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Trial Measurement of Liquid Density at NE7A

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A suitable modification was made for MAX-III, a multianvil press installed at NE7A, KEK-AR, to measure density of liquids utilizing x-ray absorption. Further improvement for quantitative measurements is now in progress.

1 Introduction

Measurement of density for non-crystalline substances, such as in a liquid or an amorphous state, *is* by far a nontrivial problem. We have tried to detect a change in density upon the phase transition in polyamorphs.

Our study revealed that tin tetraiodide exhibits the water-type polyamorphism. [1,2] It should be emphasized that the liquid-liquid critical point (LLCP) is highly expected to be located in the region centered at around 1.3 GPa and 1000 K. It is still necessary, however, for claiming that SnI_4's LLCP is located above the melting point of the crystalline phase, to detect an abrupt change in density upon compression, although we have verified *the* difference in density, which amounts to only about 0.4 g/cc, between the two liquid phases. [3]

These findings were achieved by the x-ray absorption measurements utilizing the facility installed in SPring-8. Conducting a similar experiment also at the beamlines in KEK would bring considerable benefits to the high-pressure community. This was our initial motivation for undertaking the present study.

2 Experiment

As a trial, we planned to modify the experimental setup of NE7A station of KEK-AR, where a multianvil press, MAX-III, is installed for high-pressure and temperature experiment. The first essential modification was mounting two ion chambers (ICs) just in front of and behind the press to measure the incident and the transmitted x-ray intensities, I_0 and I, respectively (Figs. 1 and 2).

Switching the imaging plate (IP) already equipped with the couple of ICs allows us to measure almost simultaneously x-ray absorption as well as diffraction. It then makes us possible to determine the absolute value for density from a measurement in a solid state.

As depicted in Fig. 3, the sample assembly is a typical one that is used in high-pressure experiments. It should be noted that the sample is located exactly at the mid-point in vertical direction. A gap between anvil blocks becomes narrower with increasing load than the diameter of the sample container. In order for the sample to be scanned across an x-ray beam, a ditch was made at the center on the anvil surface. The sample has to be located at the height of the ditch.

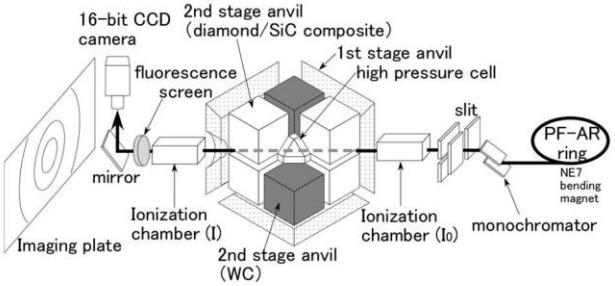

Fig. 1: The design allowing x-ray absorption and diffraction by switching the ICs and the IP.

Fig. 2: A close view of the sample stage from the incident-beam side. The IC on this side is I_0 in Fig. 1, and the IC on the other side is I. Anvil blocks are seen between the two ICs.

3 Results and Discussion

Figure 4 shows the absorption profiles obtained on a first trial. The incident beam was monochromatized to 0.2755 Å. The distortion of the profiles was found to be more or less improved by resetting the sample. Using the method described in [3], we could obtain at 2.2 GPa and room temperature (RT) the value, 3.94(17) g/cc, for density, which is appreciably smaller than 5.31 g/cc obtained through diffraction. The large discrepancy is ascribable to the ambiguity of the edge in the profile originating from the beam divergence in transvers direction. The latter effect is being tried to be deconvoluted. We also expect that a method based on the

maximum entropy method [4] will be effective for the estimation of the location of the edge.

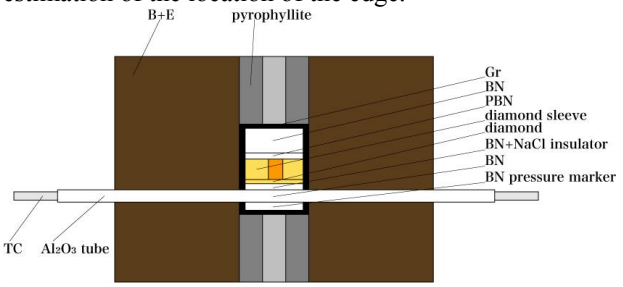

Fig. 3: Sample assembly used in the present experiment.

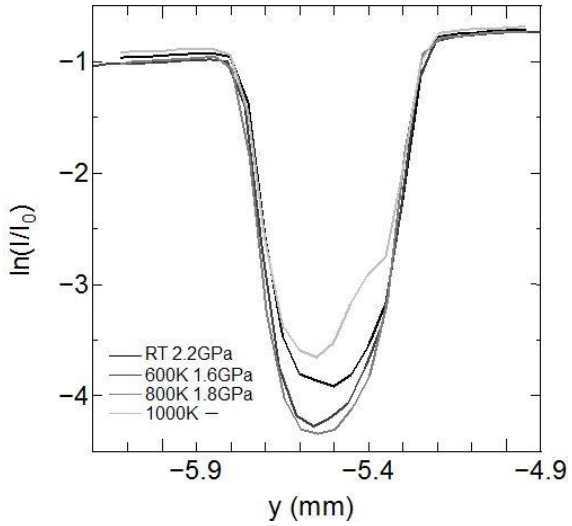

Fig.4: The ratio, I/I_0, is plotted as a function of displacement, y, perpendicular to the beam axis.

Table 1: Density of liquid SnI_4 obtained from the absorption profiles shown in Fig. 1.

temperature (K)	density (g/cc)
RT	3.94(17)
600	4.32(19)
800	4.47(8)

The x-ray absorption profiles obtained at other temperatures were similarly processed. The resulting density (including the one at RT) is listed in Table 1. Unusual temperature dependence is readily noticeable; the density increases with temperature. This unusual behavior should be ascribed not to intrinsic nature of liquid SnI_4 but to the quality of the profile. Another important reason of the poor reproducibility of the density at RT is the asymmetric feature. The profile should be symmetric in y-direction, and the analysis was made assuming the symmetry. As clear from Fig. 1, the profile recovered the expected symmetry with increasing temperature. It is anticipated in this context that the density at 800 K is close to the actual density. At 1000 K, however, the profile became irregular again. This happened because the cover (made of PBN, see Fig. 3) of the sample container penetrated into the measuring point.

The present project started in November, 2013. The measurement under the present proposal was done during June 14 through 17, 2014. Unfortunately, however, no beam time was provided in the latter half period of that year. A follow-up experiment has just been carried out, and much progress has been made. This was brought about by the improvement in the system of running gears to drive the press stage. Professor A. Suzuki took care of this improvement. Software with which to drive the system was prepared by Professor Y. Mori. The results obtained from the last measurement will be reported in the near future.

Acknowledgement

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