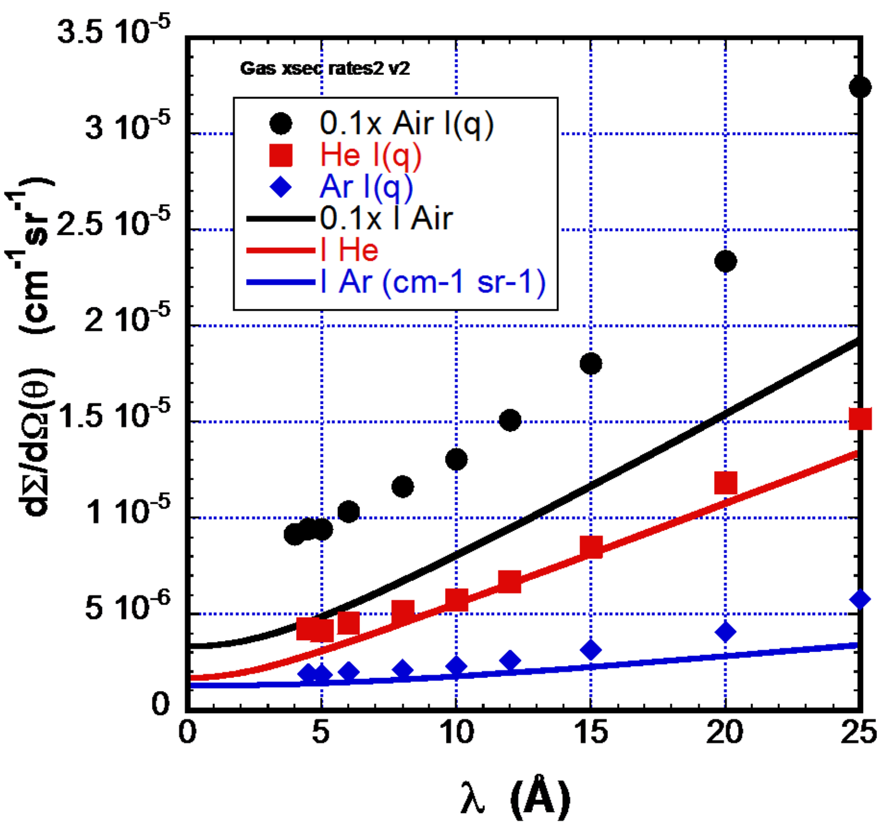
Ideal Gas law:

$$\Sigma_s = \frac{PN_{av}}{RT}\sum_{i=1}^{N} X_i \sigma_{i,s}$$

Temperature & Wavelength Dependence:

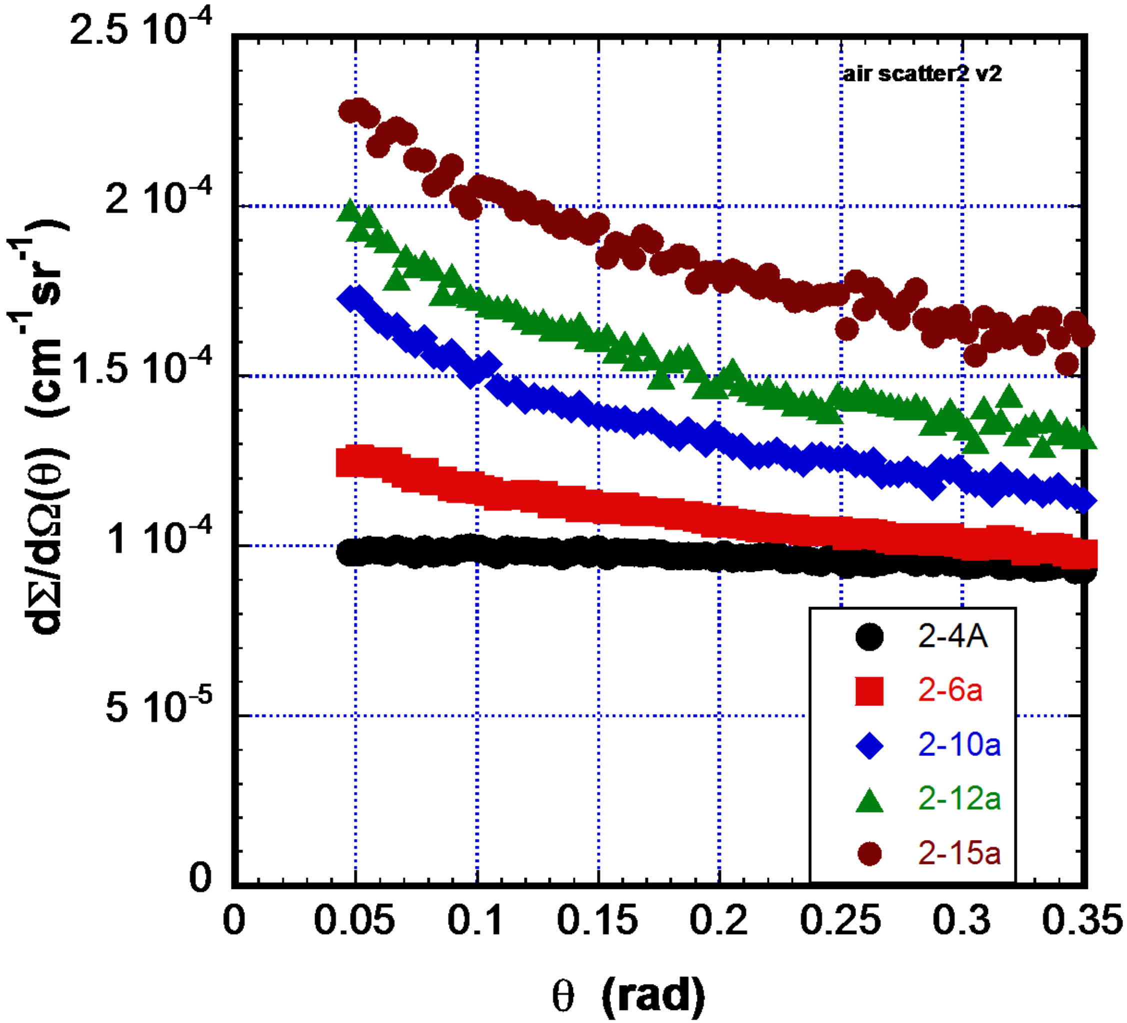
$$x^2 = \frac{A_e E_N}{k_b T} = \frac{A_e k_{E\lambda}\lambda^2}{k_b T}$$





Lines assume quasi-isotropic scattering & $G_D = G_\theta = 1$

Scattering from Air: Is not flat at large wavelengths !!!



