# The Study of Noncrystalline Material under High Pressure Using High Energy X-ray Pair Distribution Function Combined with Synchrotron X-ray Microtomography Techniques

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**Abstract.** For non-crystalline samples, such as amorphous materials and melts, due to their lack of periodicity, scattering patterns from amorphous and liquid phases are very broad. Thus, the pair distribution function (PDF) and X-ray microtomography are important methods to investigate structural change for non-crystalline samples under high pressure conditions. Here we present experimental results on liquid gallium using microtomography and Yb-based metallic glass using PDF under high pressure.

**Keywords:** High pressure, X-ray scattering, liquid-liquid transition, metallic glass.
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## INTRODUCTION

Because of lack of periodicity, scattering patterns from non-crystalline samples, such as amorphous and liquid phases, are very broad. On the other hand, the pair distribution function (PDF) describes the distribution of distances between pairs of particles contained within a given volume. The PDF method provides valuable insights into the local atomic structure underlying functional behavior in crystalline and non-crystalline materials alike; [1] x-ray microtomography can provide density and geometry information on samples regardless of their crystallinity. [2, 3] Thus, the pair distribution function (PDF) and X-ray microtomography are most important method to discuss structural change for non-crystalline samples. Here we exemplify with experiments on liquid gallium using microtomography [4] and Yb-based metallic glass using PDF method under high pressure.

## EXPERIMET ON GALLIUM USING MICROTOMOGRAPHY

The experiment was carried out at 13 BM-D of GSECARS, Advanced Photon Source, Argonne national laboratory. The GSECARS bending magnet beamline provides a 2.5 mrad fan of radiation, permitting in principal studies of objects up to 125 mm in diameter in the experimental station 50 m from the source. The vertical beam size is about 5 mm maximum, due to the narrow opening angle of the synchrotron radiation. The beamline is equipped with a Si (111) fixed-offset double-crystal monochromator, covering the range 5-70 keV. [5] The rotational Drickamer-anvil high-pressure apparatus is shown in Fig. 1. The sample was compressed and heated up to 4 GPa and 330 K, and the experimental data path in P-V diagram is shown in Fig. 2

**FIGURE 1.** High-pressure apparatus in sector 13BM-D.

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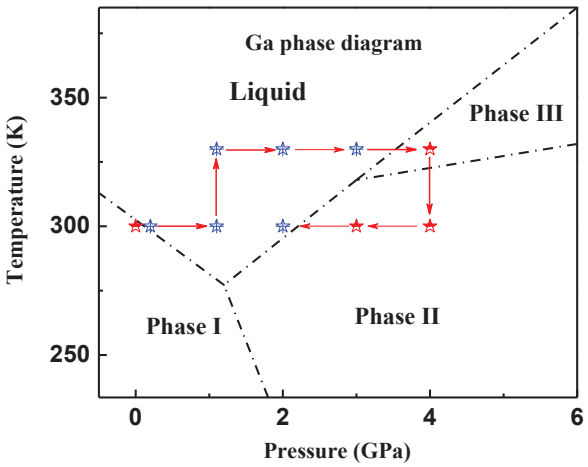


**FIGURE 2.** Pressure-temperature paths for gallium in X-ray microtomography. Phase diagram of gallium is cited from Ref. 6.

## Results

Fig. 3 shows a series of selected reconstructed 3D images of the gallium sample as a function of pressure and temperature. 3D images of each angle of gallium at 0GPa-300K are shown in Fig. 4. The relative volume change with pressure and temperature, for liquid Ga and solid Ga, can be precisely calculated. Relative volume $V/V_0$ of the gallium as a function of pressure and temperature determined by microtomography measurement at 300K and 330 K are presented in Fig. 5.And, a different slope can be observed at 3GPa-330K. This observation suggests that there is a potential liquid-liquid transition.

