

Pressure-induced structural change of liquid IV-VI alloys

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Introduction

The crystalline structure of group-V elements under pressure has recently been an interesting subject of research[1]. A theoretical study[2] suggested a possible first-order transition due to the suppression of the "Peierls distortion" in the liquid under pressure. To clarify how "Pressure-induced suppression of Peierls distortion" take place in liquids, we have experimentally examined the pressure dependence of the structure of liquid (l-) As, l-GeS, l-GeSe, and l-GeTe.

The crystalline structure of As and GeTe at ambient condition is A7 structure, whereas GeS and GeSe take A17 structure. (Therefore, it is B16/GeS structure if we distinguish between two types of atoms.) The important similarities between A7 and A17 are as follows: (1) 6 nearest neighbors (NN's) for simple cubic structure split into 3 nearest neighbors and 3 next-nearest neighbors (6 NN atoms become 3+3), and (2) the bond angle becomes slightly larger than 90 degree, whereas it is 90 degree for the not-distorted simple cubic structure. (3) In addition, these "Peierls distortion" is known to be generally suppressed by increasing pressure (3 NN +3 next-NN atoms return to become 6 NN's by compression).

The structures of these liquids had been shown to be "Peierls-distorted" near the ambient pressure. We thus expected "Pressure-induced suppression of the Peierls distortion" for these liquids.

Experimental

Synchrotron x-ray diffraction measurements were performed by use of the high-pressure apparatus MAX80 installed at the beam-line NE5C in PF-AR, KEK. We also used the SPEED1500 installed at BL04B1 in SPring-8 for the conditions above 10 GPa. Measurements were done at temperatures approximately 50K above the melting point at each pressure.

Results

Figure 1 shows $S(Q)$ for l-As at high pressures. We estimated the pair distribution function $g(r)$ for each liquid from each $S(Q)$. The pressure dependence of the ratio R_2/R_1 is shown in Fig. 2. Here R_1 and R_2 are the first and second peak positions of $g(r)$, respectively. At low pressures the ratio is higher than $\sqrt{2}$ which is shown by the horizontal dotted line in Fig. 2. This means that the bond angle is larger than 90 degrees. Importantly this ratio decreases with pressure, and changes the slope when it passes $\sqrt{2}$ at around 10GPa. This feature at 10GPa can be regarded to be the "pressure-induced suppression of the Peierls distortion" in this liquid As

because the ratio is $\sqrt{2}$ for not-distorted simple cubic structure.

We observed the same tendency for three liquid compounds as shown in Fig. 3. In Fig. 3, the value $\sqrt{2}$ is shown by the solid horizontal lines and the arrows show the point when the ratio passes $\sqrt{2}$. This pressure shown by the arrows in Fig. 3 was found to be the largest for l-GeS and the smallest for l-GeTe. We thus found that the "Pressure-induced suppression of the Peierls distortion (PISPD)" takes place for all these liquid compounds systematically. We note that this systematic PISPD does not occur in the crystalline phase for these compounds because of the different way of Peierls distortion between A7-type and A17-type.

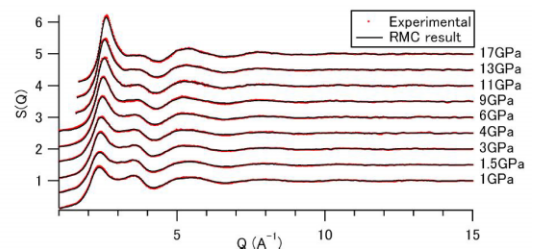


Fig. 1: $S(Q)$ of l-As at high pressures. Red dots show the experimental data and black lines show the RMC results.

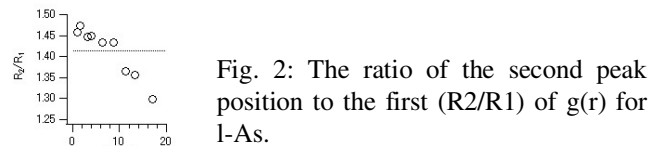


Fig. 2: The ratio of the second peak position to the first (R_2/R_1) of $g(r)$ for l-As.

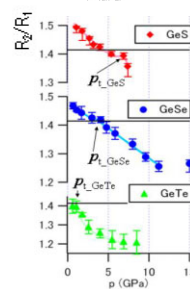


Fig. 3: R_2/R_1 for three liquid compounds. The horizontal lines show $R_2/R_1 = \sqrt{2}$, which is the value for simple-cubic structure.

References

- [1] For example, see Y. Katayama et al., Nature 403, 170 (2000).
- [2] X.-P. Li, Phys. Rev. B 41, 8392 (1990).

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