L_3-edge XAFS measurements of valence fluctuating Eu compounds

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Introduction

It is known that some Eu compounds show a valence fluctuation like Ce and Yb compounds. The valence fluctuation in Eu compounds takes place between non-magnetic Eu^{3+} and Eu^{2+} with a localized moment of $7\mu_B$. One of great interest in the valence fluctuation in Eu compounds is that the mean Eu valence strongly depends on temperature, magnetic field and pressure[1,2]. In this study, we take notice of $EuCu_2(Si_xGe_{1-x})_2$ system which was reported to exhibit a Kondo-lattice type state by Levin et al. for the first time [3]. Recently, heavy fermion behavior has been reported by Hossain et al. [4]. In order to discuss a correlation between such phenomena and the valence, we have investigated the mean Eu valence as a function of temperature by measuring L_3-edge XAFS.

Experimental

The polycrystalline samples were prepared by arc-melting under argon atmosphere and subsequent annealing in an evacuated quartz tube at 1173 K for 1 week. The XAFS measurements at the Eu L_3 edge were performed at BL-9A beamline of KEK Photon Factory using a Si(111) double crystal monochromator in the temperature range from 10 K and 300 K.

Results and discussion

All of the measured XAFS spectra at the Eu L_3 edge of the samples consist of two subspectra, the $(2p^5 4f^7 5d^*)$ and $(2p^5 4f^6 5d^*)$ final state components, as shown in Fig. 1. This directly indicates the valence fluctuating behavior. The spectra were analysed by fitting two sets of a Lorentzian and an arctangent-function. The mean valence is estimated from the relative intensity of the two subspectra. Figure 2 shows the mean valence of the samples as a function of temperature. For x 0.65, the valence shifts toward the Eu^{3+} state with decreasing temperature. For x=0.70-0.80, the T_0 means the temperature where magnetic susceptibility of the samples begins to deviate from a Curie-Weiss law. The valence at T_0 is found to be an almost common value of ~2.4, which appears to be a boundary between a non-magnetic Eu^{3+} and a magnetic Eu^{2+} characters. The valence at the lowest temperature has an intermediate value of 2.5-2.65. Similar behavior is observed also in $EuNi_2Si_2$, which has a large electronic specific heat coefficient of γ~100mJ/K^2 mol. The intermediate valence is possibly associated with the heavy fermion behavior.

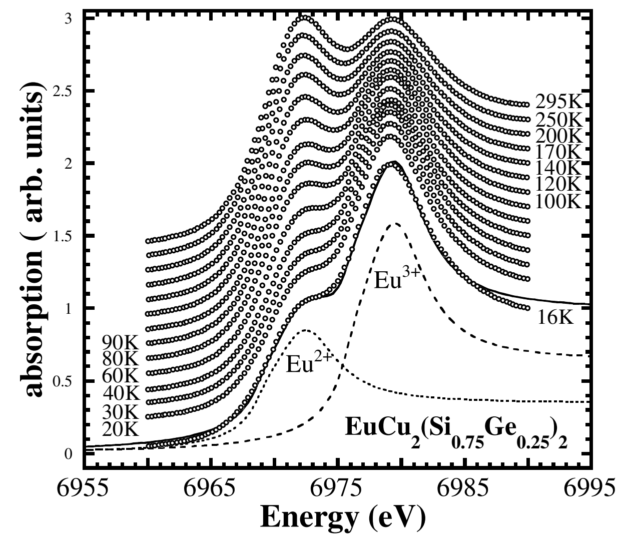

Fig. 1 The XAFS spectra at the Eu L_3-edge at various temperatures.

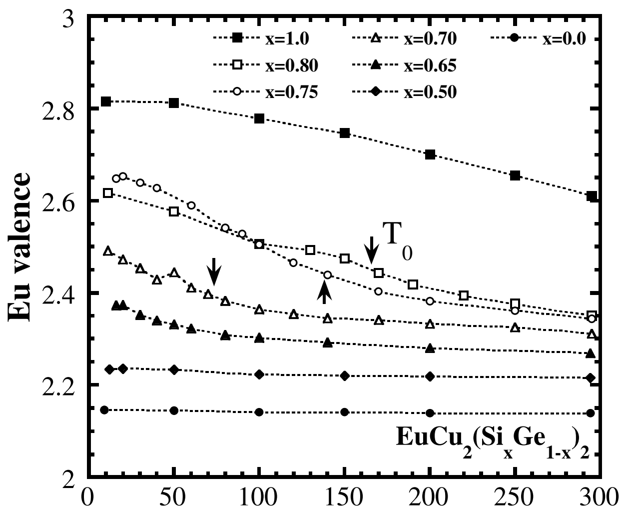

Fig. 2 Temperature dependence of the mean Eu valence.

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