NE5C/2008G078

Variation of the local structure in liquid SnI_4

Kazuhiro FUCHIZAKI*[1], Takahiro SAKAGAMI[1], Takaki HASE[2]
Yuki YAMAGATA[3], Nozomu HAMAYA[3]
[1]Department of Physics, Ehime University, Matsuyama 790-8577, Japan
[2]Altekna Co. Ltd, Nagoya 451-6004, Japan
[3]Department of Physics, Ochanomizu University, Tokyo 112-8610, Japan

Introduction

We have performed in situ synchrotron x-ray diffraction measurements on tin tetraiodide, which consists of SnI_4 tetrahedral molecules at ambient pressure, and established that the liquid forms existing above and below 1.5 GPa, where the slope of the melting curve of the solid phase changes abruptly, have different structures. This discovery offers evidence of thermodynamically stable polyamorphism in general compounds as well as in elements. A possible phase diagram that includes the two amorphous states already found is proposed based on the pseudo-binary regular solution model. The vertex-to-face orientation between the nearest molecules plays a key role in the transition from the low-pressure to the high-pressure liquid phase [1].

Our next task is therefore to judge whether there is a sharp transition between the two liquid states. To this end, in situ measurements have been performed along thermodynamic paths that pass through/near 1.5 GPa. A part of the findings will be presented elsewhere [2]. The other aspect is reported here.

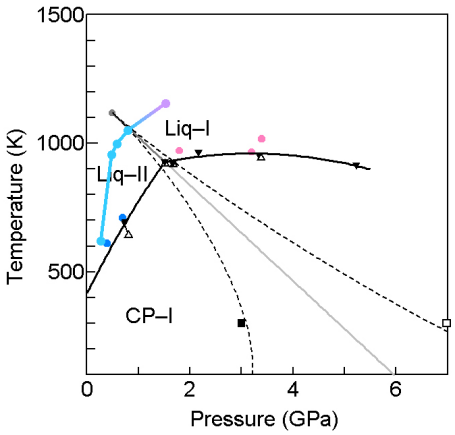

Fig. 1 One of the experimental paths crossing across the expected phase boundary between the two liquid phases, Liq-II and Liq-I. Regarding the symbols, see [1].

Experimental method

The energy-dispersive x-ray diffraction experiments were performed using synchrotron radiation. MAX80, a cubic-type multianvil press for high-pressure and high-temperature experiments, installed in BLNE5C at KEK-AR, measured *in situ* x-ray scattering at pressures and temperatures up to about 2 GPa and 1200 K, respectively. A typical thermodynamic path along which the measurements were made is depicted in Fig. 1. Tungsten carbide anvils with a center flat of 6mm were employed to generate high pressures. The difference from the previous sample assembly [1] is to place two sample containers (made of diamond) symmetrically in vertical direction. One contained the sample, SnI_4, and the other a standard pressure calibrant, NaCl, thereby estimating pressure at the sample position.

Results and Discussion

Scattered intensity distributions measured at various scattering angles were converted into the Faber-Ziman structure factor following a standard procedure [1]. The latter was then inverted via a newly devised method [3] to the reduced radial distribution function, from which real-space information regarding the liquid structure was inferred. A local order parameter (LOP) that can quantify the regular tetrahedrality was defined. The way of variation of the LOP around 1.5 GPa was estimated from the LOPs measured along the path, and is depicted in Fig.2. Currently, the number of data points is too small to allow detailed nonlinear regression for the LOP surface, although the behavior of the LOP seems to support the existence of the transition line around 1.5 GPa. Further measurements should be carried out to draw a definite conclusion.

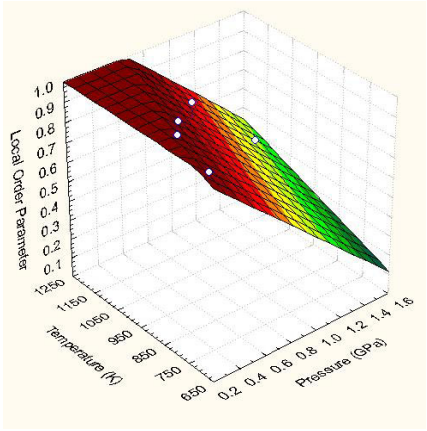

Fig. 2 Variation of LOP along the path (white dots). The surface was constructed by a linear regression.

References

[1] K. Fuchizaki et al., J. Chem. Phys. **130**, 121101(2009).
[2] N. Hamayar et al., to be presented at AIRAPT 2009.
[3] K. Fuchizaki et al, J. Chem. Phys. **127**, 064504 (2007).
* fuchizak@phys.sci.ehime-u.ac.jp