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Magnetic Field Effect on Yb-Based Heavy Fermions near Magnetic–Nonmagnetic Transition

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Recently magnetic field effects on f electron systems in the vicinity of a magnetic–nonmagnetic transition have attracted much interest. In order to investigate the mechanism of field induced modifications in heavy fermion systems, especially in terms of valent degrees of freedom, we have carried out the X-ray absorption spectroscopy measurement on the newly discovered heavy fermion material $YbCo_2Zn_{20}$ which exhibits extremely large specific heat at low temperatures. Our results show that the Yb valence of $YbCo_2Zn_{20}$ is very close to +3 over the whole temperature range of measurement. Besides, no significant change in the absorption spectrum was observed up to the field of 10 T. This contrasts to the previously reported strong field dependence in the resistivity, followed by an expansion of the Fermi liquid regime with increasing field. The obtained results therefore lead to the conclusion that the valent degrees of freedom play a minor role in the strong field dependence of the heavy fermion systems in this compound.

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1. Introduction

Many attempts to approach magnetic–nonmagnetic transitions have been made in rare-earth based heavy fermion (HF) materials, for example application of pressure, magnetic field, or chemical doping, because various unconventional properties have been observed in the vicinity of this phase boundary. Among them, recent reports of a magnetic field induced quantum critical point in ytterbium (Yb) based HF compounds have attracted much interest [1, 2]. Those studies indicate that, after antiferromagnetic ground state is suppressed by field, Fermi liquid (FL) like behavior appears and is enhanced with increasing field. However, the field effect on $4f$ electronic states near the magnetic–nonmagnetic transitions is not simple, because it simultaneously affects several degrees of freedom with small characteristic energies, for example interactions between conduction and f electrons, crystalline electric field, valence of rare-earth ions, and so on. Besides, recent theoretical works pointed out that valence fluctuations play an important role in peculiar properties in magnetic field observed in the HF materials [3]. However, from experimental point of view, it is not easy to extract the field effect on the valence. For example, in the above mentioned field induced FL states in $YbRh_2Si_2$, with increasing field, the temperature dependence of the susceptibility behaves as if $4f$ electron system became more itinerant [4], namely analogous to the behavior typically observed in mixed valence states than in a well localized trivalent Yb state. Actually, the field effect on the valence near the magnetic–nonmagnetic phase boundary has not been studied in detail so far.

In this paper, we report the studies of the X-ray absorption spectroscopy (XAS) measurement, which is known as a powerful tool for determining valence states of ions, on the HF compound $YbCo_2Zn_{20}$, and present how the Yb valence depends upon the field. In contrast to what is observed in cerium (Ce)-based HF compounds, most of the Yb-based HF materials do not show superconductivity in the vicinity of the magnetic–nonmagnetic phase boundary, which is rather advantageous in terms of investigating the criticality down to very low temperatures. $YbCo_2Zn_{20}$ crystallizes in the cubic $CeCr_2A_{20}$ structure with Yb–Yb distance ≈ 6.0 Å. One of the most unique features of this compound is an extremely large specific heat at low temperatures ($C/T = 7.9$ J/(mol K^2)), whose origin has not been clarified yet, with no sign of long range magnetic order down

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to the lowest temperature [5]. Besides, pressure induced magnetic ordering with a low critical pressure of ≈ 1 GPa is suggested by the resistivity and specific heat measurements [6, 7]. These facts indicate that this compound locates very close to the magnetic–nonmagnetic border at ambient pressure.

2. Experimental

Single crystal samples of $YbCo_2Zn_{20}$ were grown using the Zn self-flux method as reported previously [6]. The single crystal sample was finely powdered for the XAS experiments. The measurements were performed in the temperature range of 2 to 300 K and in the field range of 0 to 10 T at a beam line BL39XU of SPring-8 in Japan. The XAS spectra at the Yb L_3-edge were recorded in the transmission mode using ionization chambers. The energy was calibrated with the XAS spectrum of Yb_2O_3 at room temperature.