For **window scattering**,

thickness $d_{s,b}$, Detector distance $L_{2,b}$

$$\frac{d\Sigma_e}{d\Omega}(q) = C_f \frac{d\Sigma}{d\Omega}(q_e)$$

$$C_f = \frac{d_{s,b}}{d_s}\frac{\Delta\Omega_b}{\Delta\Omega}\left(\frac{Cos(\theta_b)}{Cos(\theta\ )}\right)^3 \approx \frac{d_{s,b}}{d_s}\left(\frac{L_2}{L_{2,b}}\right)^2$$

$$q_e = q\frac{\sin(\theta_b/2)}{\sin(\theta\ /2)} \approx q\frac{L_2}{L_{2,b}}$$

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:

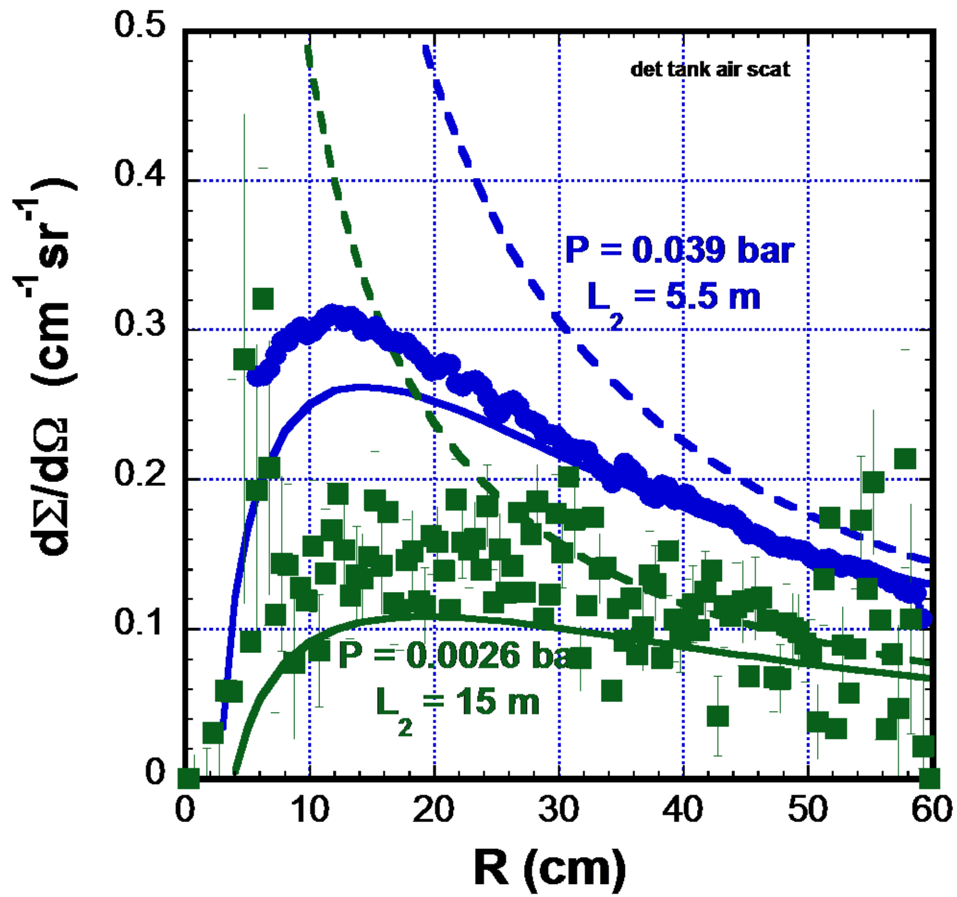
A. Brulet, et al (2007) *J. Appl. Cryst.* **40**, 165-177

Residual air in Detector vacuum tank:

Air scattering near detector is enhanced via larger solid angle

$$C_F = \frac{L_2^2}{d_s}\int_{L_{bs}+X_{bs}}^{L_2} \frac{\cos\left[\tan^{-1}(R/x)\right]^3}{x^2}dx$$

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



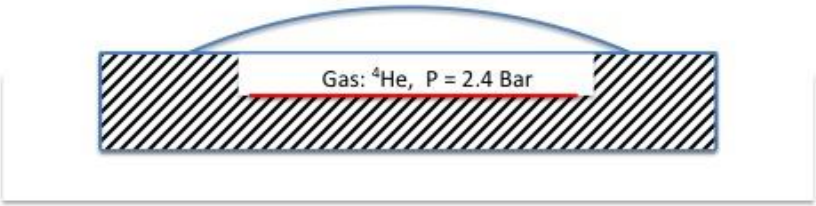
Dashed lines: beamstop located at detection plane:

