Review Article

A Review of High-Energy X-Ray Diffraction from Glasses and Liquids

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This paper summarizes the scientific trends associated with the rapid development of the technique of high-energy X-ray diffraction over the past decade pertaining to the field of liquids, glasses, and amorphous materials. The measurement of high-quality X-ray structure factors out to large momentum transfers leads to high-resolution pair distribution functions which can be directly compared to theory or combined with data from other experimental techniques. The advantages of combining highly penetrating radiation with low angle scattering are outlined together with the data analysis procedure and formalism. Also included are advances in high-energy synchrotron beamline instrumentation, sample environment equipment, and an overview of the role of simulation and modeling for interpreting data from disordered materials. Several examples of recent trends in glass and liquid research are described. Finally, directions for future research are considered within the context of past and current developments in the field.

1. Introduction

X-ray diffraction studies have long been used to obtain information on the short and intermediate range structure of glasses, since the pioneering work of Warren and coworkers in the 1930's [1–3]. This is still the case, although the measurement accuracy, data analysis procedure, and sophistication of the modeling techniques today are much more rigorous. The work of Narten and colleagues in the 1970's had a large impact on the development of data analysis techniques associated with the X-ray pair distribution function method, using conventional X-ray sources mainly to study the structure of molecular liquids, for example, [4, 5]. Around the same time, Leadbetter and Wright combined both neutron and X-ray diffraction to study the structure of network glasses [6, 7]. However the main use of the pair distribution function technique in the field of glasses and liquids centered on neutron diffraction which has been described extensively by Wright [8–10] and others. The high-energy X-ray technique (loosely defined as energies >60 keV) is the latest generation of this popular method of determining the structure of disordered materials and has its origins with the seminal work of Egelstaff and Root in

the 1980's who used γ -ray diffraction to obtain liquid structure factors [11, 12]. The breakthrough however came in the mid-90's by Neufeld and Poulsen [13–15], when they adapted this technique to synchrotron radiation using 100 keV X-rays. The triple axis high-energy X-ray diffractometer, BW5, they used for these measurements was built based on the design of the world's best neutron diffractometer at the time, D4C at the Institute Laue Langevin in France [13]. The high energy X-ray pair distribution function (PDF) technique has spread in the last decade and current synchrotron beamlines that routinely perform measurements on glasses include: SPring-8, Japan (BL04B2) [16, 17], HASYLAB [18], Germany (BW5), European Synchrotron Radiation Facility, France (ID15B), and several beamlines at the Advanced Photon Source, USA; 1-ID (stress/strain, high pressure), 11-ID-B (time resolved, chemical reactions), 11-ID-C [19] (liquid, glass, levitation). In the conference summary of the 10th International conference on the Structure of Non-Crystalline Materials in 2006, X-ray diffraction was identified as the main structural technique used by the participants.

There are two main advantages over conventional X-ray diffraction techniques when hard X-rays of ~100 keV

FIGURE 1: Plots showing the number of papers published per year and their citations from a *Thomson Reuters Web of Knowledge* database search using the key words “high energy X-ray diffraction” and “liquid” or “glass” cross-linked with several of the most prolific authors in the field. These articles were cited 1939 times by 1539 articles (excluding self-citations) with an average of 9.78 citations per item.

are used in experiments on amorphous materials: (i) the structure factors can be measured out to much higher momentum transfers, $Q > 20 \text{ \AA}^{-1}$ at smaller scattering angles (in $2\theta^\circ$), leading to higher real space resolution, and (ii) attenuation and multiple scattering effects are negligible for small samples that is, typically $\sim 1 \text{ mm}^3$ [13]. This is because the photo-electric absorption decreases as $\sim E^{-3}$ and scattering typically becomes the dominant process, under conditions similar to that of a transmission neutron diffraction experiment. High energy X-ray diffraction is very much a sister technique to neutron diffraction, and as for all glass studies, whenever possible should be combined with other structural probes to maximize the information obtained. Limitations of the technique include that it is not element specific (like EXAFs) and therefore information on low dopant ions ($<1\%$) cannot be obtained. Fluorescence energies of elements in the sample should also be avoided if possible. One of the strengths of high energy diffraction data lies in its ability to provide a rigorous test of atomistic models from computer simulation such as molecular dynamics or density functional theory. It is also often used as a model constraint in inverse computer simulation techniques, such as Reverse Monte Carlo and Empirical Potential Structure Refinement. Some current directions of high energy X-ray synchrotron radiation in glass research include high pressure studies, high and low temperature experiments combined with containerless levitation techniques, and time resolved structural measurements around the glass transition.

