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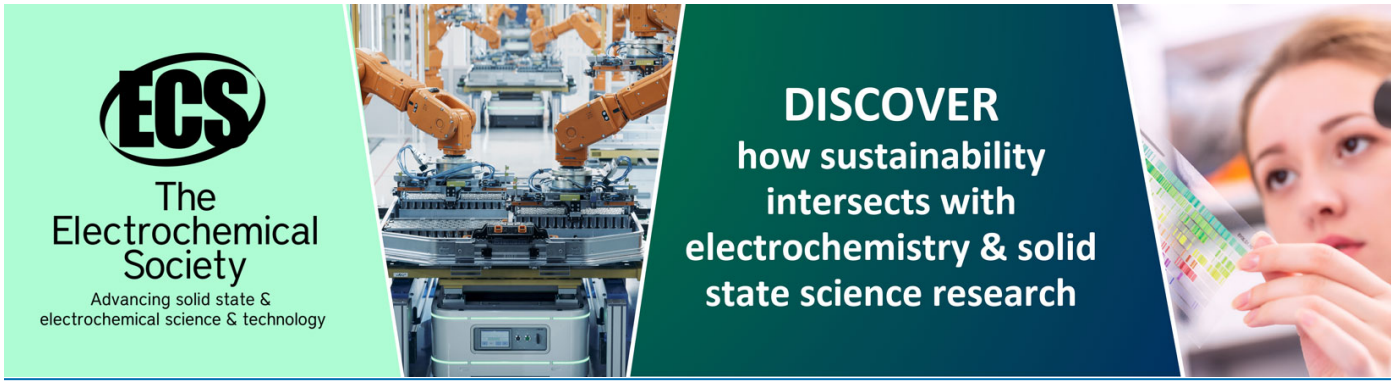

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Electronic states in the pressure-induced magnetically ordered phase in SmB_6

Takeshi Mito[1], Naoya Emi[1], Naomi Kawamura[2], Masaichiro Mizumaki[2], Takehide Koyama[1], Koichi Ueda[1], Naoki Ishimatsu[3], and Fumitoshi Iga[4]

[1] Graduate School of Material Science, University of Hyogo, Kamigori, Hyogo 678-1297, Japan
[2] Japan Synchrotron Radiation Research Institute, SPring-8, Sayo, Hyogo 679-5198, Japan
[3] Graduate School of Science, Hiroshima University, Hiroshima 739-8526, Japan
[4] Faculty of Science, Ibaraki University, Mito, Ibaraki 310-8512, Japan

E-mail: mito@sci.u-hyogo.ac.jp

Abstract. We have carried out the high-pressure measurement of X-ray absorption spectroscopy on the intermediate valence compound SmB_6 which shows magnetic ordering as well as an insulator-metal transition at critical pressure $P_c \sim 10$ GPa. The valence of Sm atom at room temperature increases with increasing pressure, however it is far below a trivalent state at P_c. In contrast to cases of pressure-induced nonmagnetic-magnetic transition in Yb compounds, which mostly occurs in the scheme of well localized $4f$ electrons, the present observation suggests that electronic system in SmB_6 still possesses strong delocalized characters at P_c.

1. Introduction

Rare-earth based compounds show variety of ground states. The ground states are often varied to another by changing pressure or magnetic field, resulting in for example a transition between nonmagnetic and magnetic states, between metallic and semiconducting states, and so on. In Ce, Sm, Eu, and Yb compounds, the valence of rare-earth ions plays a key role to control their features. According to the ground state of Hund's rule, trivalent (divalent for Eu) states show magnetic features, while divalent (trivalent for Eu and tetravalent for Ce) states are nonmagnetic. Therefore a change in the valence intimately relates to a transition between nonmagnetic and magnetic ground states in these compounds.

Pressure-induced nonmagnetic-magnetic transitions in Yb compounds have been investigated so far. In the Yb systems, pressure tends to induce the localization of $4f$ electrons from the so-called intermediate valence state. The X-ray absorption spectroscopy (XAS) is a powerful technique to measure the mean valence of the rare-earth ions even under high pressure. According to the results of the high-pressure XAS measurements performed on various Yb compounds (for example, see Ref. [1, 2, 3]), most of the pressure-induced magnetic orders appear with mean Yb valence close to 3+, namely in the scheme of well localized $4f$ electrons.

It is also known that some Sm compounds undergo a nonmagnetic-magnetic transition under high pressure. Therefore, it is useful to compare the effects of pressure on the two different systems in order to study the nonmagnetic-magnetic transition in the rare-earth compounds systematically. SmB_6 is a well-known intermediate valence compound having a

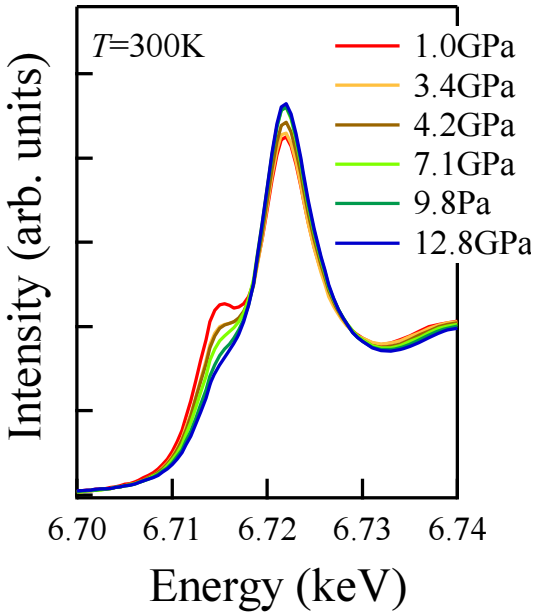

Figure 1. Sm L_3-edge absorption spectra of SmB_6 measured at 300 K and at 1.0, 3.4, 4.2, 7.1, 9.8, and 12.8 GPa.

