High Pressure Fluorescence Ce-$L_{III}$ XANES Measurement and Its Application

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Introduction

Anomalous physical properties (e.g. dense Kondo state) in Ce alloys and compounds are very sensitive to changes in external pressure. Since the anomalies are attributed to partial delocalization of $4f$ electrons of Ce, it is important to estimate the Ce valence for compounds under high pressure. We have been developed a new technique for Ce-$L_{III}$ x-ray absorption near-edge structure (XANES) measurement using a diamond anvil cell (DAC) [1]. In this study, the changes in electrical resistivities and Ce valences in β-Ce$_3$Al alloy were measured as a function of external pressure in order to understand the correlation between appearance of the dense Kondo state and the Ce valence.

Experimental

The temperature dependence of electrical resistivity under various pressures was obtained with a DAC cooled in liquid N$_2$. The detailed procedure was described elsewhere [2].

High pressure Ce-$L_{III}$ XANES spectra were measured at the facilities of BL-12C in the Photon Factory, KEK. Since the x-ray beam around Ce $L_{III}$-edge (~5.7 keV) can hardly penetrate diamond anvils of a conventional DAC, we proposed a new optical geometry for fluorescence Ce-$L_{III}$ XANES measurement with an asymmetrical pair of diamond anvils [1].

Results and Discussion

Figure 1 shows the temperature dependence of the electrical resistivity $\rho(T)$ for β-Ce$_3$Al alloy under various pressures. Under ambient pressure, $\rho(T)$ decreases almost linearly with temperature. For high pressure, the curve of $\rho(T)$ shows a negative logarithmic behavior and the negative temperature coefficient of resistivity (TCR) increases remarkably with pressure. The absolute value of the TCR at 2.3 GPa is almost equivalent to those of Pd$_3$Ce alloy at ambient pressure and of PdCe alloy at about 4 GPa. This indicates that the dense Kondo effect in β-Ce$_3$Al alloy is rapidly strengthened with external pressure.

Figure 2 shows fluorescence Ce-$L_{III}$ XANES spectra of β-Ce$_3$Al alloy under various pressures. It is clearly seen that the spectra exhibit only a single line assigned to trivalent $4f^1$ configuration. The profiles of those spectra quite agree with one another. This suggests that the Ce valence in β-Ce$_3$Al alloy holds about 3.0 up to 7.7 GPa. The present authors [3] found that the Ce valences in PdCe and GaCe alloys increase with external pressure and then the dense Kondo states in these alloys are remarkably enhanced by the pressure. We [4] also found that in the amorphous Ce$_3$Al alloy the dense Kondo effect is rapidly weakened with external pressure though the Ce valence increases with pressure. It is possible that the dense Kondo state in β-Ce$_3$Al alloy is pronounced without variation of Ce valence, or the electron transition of $4f^1 \rightarrow 4f^0$. In order to obtain more conclusive explanation of this complex phenomenon, more detailed measurements for Ce$_3$Al under high pressure are now in progress.

References


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