To my knowledge there have been three reviews on high energy photon scattering specifically aimed at investigations into the structure of liquids and glasses. The first two were by Dr. J. Neuefeind in 2002 [20] and Professor P. A. Egelstaff in 2003 [21] shortly after high energy X-ray diffraction at third generation synchrotrons was born and these were followed by Dr. S. Kohara et al. review in 2007 [22]. During

this time and since, the field has grown rapidly driven by advances in instrumentation and detectors at synchrotron sources, as illustrated in Figure 1. Here I attempt to update and rationalize these changes in the context of other current experimental and modeling methods. This paper is from my own perspective working for the past two decades in the field of liquids and glasses, and mainly describes the developments on sectors 1-ID and 11-ID-C at the Advanced Photon Source near Chicago, USA. This paper is by no means meant to be comprehensive and I apologize for those contributions to the field I have inadvertently omitted.

2. γ -Ray Diffraction

Knowing the atomic structure of a material is the starting point for explaining many macroscopic phenomena, unusual properties, or behavior. The ability to probe the bulk local and intermediate range structure of disordered materials is a powerful tool, yet for many materials science problems, more penetration is required than can be provided by conventional X-ray instrumentation. The idea of high energy (low wavelength) radiation combined with low-angle detection came from developments in γ -ray scattering, the precursor to high energy X-ray diffraction. In particular, Egelstaff and Root designed and built a γ -ray diffractometer at the university of Guelph in Canada expressly built to measure liquid X-ray structure factors [12]. A very stable 60 keV, Americium-241 source was rotated in an arc around the sample with the heavier detector at a fixed position, see Figure 2. The limiting factors associated with these experiments were the low-flux, as experiments would typically take months to complete, which was compensated for by a large sample size leading to poor resolution at low angles and significant geometric, attenuation, and multiple scattering effects.

FIGURE 2: A schematic of the 60 keV γ -ray diffractometer built at the University of Guelph, Canada.

3. High Energy X-Ray Instrumentation

In the past, spallation neutron diffraction measurements have played an important role in elucidating the bulk structure of materials over a large range of scattering vector Q , covering low scattering angles and short wavelengths, but these experiments require comparatively lengthy counting times and large samples [23]. The use of “neutron-like” photons for diffraction studies at synchrotrons uses ca. 100 keV photons to compress a wide Q -range into a small angular cone in the forward scattering direction, providing high real space resolution at low- r values comparable to the best neutron sources. For example, the high energy X-ray diffractometer 11-ID-C at the Advanced Photon Source was originally designed and built by Rütt and coworkers at the Basic Energy Sciences Synchrotron Radiation Center (BESSRC) in 2001 [19]. It was designed as a triple axis machine using an elliptical multipole wiggler. In 2007 the wiggler was replaced by two in-line undulators in a tandem configuration, one full length undulator A (downstream) and a full length high energy undulator with a period length of 2.3 cm, providing simultaneous high brilliance flux to all three 11-ID stations (B, C, and D). The undulator A is limited to a minimum gap equivalent to $k = 2.11$ (minimum energy of 4.5 KeV) and the high energy undulator is used at fixed closed gap of 10.5 cm. During a 2010 upgrade the 11-ID-C energy remained fixed at 115 keV using a Laue Si(311) crystal reflection at 1.8 degrees, providing a highly penetrating beam and allowing a wide coverage of reciprocal space over a small angular scattering range. The flux is typically 10^{11} photons/sec using a beam size of $1 \text{ mm} \times 1 \text{ mm}$. In 2010, the end station was redesigned to take advantage of the advances in detector technology and rebuilt around a large moveable flat-plate area detector, see Figure 3. The open design, heavy duty sample stage, and extensive range of detector motion on 11-ID-C distinguishes it from beamlines performing similar measurements at other synchrotrons, such as BL04B2 and BL08W at SPring-8 [17] and ID-15B at ESRF. The use of short wavelength X-rays are well suited to experiments that require bulky sample environments and much of the science program at beamline 11-ID-C revolves around studying the

structure of phase transitions or materials characterization at nonambient conditions.