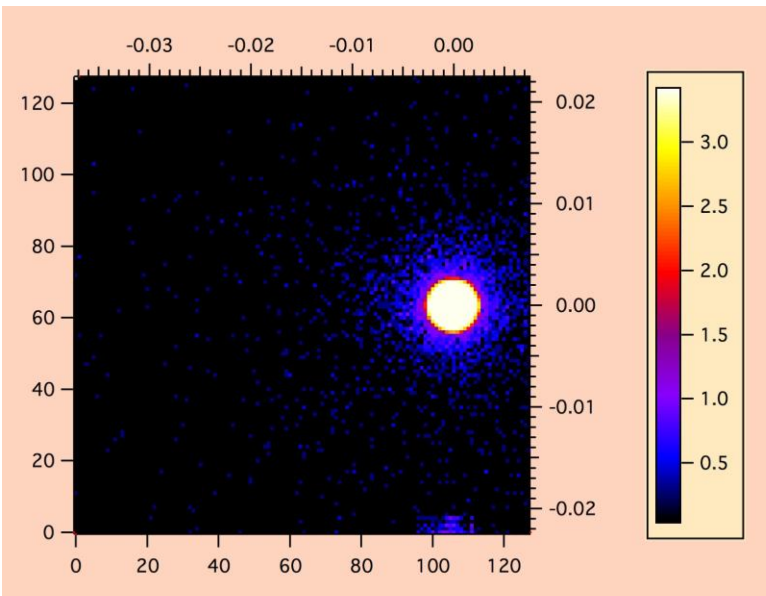
$X_{BS} = 0$

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





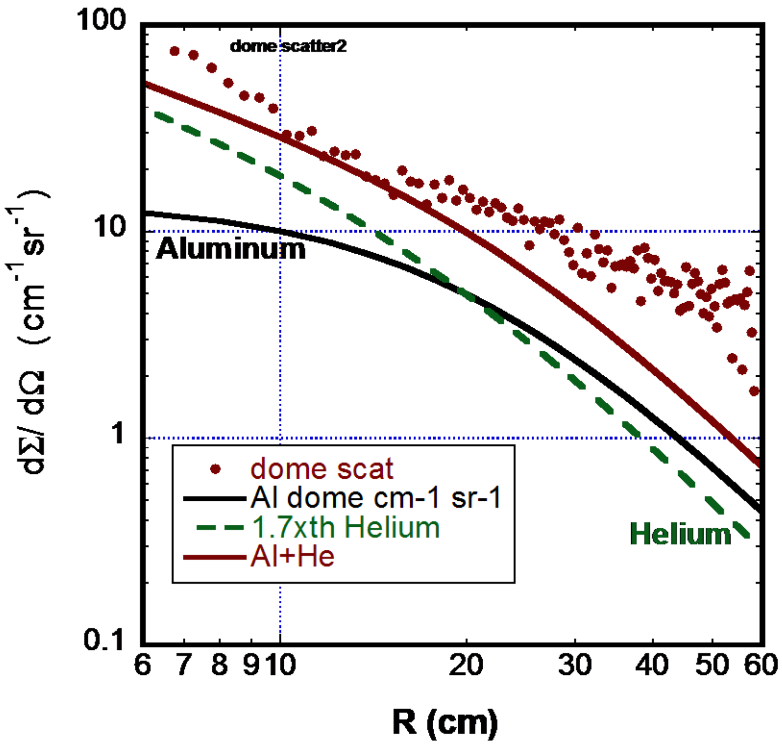


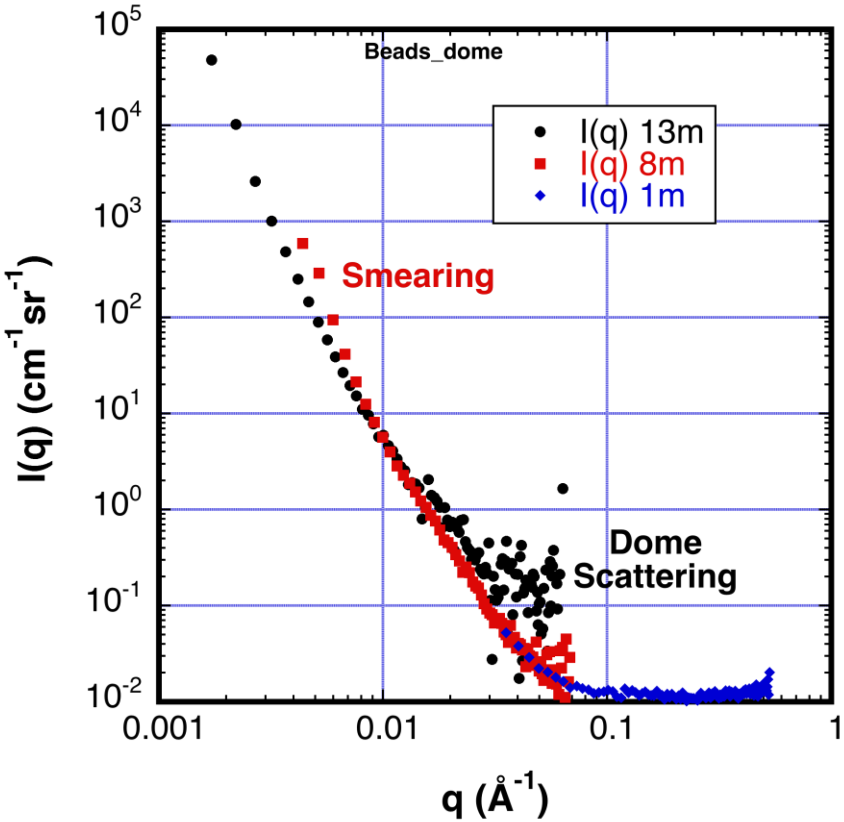
Figure 3.9. Measured scattering (No beam stop) from dome of the detector, and calculated contributions from helium and aluminum window.

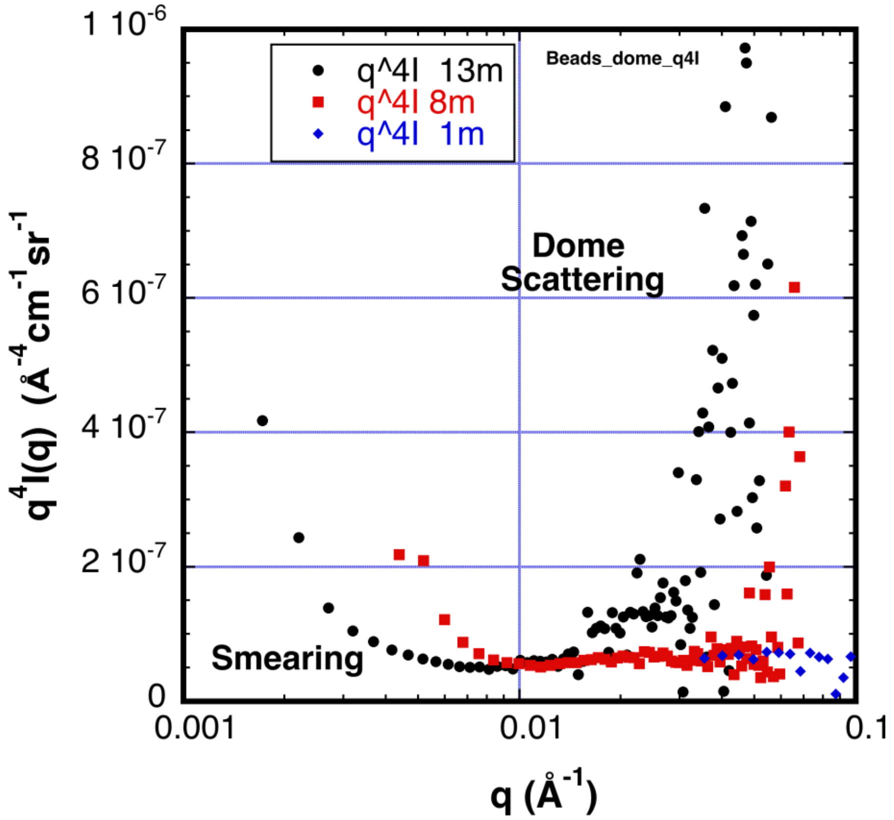
Porod Scatterer: Spherical glass beads

$I(q) = Aq^{-4}$

d_s = 2 mm, 35 μm diameter

Scattering should be horizontal line:



