

Pressure-induced electronic transition in liquid Rb

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Introduction

Rubidium is considered typical free-electron metals because of the single s electron in the valence bond. However, this simple element shows unexpected behavior when it is subjected under pressure. For example, x-ray diffraction studies have shown that it undergoes a sequence of phase transition leading to low symmetry instead of close packed structure [1]. The pressure-induced complexity is attributed to the increase role of the d electrons becoming more appreciable with increase in electron density. Such behavior would occur not only in crystalline phases but also in liquid states. Actually, the melting curve shows maxima around 2 GPa and the deviation of the contraction behavior from uniform contraction model was reported at the higher pressure [2]. However, it is subject of debate because of a lack of the structural data at pressures where the s - d electronic transition occurs. The lack stems from the experimental difficulty due to the high reactivity of alkali metals as well as the highly fugitivity of the sample. To avoid these problems, we have developed externally heated diamond anvil cell (DAC), which prevents the reaction and leakage of the sample. We investigate the pressure-induced change of liquid Rb up to about 12 GPa using this device on the basis of the afore-mentioned motivation.

Experimental

The schematic view of the high-pressure cell is shown in Fig. 1. The externally heated DAC is equipped with the coil heater located inside the piston of DAC and the larger heater surrounding the piston. We put the sample inside the hole of the Re gasket without any pressure medium or pressure marker to avoid reaction of the sample. We controlled the pressure from the outside of the experimental hutch by regulating pressure of the gas loaded in the bellows which is attached to the body of DAC. This system enables us to prevent the pressure decrease on heating as well as the change of the sample position during compression or decompression.

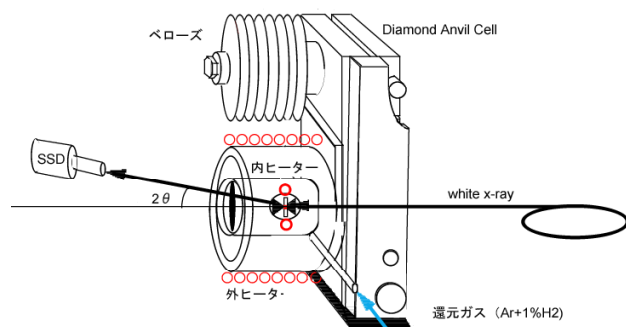


Fig. 1 Schematic view of the externally heated diamond anvil cell equipped with a gas pressure controller.

Pressure was estimated from the position of the main peak Q_1 in the diffraction pattern on the basis of the results of the previous study [2]. The pressure in the region where the investigation has not been conducted was estimated from the extrapolated curve. The x-ray diffraction intensity was measured by an energy-dispersive method using a pure-Ge solid-state detector.

Results and Discussion

The relation between the position of the main peak Q_1 and the full width at half maximum in diffraction patterns at high pressures is shown in Fig. 2. With increase in Q_1 up to 1.85 (with increasing pressure up to 4 GPa), the FWHM remains almost constant. However, the value increases on the further increase in Q_1 . When the liquid contracts uniformly the value should become almost constant because in such case all the profile at high pressures can be scaled by a Q value. This finding suggests that the local structure change of above 4 GPa. The high-pressure sequence in crystalline Rb helps us understand this behavior. When the pressure-induced s - d transition occurs, the average atomic radius becomes smaller because of the smaller d -orbital compared s -orbital. In the crystalline phase, the contraction of the atomic radius takes place discontinuously. In contrast, such change occurs gradually in liquid since the local structure gradually changes over wide pressure region. In the transient states, two species with different atomic diameter coexist and therefore, the main peak becomes broader. The increase in FWHM observed above 4 GPa implies the occurrence of s - d transition in liquid Rb, and the s - d transition does not complete at 12 GPa, which corresponds to $Q_1 = 2.1 \text{ \AA}^{-1}$.

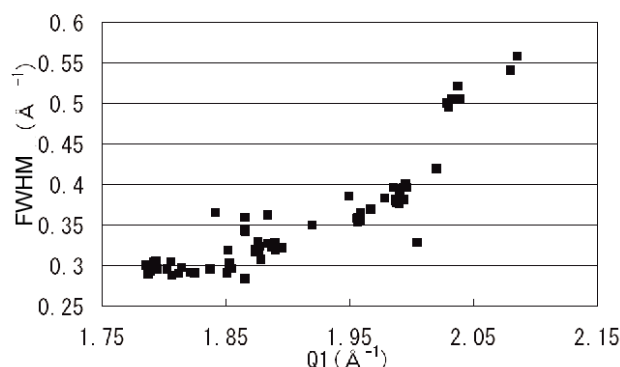


Fig. 2 The relation between Q_1 and the FWHM in diffraction patterns at high-pressures.

References

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- [2] Tsuji et al., J. Non. Cryst. Solids 205-207, 295 (1996).

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