4. Data Analysis and Corrections

In the data analysis procedure we can consider three different origins of effects which can affect the accuracy of the extracted X-ray static structure factor [16, 24]: (i) effects related to the source for example, polarization, energy resolution, and relativistic effects; (ii) sample and environmental effects for example, container, attenuation, multiple scattering, fluorescence and (iii) detector effects, for example, geometrical arrangements, oblique incidence, detector efficiency, flat field, dark currents. Many of these have been covered in the literature and can be found here: [16, 24–26]. These corrections and the order they are applied must be considered together with the removal of the background scattering (air or vacuum plus any windows) and the composition dependent Compton scattering. Removal of the “self scattering” (Compton plus X-ray form factor) allows the extraction of a pseudo-nuclear total X-ray structure factor, $S(Q)$ to be obtained. Provided a wide range of reciprocal space is covered this function can be Fourier transformed into real space to provide an average probability function of all the atomic positions in the material called the radial or pair distribution function $G_X(r)$ [1]. The $G_X(r)$ function can be used to extract bond distances, local coordination numbers, average bond angles, and provide a rigorous test of structural models. In addition, the first sharp diffraction peak at position Q_1 in the X-ray (or neutron) diffraction patterns is associated with the existence of intermediate or medium range order in glass with a periodicity of $2\pi/Q_1$ (although its origin is still controversial). Medium range order has been defined as covering the region $\sim 5\text{--}10 \text{ \AA}$, although longer “extended chemical ordering” up to $40 \text{ \AA}$ has also been found in network glasses.

The sample dependent absorption corrections to the scattered X-ray intensity $I_X(Q)$ from a liquid or glass can be reliably applied using the method of Paalman and Pings [27], by independently measuring the sample scattering in a vessel I_0^{SV} , the empty vessel I_0^V and the background B ,

$$I_X(Q) = K \left(\frac{I_0^{SV} - B}{A_{S,SV}} - \frac{A_{V,SV}(I_0^V - B)}{A_{S,SV}A_{V,V}} \right) - \langle f^2(Q) \rangle M_{SV}, \quad (1)$$

where the coefficient $A_{S,SV}$ represents the attenuation of the sample in the presence of the sample plus vessel, $A_{V,SV}$ the attenuation of the vessel in the presence of the sample plus vessel, and $A_{V,V}$ the presence of the vessel in the presence of the vessel. The average scattering is given by

$$\langle f^2(Q) \rangle = \left[\sum_{i,j=1}^n c_i f_i(Q) \right]^2. \quad (2)$$

However, for thin samples with few electrons the ratio of multiple to single scattering [28, 29] of the sample in the vessel M_{SV} is negligible, so the second term in (1) can often

FIGURE 3: The high energy X-ray diffractometer 11-ID-C at the Advanced Photon Source with an aerodynamic levitator system installed.

be discarded. K is the normalization factor [30] required to formalize $I_X(Q)$ to the number of electrons (plus any residual self-scattering, see (2) below) such that $S_X(Q)$ oscillates about unity at high Q values. For homogeneous liquids the normalized $I_X(Q)$ tends to the isothermal compressibility as Q tends to zero as given by

$$I(Q \rightarrow 0) = \rho k_B T \chi_T Z, \quad (3)$$

where Z is the total number of electrons in a single molecule, k_B is the Boltzmann constant, T is the absolute temperature, and χ_T is the isothermal compressibility.

There are many different formalisms for the total structure factor $S_X(Q)$. Experimentally, they all derive from the measured elastically scattered intensity $I_X(Q)$ by subtracting a form factor $f^2(Q)$ approximating the electron density (self scattering of the atoms or molecules) and the Compton scattering contribution, $C(Q)$. The most common is to divide by the average scattering $\langle f^2(Q) \rangle$,

$$S_X(Q) - 1 = \frac{I_X(Q) - (\sum_{i=1}^n f_i^2(Q)) - C(Q)}{\langle f^2(Q) \rangle}, \quad (4)$$

where i and j represent the different atomic species in the molecule [31–33]. For molecules or well defined “molecular units” we can define a function,

$$S_{\text{molecule}}(Q) = \sum_{i,j=1}^m \frac{N c_i c_j f_i(Q) f_j(Q)}{2} \text{sinc}(Q r_{ij}) \exp(-Q^2 \sigma_{ij}), \quad (5)$$