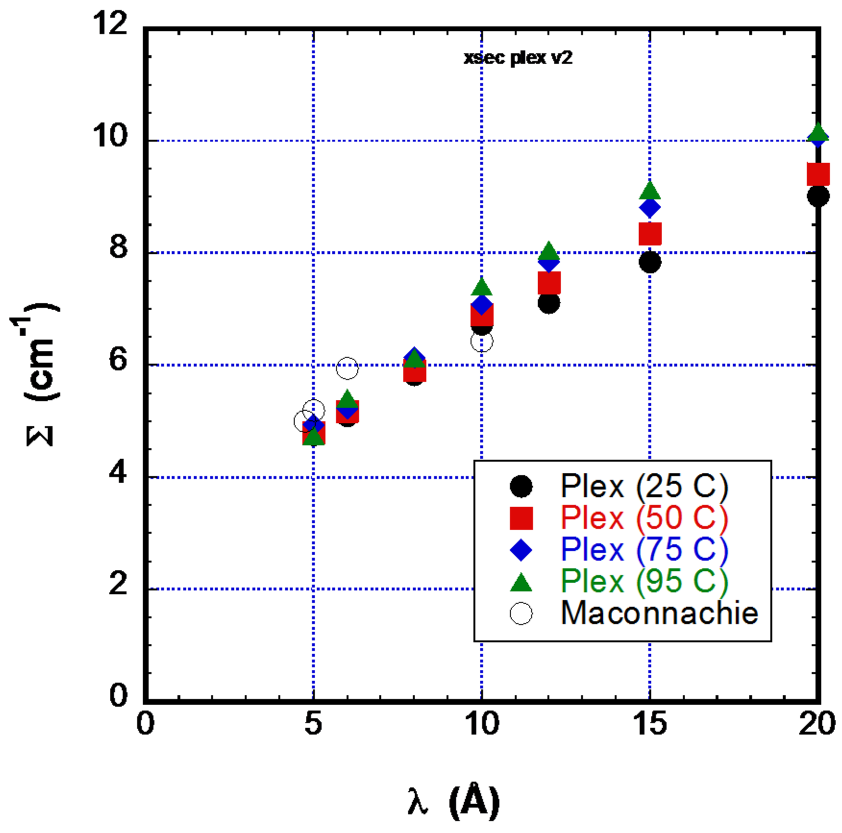


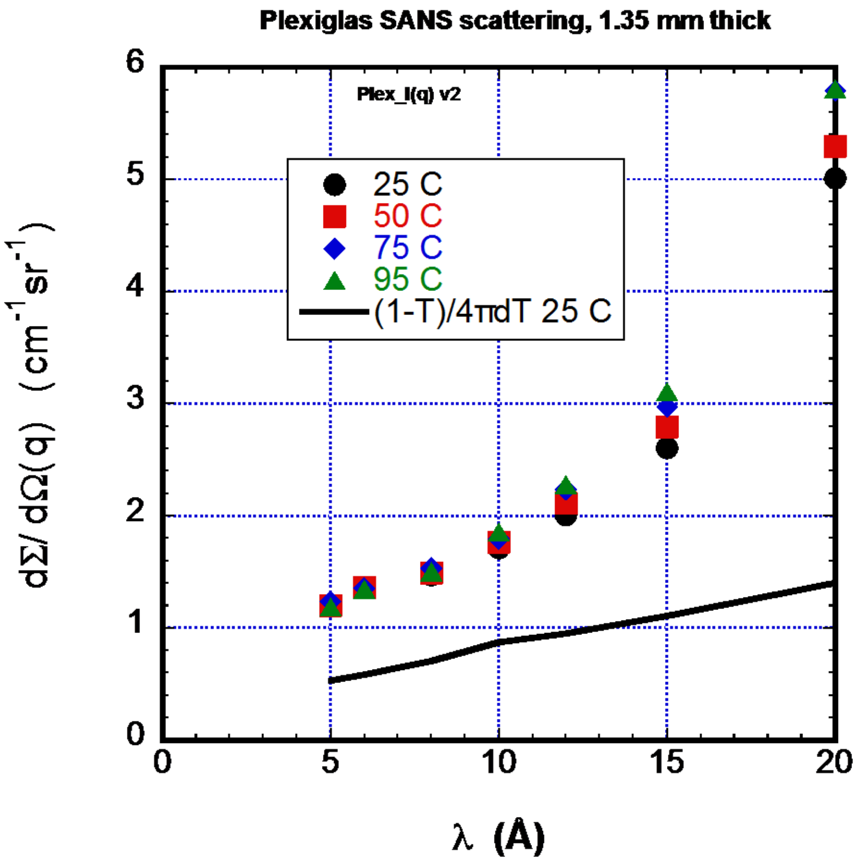
Scattering from PMMA (Plexiglas) 1.35 mm thick

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



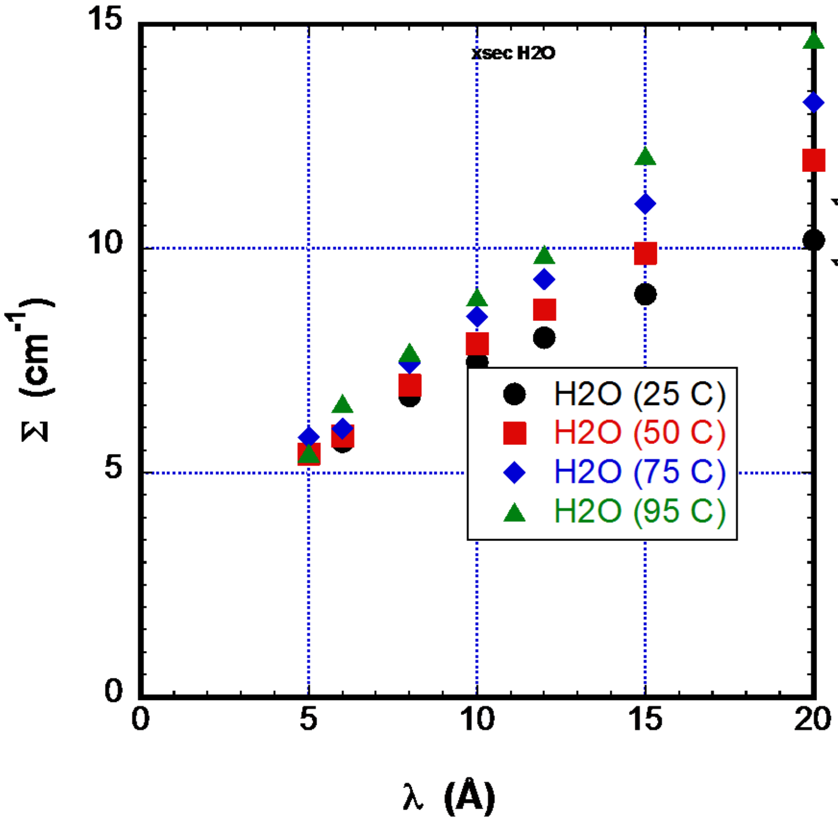


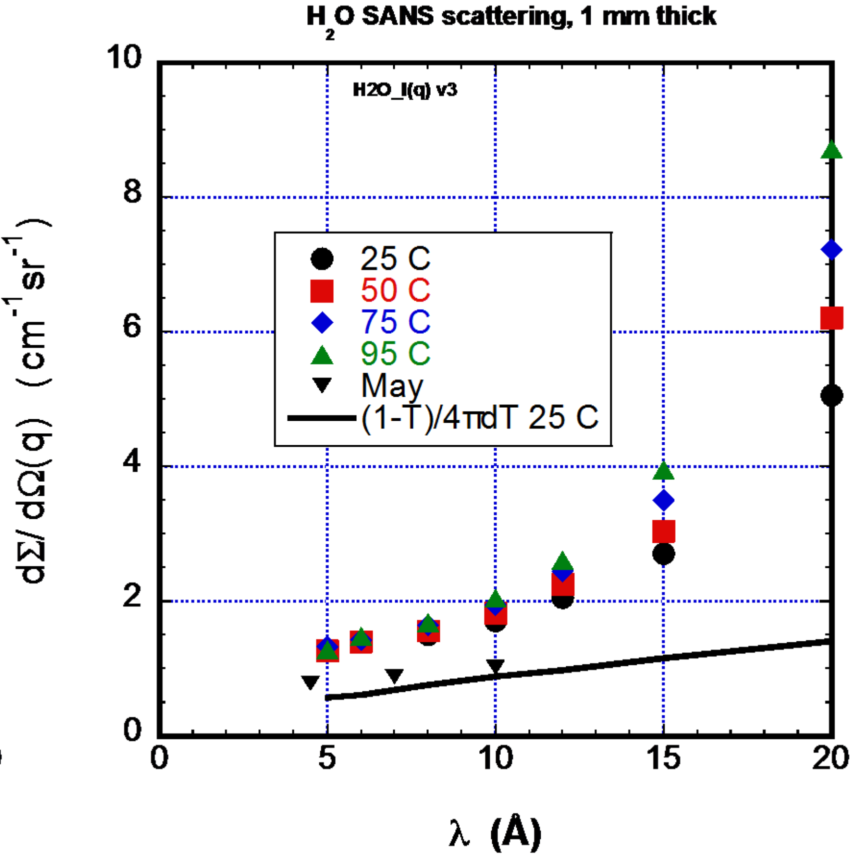
Scattering from H_2O 1 mm thick

Total cross-section → 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_2O 4 mm thick

