A time resolved high energy X-ray diffraction study of cooling liquid SiO$_2$

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Physics 570
Introduction

- **Purpose:** To study the structure of liquid SiO2 as it cools through its transition phase to glass.
- **Why Synchrotron:** Negligible attenuation and multiple scattering effects on mm sized samples, can access large momentum transfers, high energy, high temporal resolution.
Methods

• Synchrotron Specs
  o Beamline 11-ID-D at APS, Argonne National Lab
  o 114.78keV incident energy
  o 0.5x0.5mm beam size

• 3 experimental runs
  o (1) 2mm diameter spherical SiO2, suspended and melted in aerodynamic levitator
  o (2) “Candlestick” set up
  o (3) Same set-up as 2, but double the sample-detector distance
  o All: Heated from above with 240W CO2 laser
  o Levitation: Levitator enclosed in stainless steel chamber with Kapton windows that transmit X-rays with low attenuation or scattering.
Theory/Analysis

1. Intensity at each temperature was analyzed using software to produce $S_x(Q)$

$$S_x(Q) = \frac{I(Q)}{Nf(Q)^2}$$

2. Evaporative loss: normalize each temperature measurement to the X-ray form factor

3. Each Structure Fourier transformed

$$g_x(r) = 1 + \frac{1}{2\pi^2 r \rho} \int_{Q_{min}}^{Q_{max}} Q[S_x(Q) - 1]M(Q, \Delta(r)) \sin(Qr) \, dQ$$

$$M(Q, \Delta(r)) = \text{Modification function}$$

$$\Delta(r) = \text{width of R-dependent real space broadening}$$

4. Minimize systematic errors from slowly varying backgrounds

Un-physical features in real space set to 0 in $g_x(r)$ and Fourier back-transformed into reciprocal space.
Theory/Analysis

5. Fitting First Sharp Diffraction Peak (Lorentzian function)
\[ L(Q) = \frac{Ad}{\pi} \left( \frac{1}{(Q - Q_1)^2 + d^2} + \frac{1}{(Q + Q_1)^2 + d^2} \right) \]

5. Determine Average Si-O coordination number (remove Q-dependent weighting factor)

6. Width of Si-O peak is a result of convolution of data window and x-ray weighting factor with true distribution of atom positions

7. Measured Q-space oscillations due to nearest neighbor Si-O peak (fit to back transformed pattern to measure Si-O peak position, width and coordination number.

8. Emphasize intermediate range order, remove bulk density to get differential real space function.
\[ D_X(r) = 4\pi\rho r [g_X(r) - 1] \]
Results
Results
Conclusions

- **Density maximum observed** at 1750K – correlated to onset of slight reduced intermediate range order and structure coherence.
- **Unexpected decrease in both position and width** of average tetrahedral Si-O peak at 1500K, corresponding to glass transition temperature for pure silica and density minimum.
- **Tetrahedral coordination confirmed** – average coordination number of near neighbor slits at 3.96.
References