where

$$S_X(Q) = S_{\text{molecule}}(Q) + S_{\text{inter-molecule}}(Q). \quad (6)$$

N denotes the coordination number, r_{ij} are the intra-molecular atomic separations, and σ_{ij} are the associated mean squared displacements. The nonspherical shape of the electron cloud in liquids or glasses with few electrons, such as hydrogenous materials, requires a redistribution of charge to be taken into account. This affects the shape of the $S(Q)$ at low Q -values that is, $Q \leq 2 \text{ \AA}^{-1}$. In the case of water the spherical independent atom approximation form factors have been successfully modified to give the Modified Atomic Form Factor (MAFF) through [34],

$$f_{\alpha}^{\text{MAFF}}(Q) = \left[1 - \frac{Z_{\alpha}}{f_{\alpha}(0)} \exp\left(-\frac{Q^2}{2\delta_{\alpha}^2}\right) \right] f_{\alpha}(Q), \quad (7)$$

where Z is the fractional electron charge on the α atoms and it is required that $\sum_{\alpha} Z_{\alpha} = 0$ to conserve charge.

Given the electron density approximation, a (pseudo-nuclear) pair distribution function $G_X(r)$ can be defined via a Sine Fourier transformation,

$$G_X(r) - 1 = \frac{1}{2\pi^2 r \rho} \int_{Q_{\min}}^{Q_{\max}} Q [S_X(Q) - 1] \sin(Qr) dQ, \quad (8)$$

where $Q_{\min}$ and $Q_{\max}$ represent the finite range in reciprocal space over which the X-ray data are measured and ρ is the atomic (or molecular) number density in $\text{\AA}^3$. Of these two limits the $Q_{\max}$ often has the most noticeable effect on the Fourier transform, especially at low- r , as the $S_X(Q)$ curve should be truncated at precisely 1.0 at a node, to avoid transforming a step function. A Lorch [35] or other modification function is often used to minimize the oscillations generated during the Fourier transform over a finite Q -range. Some useful software packages used to reduce the measured X-ray diffraction pattern to the structure

factor and pair distribution function include *ISOMERX* [36], *FIT2D* [37], *PDFGETX2* [25], and *GSAS-II* [38].

5. Advantages

High energy X-rays or “hard” rays typically have energies of 60 keV–150 keV, about one order of magnitude higher than conventional X-rays [39]. The main advantages of conducting these experiments on liquids and glasses using high energy photons include the following.

- (i) High-momentum transfers can be accessed, leading to high-real space resolution at short distances in the pair distribution function. This can aid in accurately distinguishing between two average bond distances which are very close together, for example.
- (ii) The high penetration allows experiments to be conducted in air and in transmission geometry, in which the scattering is concentrated in the forward direction with minimal polarization effects. This allows the use of a variety of sophisticated sample environments and a straightforward detector arrangement.
- (iii) Photo-absorption strongly depends on the atomic number of the material and is greatly reduced at higher energies, so heavy element containing samples can be studied.
- (iv) Radiation damage, particularly from biological samples, is greatly reduced.
- (v) The measured X-ray structure factors and corresponding pair distribution functions are directly comparable to neutron diffraction studies measured over similarly wide Q -ranges.

Of all these, the two most exploited advantages at the APS have been (ii) on liquids at extreme conditions and (v) on glasses where a section of the same sample can be used in both experiments.

6. Experiments on Glasses and Liquids

Early papers in the field of glasses using high energy X-ray diffraction focused on classic network glass formers such as SiO_2 [13], GeO_2 [14], P_2O_5 [40], and B_2O_3 [41], as well as tellurite [42], iron phosphate [43], and calcium aluminate glasses [44]. The structure of liquid ZnCl_2 [45], iron trichloride [46], and methanol [47] were also studied. The field of isotopic quantum effects in hydrogen bonded molecular liquids originally studied using gamma-ray diffraction was revitalized, focussing on the deuterium effect in water [48] and methanol [49–51]. The high energy X-ray diffraction studies of glasses [52–55] can be broadly split into three main areas, oxides (e.g., silicates, germanates, borates and aluminates, etc.), chalcogenides and chalcogenhalides, and Bulk Metallic Glasses (BMG's).