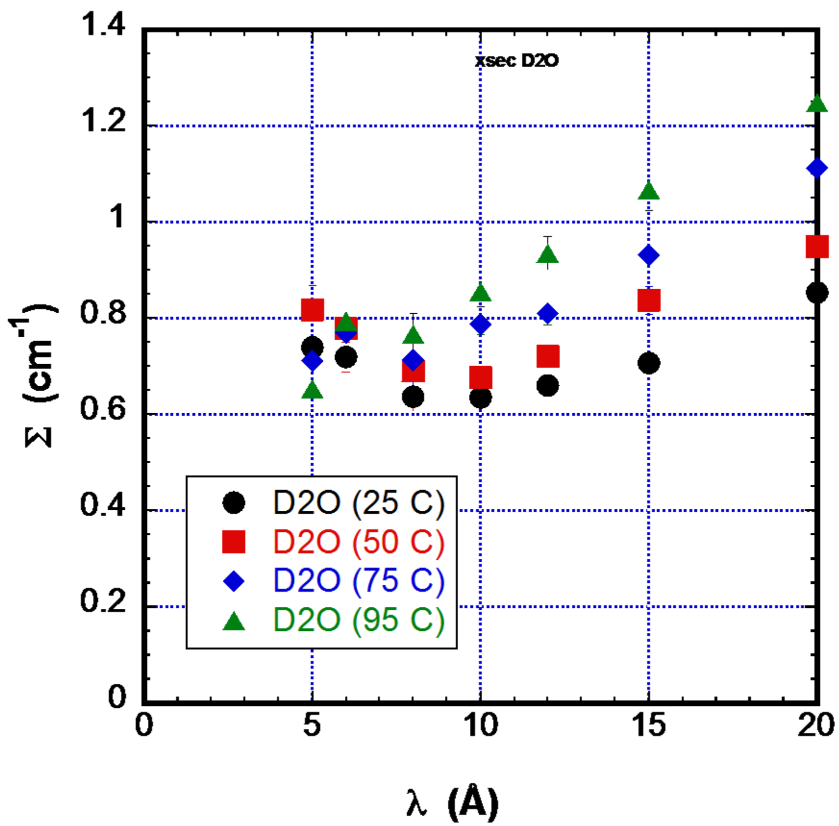
Total cross-section → Transmission

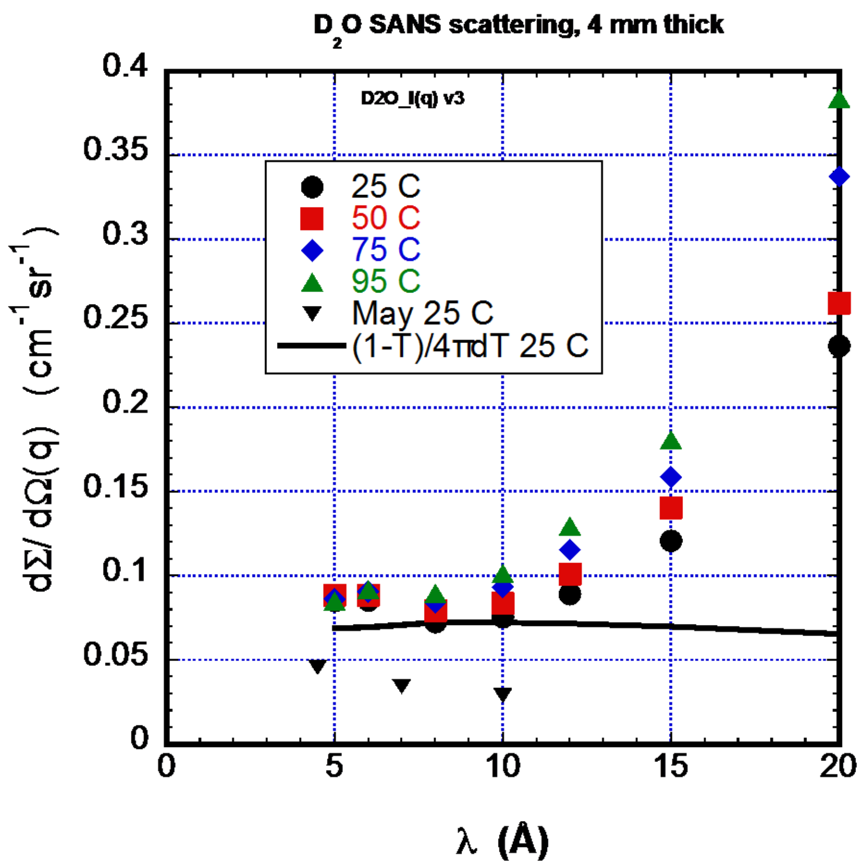
Forward cross section (5˚ to 20˚)

• 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 ??)