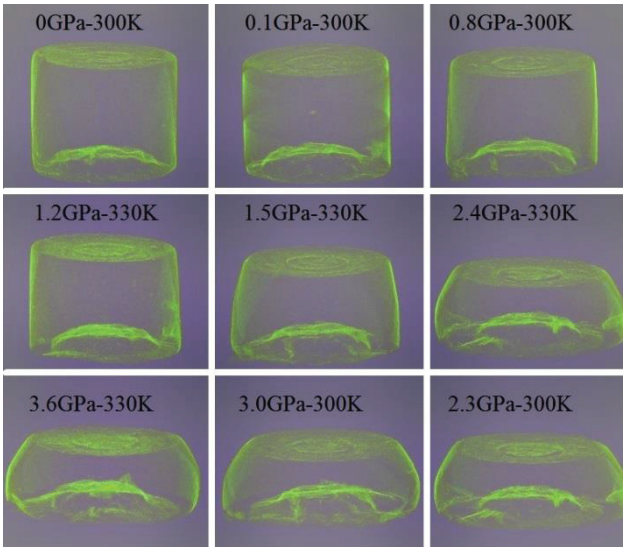


**FIGURE 3.** 3D images for the gallium as a function of pressure and temperature.

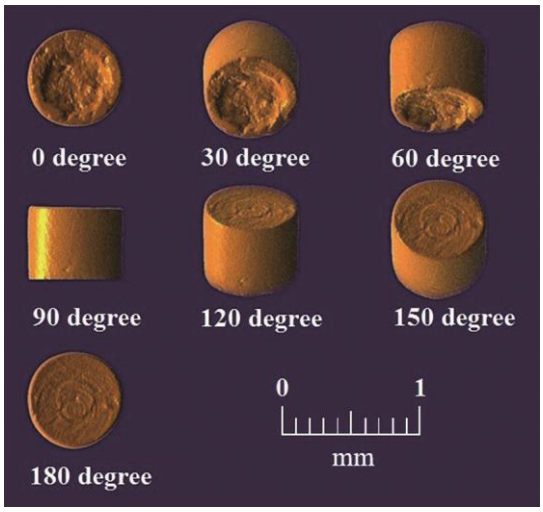


**FIGURE 4.** 3D images of each angle for gallium at 0GPa-300K.

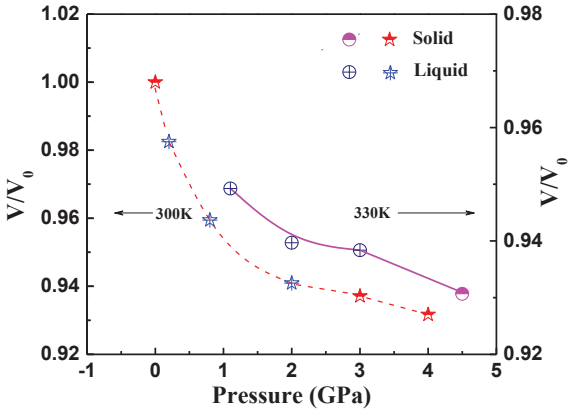


**FIGURE 5.** Relative volume $V/V_0$ of the gallium as a function of pressure and temperature at 300K and 330 K.

Combined the relative volumes of the gallium measured in this work with a starting density, the densities for gallium can be calculated. The density of liquid gallium was calculated as 6.30g/cm3 at 0.8GPa-300K, which is close to the previous value worked by Yagafarov et al. Additionally, at 3GPa-300K, the density of the solid gallium was determined as 6.57g/cm3 which accord with the previously reported density of 6.57 g/cm3 measured at 2.8GPa-298K. All of these agreements illustrate that relative volume measurements from tomography imaging method under pressure indeed provide reliable densities information on liquid and solid gallium.[4, 7-8]

## EXPERIMET ON YB-BASED METALLIC GLASS USING PDF ANALYSIS

The data was collected at sector 11 ID-B, Advanced Photon Source, Argonne national laboratory. The beamline 11 ID-B is located on a 2.3 cm-period undulator, a device that produces high fluxes of high-energy X-rays. It routinely operates at two wavelengths, 0.2128Å (58 keV) and 0.1370Å (90 keV), which correspond to the 311 and 511 reflections from the Si Laue monochromator crystal, respectively. [1] The apparatus performing PDF experiment is shown in Fig. 6. Yb-based metallic glass was measured using PDF method under high pressure up to 2.7GPa.

**FIGURE 6.** The apparatus performing PDF experiment

## Results

S(Q) is Fourier transformed to retrieve real space G(r) information about atomic structure. Vice versa, G(r) is obtained from the S(Q), by a Fourier transformation according to

$$G(r) = 4\pi r[\rho(r) - \rho_0] = \frac{2}{\pi}\int_{Q_{min}}^{Q_{max}} Q[S(Q) - 1]\sin Qr\, dQ$$

where ρ(r) is the microscopic pair density, and $\rho_0$ is the average number density. The Fourier transform is a linear transform, and there is a unique relationship between S(Q) and the resulting G(r). No information is added or removed in the transform, [9, 10] if all relative experimental parameters are in ideal conditions. In practice, what is transformed is the corrected $S_{exp}(Q)$ determined by experiment, which are assumed to be very close to the actual S(Q):

$$G_{exp}(r) = \frac{2}{\pi}\int_{Q_{min}}^{Q_{max}} Q\left[S_{exp}(Q) - 1\right]\sin Qr\, dQ.$$

There is still a unique transformed relationship between $G_{exp}(r)$ and $S_{exp}(Q)$ if the transform is carried out over the same Q-range. [10]