Oxide glasses have particular relevance in geoscience and technology and include soda-lime window glass, borosilicate

pyrex glass, lead oxide glassware, aluminosilicate fiberglass, and aluminate glass fiber optics. Much of the research has focused on the short and intermediate range order in silicates [57–65], Na-silicates [66, 67], germanates [68], borates [69–71] aluminates [72–74], and aluminosilicates [44, 75, 76]. XRD PDF data on oxide glasses is highly complimentary to neutron diffraction PDF data since neutrons are more sensitive to the oxygen atoms, often providing a stark contrast in the partial structure factor weighting factors. Phosphate glasses [77–84] represent an important class of polymeric and molecular oxide glasses with similar applications to silicates, such as radioactive waste containment and optical fibers as well as biochemistry. Extensive studies have been carried out by Hoppe and coworkers on phosphates doped with rare-earth ions including; La [77], Ga [78], Fe [79], Pb [80], Zn [81], and Ti [82]. Hoppe et al. [77–82] developed a set of rules for network changes upon the addition of modifying atoms to describe the disruption of P-O-P bonding units as a function of P_2O_3 content.

Chalcogenide glasses contain a major constituent from a group 16 element of the periodic table. These network glasses are covalently bonded like oxides but can also form homopolar bonds making a richer combination of structural motifs. Modern technological applications include mouldable intra-red optics and fibers, lenses, memory devices, and phase change materials. High energy X-ray diffraction studies have been carried out on selenides [85–91], for example, see Figure 4, sulfides [92–98], arsenides [96–98], tellurites [99–104], and chalcogenhalides [105–107]. Bychkov et al. found a complex variation in network formation and destruction versus composition in binary chalcogenides [85]. Chemical ordering effects [98] and conduction pathways in fast-ion conducting chalcogenide glasses have also been an area of significant interest relating structure to changes in ion transport [87, 88, 94, 95, 105–107].

Bulk metallic glasses are generally made in small batches by rapid cooling methods to produce strong materials with good electrical conductivity. High energy X-ray diffraction is well suited to the study of BMG's [108–130] because of the high signal/background signal and both binary [113–117] and multicomponent [118, 121] systems have been studied extensively using the HEXRD technique. Of particular interest has been structural ordering upon supercooling [119, 120], thermal behavior [121, 122], and correlating the mechanical or tensile behavior with local distortions in the local structure [123–126]. Attention has also been paid to their glass forming ability [127, 128] and an important theme has been identifying structural heterogeneities in the deeply supercooled melt [129, 130].

In the case of liquids, several metals and alloys [131–134] have been studied in the molten state, along with Ge [135], tellurites [136, 137], aluminates [138, 139], silicates [140], and aluminosilicates [141]. Molecular liquids have also been investigated extensively with high energy X-ray diffraction, including the electron density in water [142], the effect of strong hydrogen bonding in deeply supercooled water [143], fluorides [144–150], the structure of molten

FIGURE 4: High energy X-ray diffraction data on GeSe_2 glass measured on beamline 11-ID-C at the Advanced Photon Source. (a). The X-ray structure factor. (b). The corresponding pair distribution function. Courtesy of L. Skinner.

salts [151, 152], as well as temperature and composition-dependent intermediate range order in ionic liquids [153–163]. The structural chemistry of actinide solutions has also been explored [164–168]. Experiments on these radioactive liquids and highly corrosive acids [146, 147, 150] have led to the design of specialized containers [169] which the high energy X-rays are able to penetrate.

High energy X-ray scattering techniques have been effectively used to measure the electron density distribution differences between light and heavy water [48, 142, 170–172], benzene [173], methanol [49–51, 174], ethanol [175], as well as amorphous hydrides [176, 177]. Since the electronic scattering is essentially the same for both H and D, changes in the zero point energies lead to subtle changes in both the intra- and intermolecular structures. These isotope quantum effects obtained by comparing X-ray diffraction from the hydrogenous and selectively deuterated forms of molecular liquids have important implications for the accuracy of the H/D substitution technique in neutron diffraction. The deuterated liquids have a more rigid (ordered structure), and the hydrogenous liquids more quantum mechanical. Computer simulations suggest that the substitution is essentially equivalent to reducing the quantum mechanical effects by half in liquid water upon deuteration [48]. This has led to comprehensive studies of temperature [170, 171] and density [178] dependent deuteration effects (see Figure 5) which will hopefully lead to the development of better intermolecular potentials.