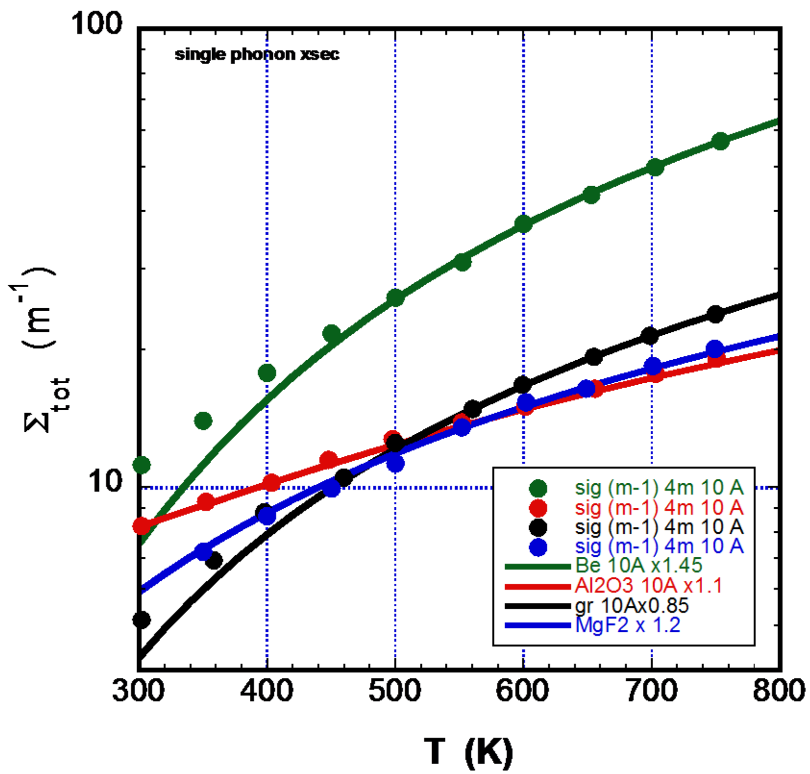


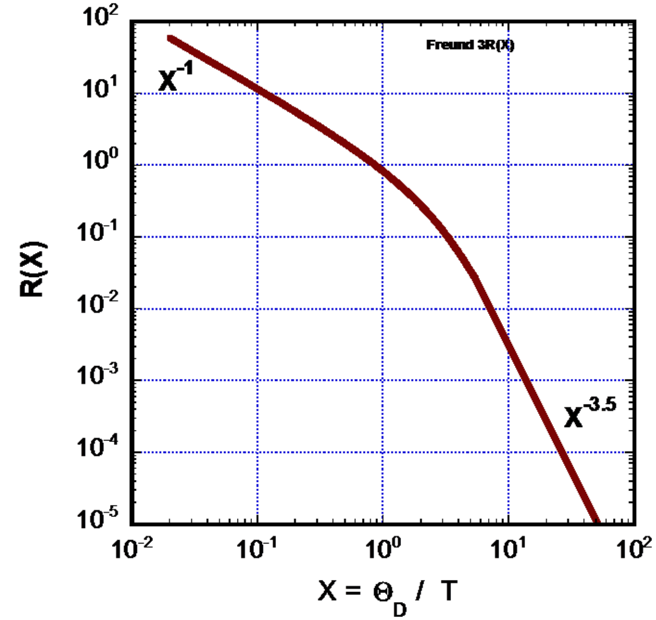
Phonon Scattering from Solids

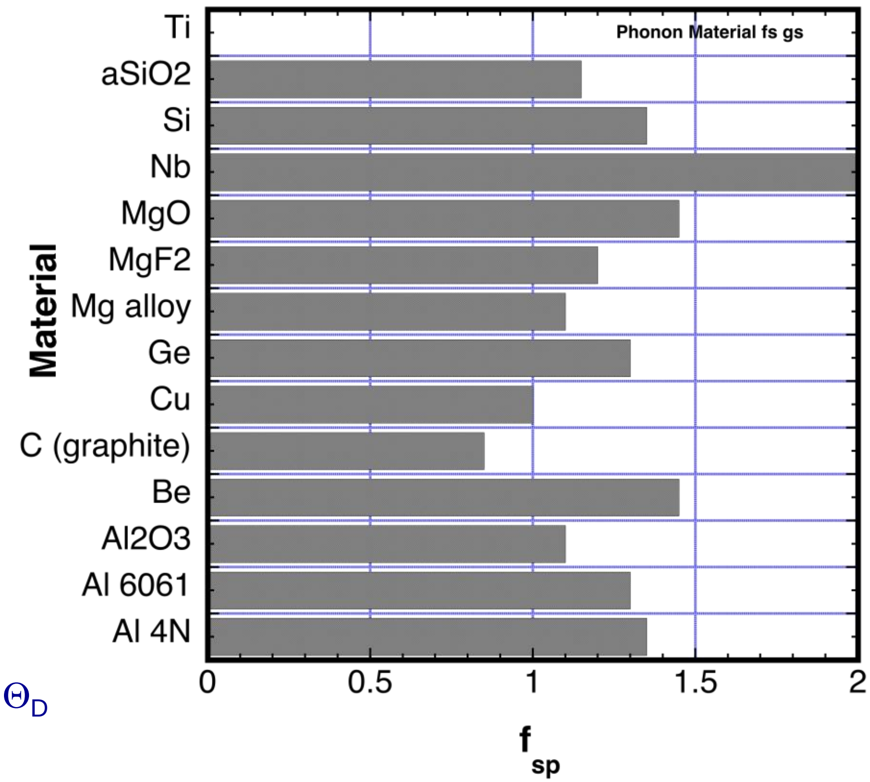
Steyerl (1977) has given an expression for the calculation of the temperature-dependent single phonon scattering as a function of temperature T as

$$\Sigma_{sph}(\lambda, T) = \frac{C_{sph}\Sigma_{bat}\Theta_D^{1/2}\lambda}{M_e} R(x) \qquad \Sigma_{bat} = \rho_m N_A \frac{\sum_1^N x_i \sigma_{s,i}}{\sum_1^N x_i M_i}$$

$$\Sigma_{tot}(\lambda, T) = \Sigma_{abs}(\lambda) + f_{sph}\Sigma_{sph}(\lambda, T)$$