small semiconducting gap of $50-100$ K [4, 5]. The formation of the gap may be associated with hybridization between conduction and $4f$ electrons, however the details are still controversial. Recent nuclear forward scattering, specific heat, and resistivity measurements under pressure in SmB_6 give evidences for the appearance of magnetic ordering simultaneously with an insulator-metal transition at critical pressure $P_c = 7-10$ GPa [6, 7, 8]. No structural anomaly has been found so far under pressure [13]. However, the detailed electronic states at the transition, especially whether $4f$ electrons are in a well localized state just like in many Yb compounds, have not been clarified yet. We have therefore carried out XAS measurements in order to investigate the pressure dependence of Sm valence in SmB_6.

2. Experimental details

Single crystalline samples of SmB_6 were grown by a floating-zone method using an image furnace with four xenon lamps [9]. The XAS measurements near the Sm L_3-edge (6.72 keV) were performed in the pressure range of 1 to 13 GPa at room temperature, at the beamline BL39XU of SPring-8, Japan [10]. For the high pressure measurements, the sample was loaded in a diamond anvil cell (DAC) filled with a mixture of 4 : 1 methanol : ethanol as a pressure-transmitting medium. Nanopolycrystalline diamond anvils were used to avoid glitches in XAS spectra [11]. The pressure was calibrated using fluorescence from ruby chips mounted with the sample inside the DAC.

3. Results and Discussion

Figure 1 shows the Sm L_3-edge absorption spectra of SmB_6. The shoulder like structure at 6.715 keV, corresponding to a divalent component, is gradually suppressed with increasing pressure. However, a finite spectral weight persists even at the maximum pressure of the present experiment. Simultaneously, the intensity of the main peak at 6.72 keV, corresponding to a trivalent component, increases with pressure. These results clearly indicate that pressure brings

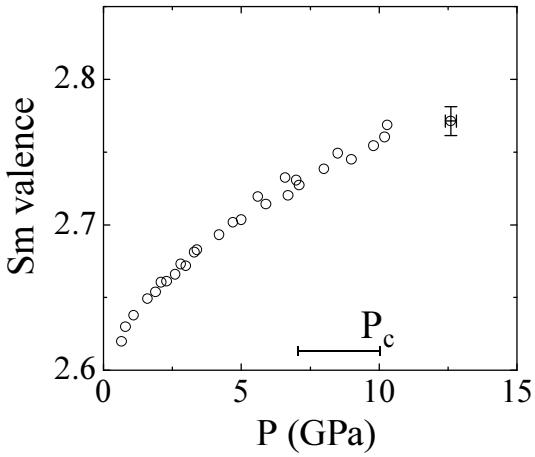

Figure 2. Estimated mean Sm valence at 300 K as a function of pressure. P_c somewhat distributes depending on the experiments [6, 7, 8].

about a shift of the Sm valence toward the trivalent state, consistent with the results of previous Raman scattering and nuclear magnetic resonance measurements [12, 13].

The mean valence of the Sm ions is estimated from the relative intensities of the Sm^{2+} and Sm^{3+} components in the XAS spectra shown in Fig. 1. Each component was modeled by the sum of a Lorentz function and an arctangent function representing the continuum excitations. The evaluated Sm valence v_{Sm} is illustrated in Fig. 2 as a function of pressure. As pressure increases, v_{Sm} increases and the slope dv_{Sm}/dP gradually decreases. We here compare the present observations with various cases of pressure-induced nonmagnetic-magnetic transition in the Yb compounds (for example, see Ref. [1, 2, 3]). The weak saturation of v_{Sm} at high pressures in SmB_6 is analogous to the pressure dependence of Yb valence in the Yb compounds. However, a striking contrast between the two systems is observed in the valence at the critical pressure: at the nonmagnetic-magnetic transition in the Yb compounds, the mean Yb valence is very close to 3+, while v_{Sm} in SmB_6 is far below 3+, *i.e.* $v_{\mathrm{Sm}} < 2.8$ at P_c. The result indicates that electronic system in SmB_6 still possesses strong delocalized characters at P_c. The remarkable difference in the valence of the rare-earth atoms should arise from underlying electronic structures and correlations unique to each rare-earth compound at the critical pressure.

4. Summary

We have carried out XAS measurements on the intermediate compound SmB_6 in the pressure range up to 13 GPa at room temperature. The Sm valence increases with increasing pressure, however the valence is far below the trivalent state at the critical pressure where magnetic ordering occurs. With respect to the mean valence of rare-earth atoms, the present observation in SmB_6 strikingly contrasts with what is mostly observed in the Yb compounds: namely the Yb valence is very close to 3+ at critical pressure. The result suggests that electronic system in SmB_6 still possesses strong delocalized characters at P_c.

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