Amorphous solids usually refer to disordered materials which are produced by means other than quenching the liquid (chemical reactions, vapour deposition, etc.) and do not exhibit a glass transition temperature. High energy X-ray diffraction studies of technological importance on amorphous materials include calcium-silicate hydrate cements [179–182], bioactive foams [183], amorphous zeolites [184, 185], spider silk fibers [186], and electronic materials [187, 188].

FIGURE 5: The difference between the electronic distribution functions of light and heavy water as a function of temperature.

7. Extreme Environments

Increasingly, materials need to be measured under realistic, sometimes extreme, conditions and over a range of timescales.

The effect of high pressure on the structure of glass have been of interest since the pioneering work of Bridgman in the 1950's [189]. Densified glasses subjected to high pressures and recovered to ambient pressures generally show a significant loss of intermediate range order but very little change in short range ordering [190–193]. Moissanite [194] and Diamond Anvil Cell measurements [195–208] on glasses

FIGURE 6: A schematic of the high pressure Perforated Diamond Cell set up on 1-ID. Adapted from a talk by E. Soignard.

FIGURE 7: X-ray structure factors for GeSe_2 glass at high pressure measured in a Diamond Anvil Cell at beamline 1-ID at the APS.

have been able to achieve several tens of GPa's leading to much larger changes in structural packing as well as local coordination number changes, for example, see Figure 6. In addition to the single crystal Bragg scattering which has to be removed during the data analysis procedure, the large Compton scattering contribution from the diamond has been greatly reduced by perforating one of the diamonds [209]. The study of silicate glasses at high pressure has attracted interest from the geological community from the point of view of understanding the atomic structure and transitions that occur in magmas within the Earth's mantle [202, 206]. The densification of chalcogenide glasses [194, 195, 198, 200, 206] with pressure are more complex due to the propensity of homopolar bonding (see Figure 7) and offer the possibility of forming high-density glasses which may be retained at ambient pressure [194]. Some current trends include measurements under hydrostatic conditions [203] and the development of new pressure cells to explore new regions of the liquid phase diagram not previously accessed by high energy X-ray diffraction [209]. These include the a hydrothermal (HDAC) cell design aimed at accessing low pressures and high temperatures [209], and gas pressure cells with low-background amorphous carbon windows ideal for studying poorly scattering aqueous solutions. Measuring

strain distributions in amorphous materials also continues to be a topic of significant interest [210–212].

For glasses, the study of (usually) small structural changes around the glass transition temperature which may be associated with large changes in the dynamical behavior occurring has been of particular interest [56, 213–229]. This has led to the development of rapid data acquisition methods (up to ~ 100 ms) applied to glass forming liquids upon cooling from very high temperatures [215–219] as well as studies of the stable melts [220–224]. Time-resolved measurements have been made on supercooled oxide liquids held in aerodynamic levitators from temperatures up to 2500°C [225–227] (see left hand side of Figure 8) and liquid metals suspended in electrostatic levitators [228, 229]. Recently acoustic levitation combined with high energy X-ray diffraction (see right hand side of Figure 8) has been used to study the structure of supercooled, glassy, and amorphous organic liquids and solutions at temperatures down to -20°C [230]. An interesting new application using this technique is the application for making and characterizing fast acting amorphous drugs [231, 232]. The ability to model different molecular conformations of amorphous drugs made using different methods and compare the results directly to the pair distribution function could provide useful information to the pharmaceutical industry.

It has long been known that glasses and amorphous solids are by definition inherently polyamorphic, since their exact structure depends on the route by which they were produced. However in recent years the term “polyamorphism” in liquids has come to be associated with a first order phase transition between two distinct structural forms of the same material [233–235]. Since many of the proposed transitions occur at extreme conditions, high energy X-ray diffraction has been used extensively in this area to identify different structures of potential polyamorphic materials. The abrupt transition(s) observed between different density forms of amorphous ice has often been used as an analogy with what could occur in the liquid state, and the X-ray diffraction patterns at different densities are strikingly different [236–241]. High temperature aerodynamic levitation has also been used to investigate the two phase structure of liquid and glassy yttria aluminates [242–247], for example, see Figure 9, and low temperature measurements have been carried out to characterize the so-called glacial state of the molecular liquid triphenyl phosphite [248–250].