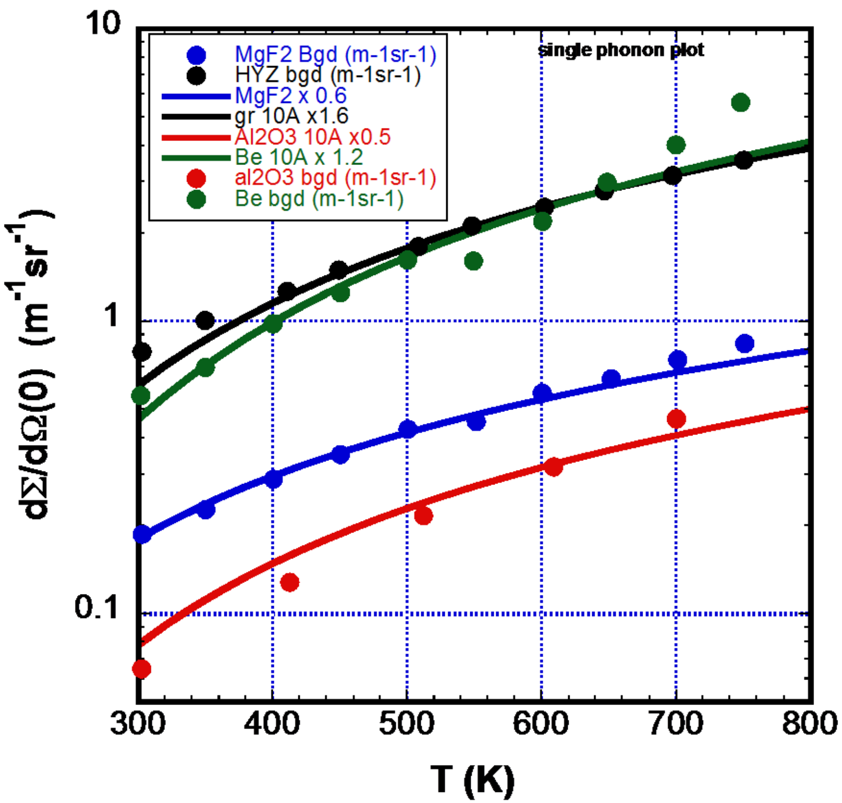


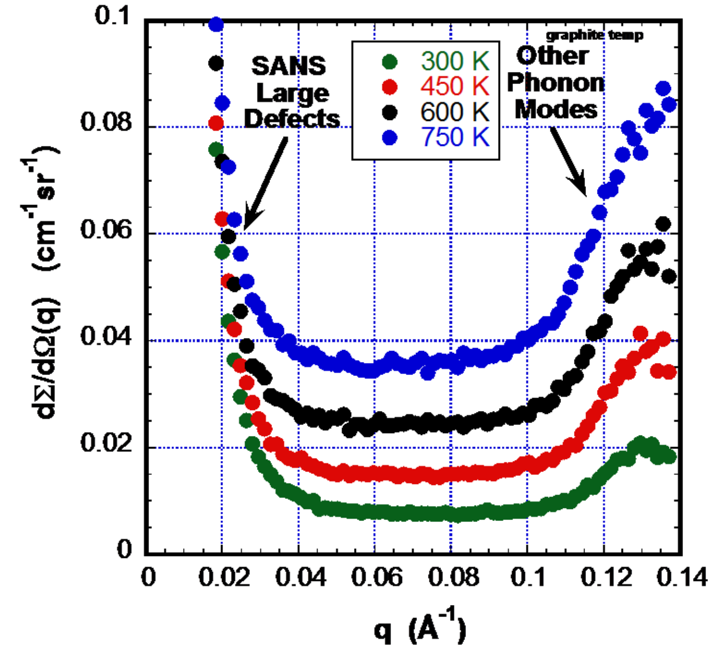


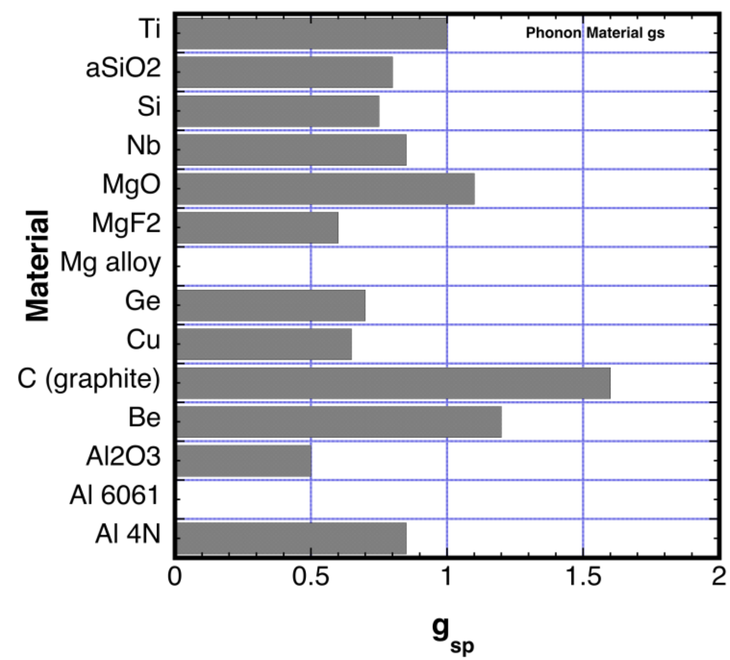
Fit f_{sp} depends upon choice for Debye Temperature Θ_D

Assume Phonon scattering is quasi-isotropic:

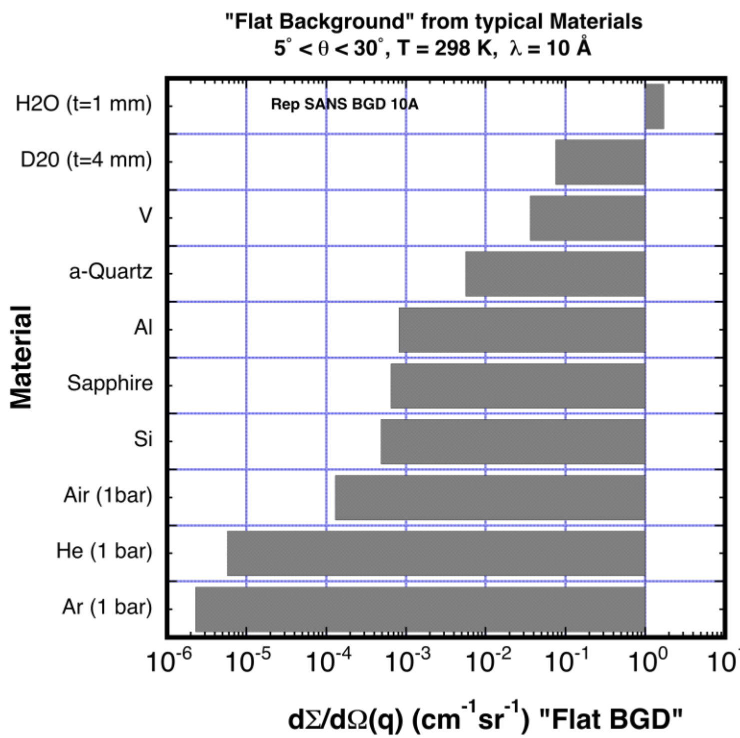
$$\frac{d\Sigma}{d\Omega}(0) = g_{sph}\,\frac{(\Sigma_{sph} + \Sigma_{inc})}{4\pi}$$