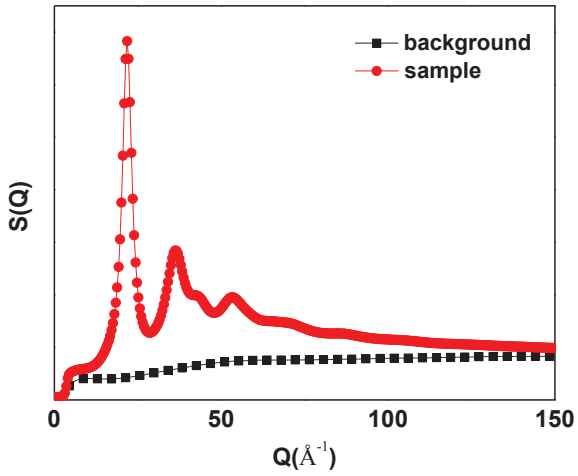


**FIGURE 7.** A selected PDF data S(Q) at ambient conditions.

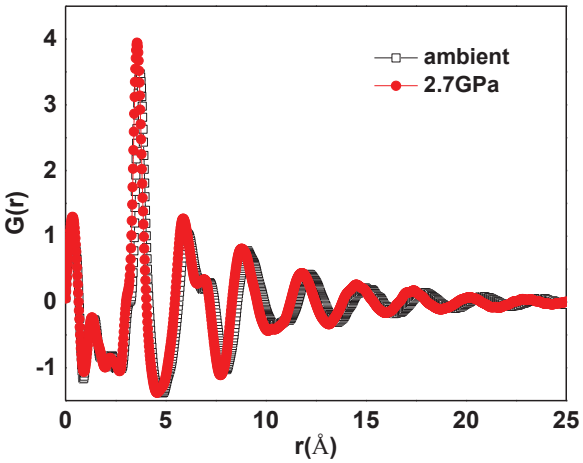


**FIGURE 8.** G(r) of Yb-based metallic glass under ambient pressure and 2.7GPa.

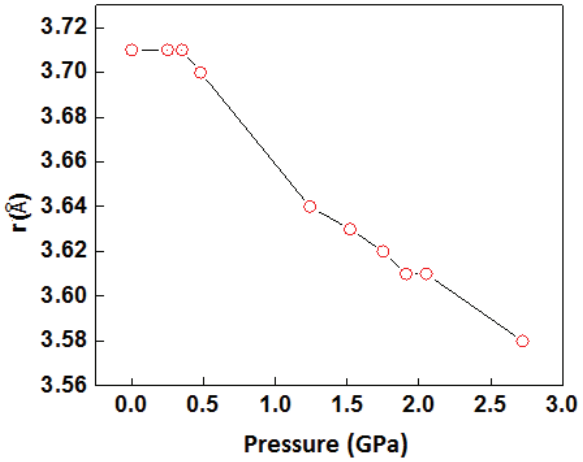


**FIGURE 9.** Relative distance between nearest-neighbor atoms as a function of pressure at ambient temperature.

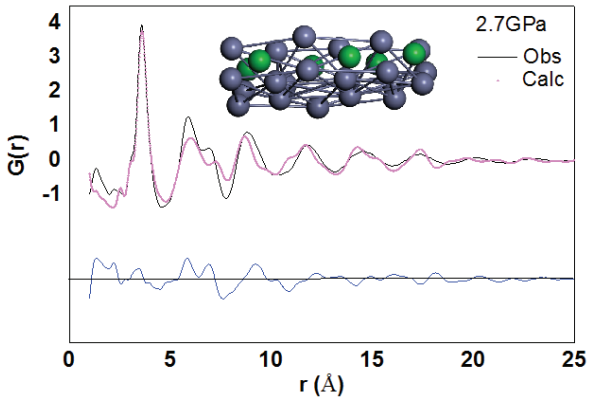


**FIGURE 10.** Experiment date from Yb-based metallic glass (black line) and calculated date using PDFFIT (pink line). Below is plotted a difference curve.

A selected PDF data S(Q) and G(r) are shown in Fig.7 and Fig.8 respectively. The distance between nearest-neighbor atoms can extract from G(r) and the relative distance change with pressure are presented in Fig. 9. A different slope can be observed at 0.4 GPa obviously. A previous work report that Yb-based BMGs do exhibit extremely soft features and strong liquid fragility. [11] However, there is an abnormal phenomenon that Yb-based metallic glass exhibit a hard feature at region of low density compared to that of high density. This may result from change of prosperity due to residual stress change induced by pressure. To get 3D structure from 1D PDF data, we model it using PDFFIT and the result is shown in Fig. 10. The insect of Fig. 10 show the model determined by PDFFIT. The initial configure for fitting is from crystal structure corresponding to Yb-based metallic. A good fitting within 5Å between experiment data and calculated data observed from Fig. 10 imply that the Yb nearest-neighbor coordination environments in Yb-based metallic glass is close to that of crystalline. The peaks of G(r) oscillate around zero and decrease with r increasing and beyond of 18 Å, the oscillation is close to zero. All of these imply that Yb-based metallic glass is median-range order and long range disorder.

## CONCLISIONS

In this work, liquid gallium and Yb-based metallic glass were study using microtomography and PDF methods under high pressure conditions, respectively. The relative volume of the gallium as a function of pressure were observed using microtomography, and obtain the information on volume and density of gallium. Moreover, the distance between Yb atoms as a function of pressure were revealed. Modeling it using PDFFIT, a median range structural information is obtained.

## ACKNOWLEDGMENTS

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