8. Data Interpretation

The most common way of interpreting total scattering data is to use the Faber-Ziman formalism [251] which defines element specific partial structure factors. Other formalisms are occasionally used in the literature by various groups and have been summarized by Keen [252]. There are also several different variations for representing the Faber-Ziman partial structure factors and partial pair distribution functions, used by different communities to highlight different aspects of the patterns. Here we describe the Hannon-Howells-Soper

FIGURE 8: (a) A laser-heated sample in the aerodynamic levitator chamber. The chamber totally encloses the X-ray beam to enable a class I system operation despite containing an embedded class IV laser. (b) An acoustic levitator, comprising of two transducers (blue) with a camera focused on the levitated droplet and an area detector in the background.

FIGURE 9: A time dependent sequence showing the melting and subsequent vitrification of an oxide melt in the aerodynamic levitator.

formalism [252] commonly used for liquids and glasses where for X-rays,

$$S_X(Q) = \sum_{\alpha,\beta} \frac{c_\alpha f_\alpha(Q) c_\beta f_\beta(Q)}{\langle f^2(Q) \rangle} [S_{\alpha\beta}(Q) - 1], \quad (9)$$

where α and β represent different atom types. A discussion of the density related behavior of the intensity of the so-called “first sharp diffraction peak” related to intermediate range ordering compared to that of the second diffraction peak (related to chemical or extended range ordering) in several binary liquids and amorphous materials has been given by Benmore et al. [253]. The Sine Fourier transform given in (8) yields the pair distribution function $G_X(r)$ which is commonly used by the liquids community to emphasize local structure. The neutron glass community on the other hand tends to use the distribution function $T_X(r) = 4\pi r G_X(r)$

since the resolution function is symmetric leading to more accurate fitting for extracting coordination numbers [23]. However, for X-rays peak and coordination number fitting is often done in Q -space using equation (6) due to the need to include Q -dependent form factors [33]. The differential distribution function $D_X(r) = 4\pi r [G_X(r) - 1]$ with the bulk density removed is used for glasses and molecular liquids to highlight ordering at longer distances or for systems where the density is not known [23]. Expressing the $G_X(r)$ representation in terms of partial distribution functions is not so straight forward as for neutron diffraction because the element specific partial weighting factors in reciprocal space are Q -dependent. The expression can be simplified by using the approximation $f_\alpha(Q)/\langle f^2(Q) \rangle \simeq f_\alpha(0)/\langle f^2(0) \rangle$, where $f_\alpha(0) = Z_\alpha$ as has been described by Neufeld and Poulsen [13] and Keen [252].

The Bhatia-Thornton formalism [254] is also sometimes used to provide information on the topology and chemical ordering in a liquid or a glass. For a binary system the Bhatia-Thornton representation is straight-forwardly linked to the Faber-Ziman formulation using linear equations which convert the element specific partials to the number-number, concentration-concentration, and the cross term number-concentration partial structure factors. The Bhatia-Thornton formalism has been successfully used to explore the extent of longer range correlations in real space known as “extended range ordering” [255]. However the magnitude of these oscillations show that the degree ordering only represents a very small fraction of the bulk material [256].

Partial extraction or elimination of one or a group of partial structure factors from the measured total is most commonly achieved experimentally by combining high energy X-ray diffraction data with other techniques, such as neutron diffraction or anomalous X-ray diffraction.

Various first or even second order difference functions may be extracted if there is sufficient contrast, to extract information on a particular feature of interest. EXAFS is also commonly used to help remove element specific peaks of a known coordination number from the total pair distribution function. Nuclear Magnetic Resonance and to some extent Raman scattering provides complementary information on the speciation of certain elements which cannot be directly accessed using diffraction data alone, but may be used a strong constraints on the interpretation of X-ray PDF data.