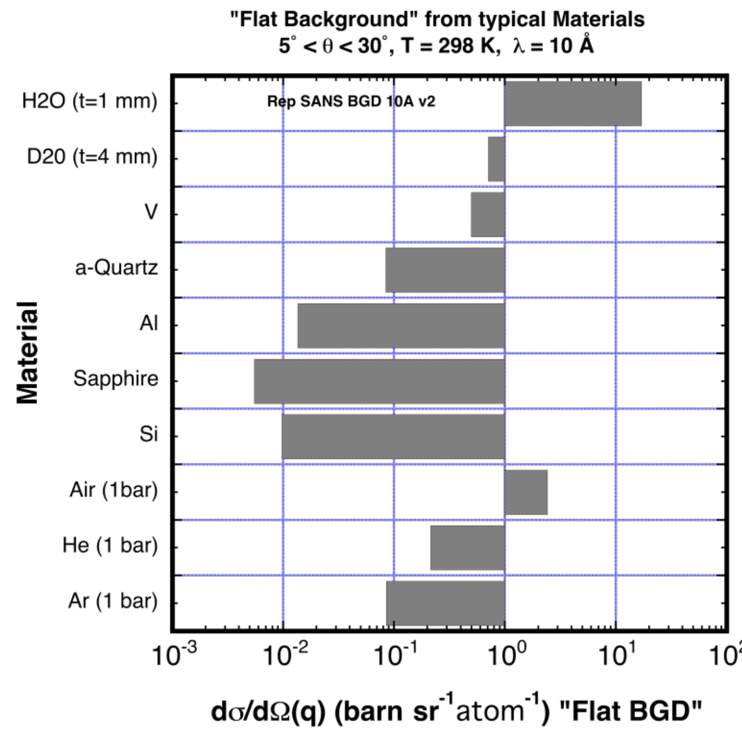






Measured “Flat Background from 5˚ to 20˚ for all materials: T =298 K, λ = 10 Å

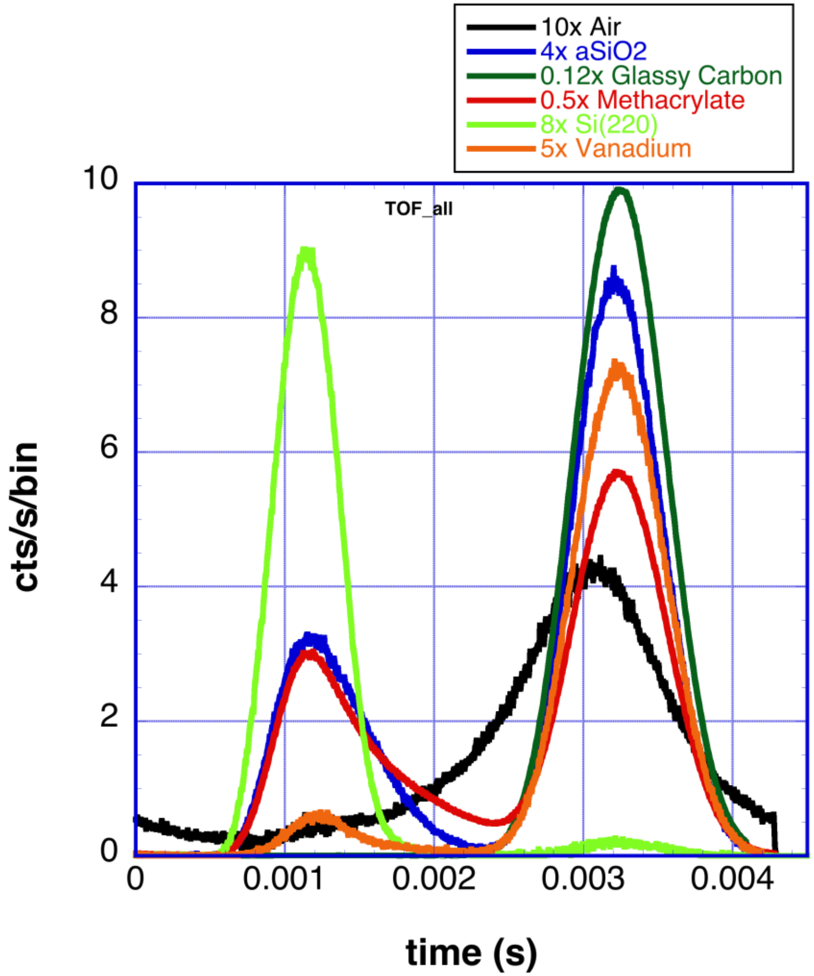


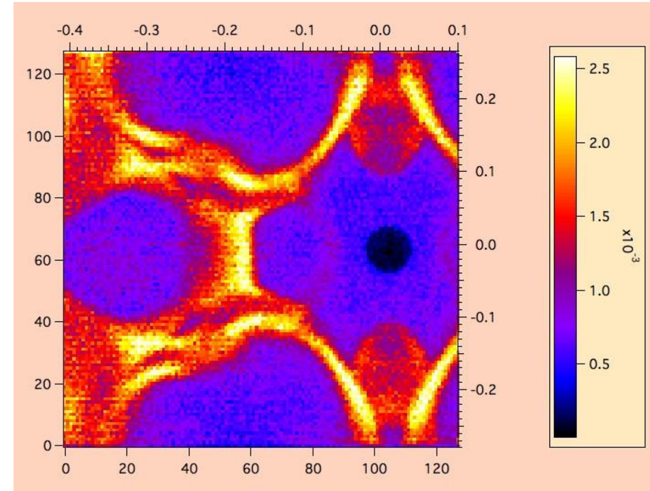


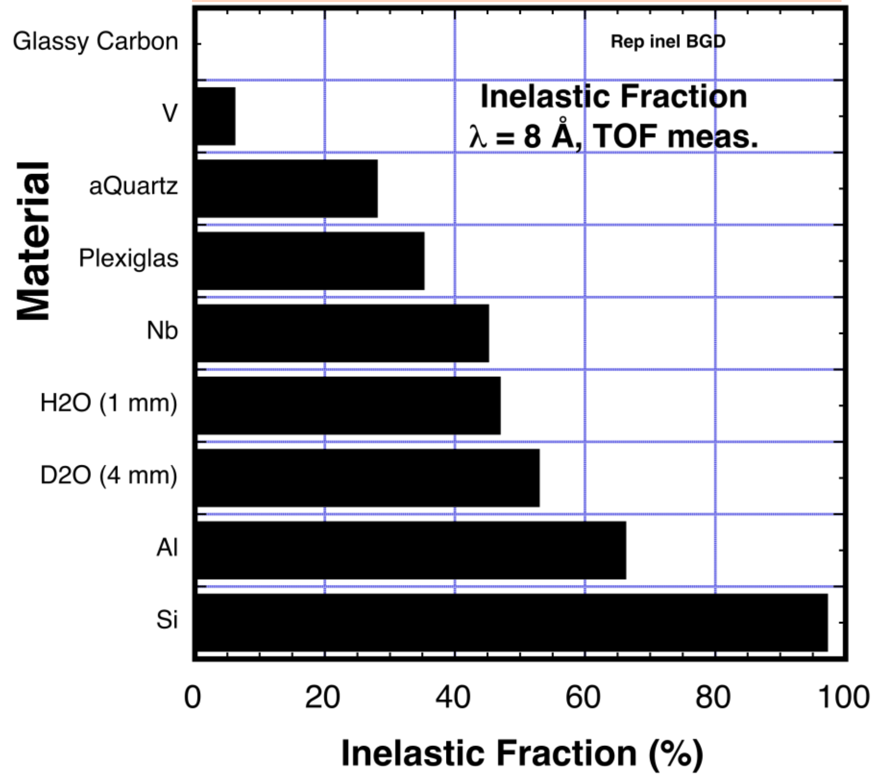
Scattering by thickness:
Strongest → H_2O, D_2O
Weakest → Ar, He gas (1 bar)

Scattering per atom:
Strongest → H_2O, Air
Weakest → Sapphire, Silicon

TOF measurements with disk chopper to separate inelastic from quasi-elastic scattering:





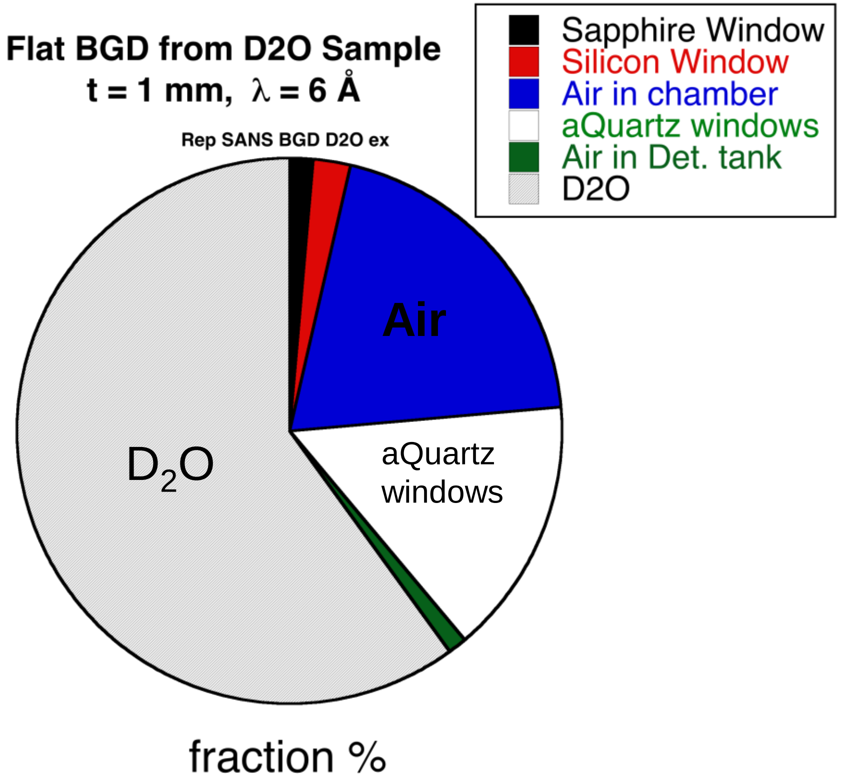


Flat Background from 1 mm thick D_2O Sample:

Material	Thickness
Air	150 mm
aQuartz	3.2 mm
Sapphire	3.2 mm
Silicon	6.4 mm
Vacuum (0.1 Torr)	4000 mm

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 wit**h 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 {Lense config.}

Future Work:

Resolve discrepancy in Bgd measured at different facilities