Temperature Variations in the Reduced Distribution Function of Liquid Metals and Metallic Glasses

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Introduction

Third generation synchrotron sources, in conjunction with area detectors, have reduced data collection times to as little as a few seconds or less [1,2]. This allows for rapidly obtaining hundreds of data sets. These time-resolved measurements allow, for instance, the study of chemical and topological ordering of metallic glasses and how local fluctuations may control devitrification [3], or subtle local atomic rearrangements proceeding phase transitions. All rely on measuring small changes in the total scattering function. The pair distribution function (PDF) analysis provides a means of recovering the average real-space bonding without a priori knowledge of the atomic structure. However, the amount of corrections of the data for accurate pair distribution function (PDF) analysis greatly complicates automation. Tracking changes during supercooling of a liquid or if clusters form proceeding devitrification of a glass all hinge on measuring how the local order is evolving as a function of time or temperature. Concentrating on the change rather than the absoulte values 1) greatly simplifies the data analysis for detecting changes in the PDF and 2) allows for rapid processing of large data sets.

Methods and Materials

The observed x-ray scattering intensity ($I^{obs}(Q)$) is measured in terms of the wave momentum number and contains a number of contributing components;

$$I^{obs}(Q) = PAG[I^{c}(Q) + I^{i}(Q) + I^{m}(Q)] + \mathrm{B} \qquad \text{eq. (1)}$$

where **B** is background, **P** is the polarization, **A** is absorption and **G** is geometrical corrections while $\boldsymbol{I^{c}}$ is the coherent, $\boldsymbol{I^{i}}$ is the incoherent and $\boldsymbol{I^{m}}$ is the multiple scattering. Assuming that the sample cross section and the flux on the sample has not changed, the difference is

$$\Delta I^{c}(Q) \approx C * \Delta I^{obs}(Q) \qquad \text{eq. (2)}$$

where the $\lim_{Q\to\infty} \Delta I^{c}(Q) = 0$. The contribution from **P**, **A** and **G** should be invariant for the experiment. Using the Faber-Ziman formalism for the total scattering function S(Q), the change in S(Q), ΔS(Q), can be easily be written as;

$$\Delta S(Q) = \frac{C\Delta I^{obs}(Q)}{< b >^{2}} \qquad \text{eq. (3)}$$

where $< b >= \sum_{i=1}^{n} a_i f_i(Q)$. The change in the PDF is then given by,

$$\Delta G(r) = \frac{2}{\pi}\int_{0}^{\infty} Q[\Delta S(Q)]\sin(Qr)dQ \qquad \text{eq. (4)}$$

Results

Figure 1 is a comparisions of $\Delta G(r) = G(r)^{973} - G(r)^{T}$ for a pure liquid Al (T=1073 and 1274 K) shown as open symbols (i.e., standard analysis). The ΔG(r)'s derived by applying eq. (4) to $\Delta S(Q) = (I(Q)^{973} - I(Q)^{T})/<b>^{2}$ is shown using solid lines. The scattering data were collected at the MUCAT 6-IDD beamline at the Advanced Photon Source using an energy of 98.107 keV in transmission mode using a MAR345 image plate detector.

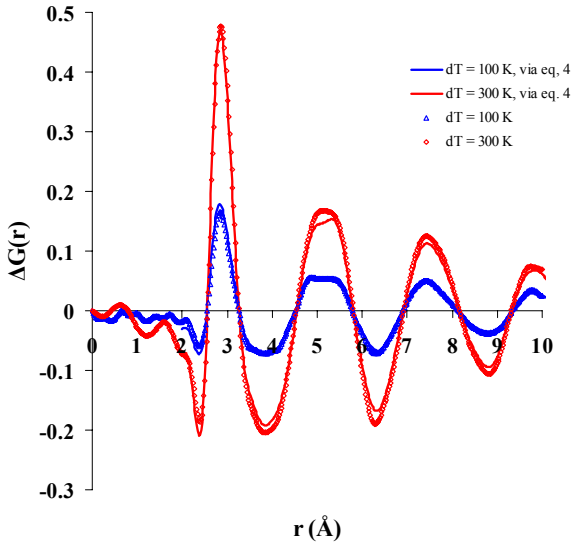

Fig. 1. (see text for details)

Discussion

The difference in the S(Q) and G(r) can be readily determined from the raw I(Q) if the experiments are properly designed. The difference plots obtained from performing the full standard data analysis versus applying eqs (2)-(4) are nearly identical. This methology provides a rapid means of quickly analyzing time-resolved data which may involve hundreds of scans and a unique insight to how the pair correlations change under any number of external conditions. These external inputs can be thermal, mechanical or even magnetic. The perturbations of the PDF can be either short ranged or long ranged.

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