9. Main Limitations of the Technique

By far the largest limitation of high energy X-ray scattering is the rapidly decaying X-ray form factor signal compared to the growing Compton scattering (background) contribution at high- Q values, making the accurate extraction of $S_X(Q)$ over the widest accessible Q -range problematic. At the highest Q -values, for example, $>30 \text{ \AA}^{-1}$, small corrections of less than a percent from the source, sample, and detector become magnified, leading to greater statistical and systematic errors, such that accurate normalization in absolute units becomes increasingly difficult. The situation is worst for low- Z materials. In addition, currently the most information that can be reliably extracted from an X-ray experiment from an isotropic glass or liquid is a one-dimensional distribution function from which the three-dimensional structure cannot be directly generated. The X-ray data therefore represent an average overview of the structure from which only the first few peaks in the pair distribution function can typically be used to extract accurate atom-atom distances and coordination numbers.

10. Simulation and Modeling

In practically all cases a complete structural picture of the liquid or glass can only be achieved with the help of some sort of computer modeling or simulation. Inverse methods such as Reverse Monte Carlo (RMC) [257, 258], Empirical Potential Structure Refinement (EPSR) [34, 172, 259] are now widely used to interpret high energy X-ray and neutron diffraction data since they provide a 3-dimensional atomic (or molecular model) which exactly fits the measured data within the errorbars. To a lesser extent, disordered crystal models for fitting pressure induced amorphization diffraction patterns have also been successfully employed [260]. RMC works when the interactions in the system are pairwise additive and has been mainly used for studying glasses, amorphous materials, and melts, for example, [66, 70, 261]. The process tends to produce the most disordered structure associated with the measured data and the more constraints that are used the more realistic the model becomes. EPSR was designed primarily for interpreting diffraction data from molecular liquids but uses an empirical potential to drive the fitting process. Both methods can be considered as analogues of the Reitveld fitting process in crystallography and will provide partial structure factors, bond angle distributions, and ring statistics information.

FIGURE 10: The measured X-ray pair distribution function for liquid CaSiO_3 (black solid line, top) compared to that calculated from a molecular dynamics simulation (red dotted line, top) both truncated at a $Q_{\text{max}} = 19 \text{ \AA}^{-1}$ and Fourier transformed (E.T.) with the same Lorch modification function applied. The bottom set of curves shows the individual X-ray weighted partial pair distribution functions calculated directly from the MD simulation box [56].

FIGURE 11: A schematic of the nature of different simulation techniques versus the number of atoms in the model box.

To model the underlying physics of the inter-atomic or intermolecular interactions Molecular Dynamics (MD) [262, 263] or Density Functional Theory (DFT) [264] have commonly been used. Accurate inter-atomic potentials for both oxide and nonoxide glasses have been developed that closely resemble the measured X-ray and neutron structure factors and pair distribution functions (see Figure 10). Hopefully these potentials, which are based on a realistic structural model, may be relied upon to predict reasonable dynamical behavior. Both MD and DFT have been combined with an atomic starting configuration based on RMC models

FIGURE 12: A schematic of my personal view of the birth and development of high energy X-ray diffraction over the past few decades and future opportunities.

[74, 239, 265]. These combined theoretical and experimental approaches represent a major step forward as the electronic structure and bond ordering information can be obtained from the DFT calculation as well as a prediction of the NMR spectrum.

A schematic of the different simulation methods as a function of the number of atoms is shown in Figure 11. These range from detailed quantum many-body method calculations to large scale mesoscopic and macroscopic phenomenological models.

11. Future

A schematic of the past, present, and future developments of high-energy X-ray diffraction from liquids and glasses is shown in Figure 12. The figure identifies some major breakthroughs in the field and predicts opportunities for further growth such as detector and software development.

The future of high energy X-ray diffraction science at 11-ID-C at the Advanced Photon Source lies in the design of complex sample environments for the study of energy-related materials under realistic conditions. The combination of magnetic and/or electric fields, pressure, and low/high temperature as well as time resolved studies is already in progress. The advances in time-resolved measurements will allow chemical reactions in liquids to be followed *in situ* on the beamline. Also, the question of structural heterogeneity between the ergodic and nonergodic regimes during glass formation may now be accessed for a wider range of glass-forming liquids. To achieve this smaller, more intense photon beams are required with energy discriminating large area detectors, which are not currently available. Nanoparticles

have not been mentioned in this paper but as the interface between lengthscales blends together the study of bulk versus surface effects represents another area of research which maybe extended to glasses and amorphous materials at interfaces. Lastly, more sophisticated software is needed to process the huge amount of data generated. Advances in computational scattering science are necessary to make full use of the data we already measure and interpret the ever increasing complexity of our experiments [266].

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