3. Results and discussion

Figure 1 shows the L_3-edge absorption spectra in zero field measured at 286 and 2 K. The main peak at 8.947 keV corresponds to the trivalent component. The signal for the divalent state, which generally appears around 8.939 keV [8], is not clearly observed in the present spectra. The Yb valence therefore should be very close to +3, although the precise absolute value could not be determined because very little spectral weight of the divalent component led to the difficulty in fitting the data. When temperature is lowered from 286 to 2 K, there is no difference in the lineshape within experimental error. This is consistent with the fact that the susceptibility of $YbCo_2Zn_{20}$ follows the Curie–Weiss law down to 2 K with an effective moment near the full free-ion moment of Yb^{3+} [5, 7]. We also note that, in the temperature dependence of C/T, the FL behavior with the heavy mass is seen only below 0.2 K [5, 7].

We have found that the absorption spectrum is insensitive to the application of field. As shown in Fig. 2, the spectrum measured at 2 K does not reveal any changes in shape when the field is applied up to 10 T, implying that we observed no evidence that the original Yb trivalent valence changes in the magnetic field. Here the resolution of the absorption spectrum corresponds typically to the valence of 0.01 or less.

This trend obviously contrasts to strong field dependences observed in the resistivity and the susceptibility (M/H). As field increases, the FL regime in the resistivity, where the resistivity ρ follows $\rho = \rho_0 + AT^2$, rapidly extends to higher temperature range, and the A value becomes about 10^2 times smaller at 6 T [9]. M/H deviates downward from the Curie–Weiss law with increasing field above 1 T (M/H at 7 T is $\approx 50\%$ of that at 1 T), and becomes less temperature dependent at low temperatures (not shown). Such field induced FL behavior is also observed in $YbRh_2Si_2$ [2, 4, 10]. From these experimental facts, we conclude that the significant field effects

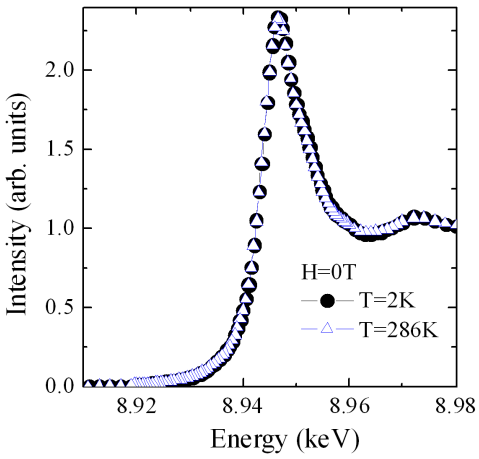

Fig. 1. L_3-edge absorption spectra of $YbCo_2Zn_{20}$ powder sample taken at 2 and 286 K in zero field.

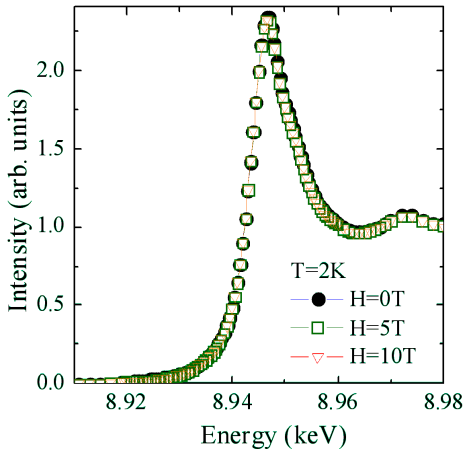

Fig. 2. Field dependence of the L_3-edge absorption spectrum of $YbCo_2Zn_{20}$ measured at fields of 0, 5, and 10 T and at the constant temperature of 2 K.

in $YbCo_2Zn_{20}$ near the magnetic–nonmagnetic transition result from the interactions not between the field and the valence of Yb ions, but between the field and magnetic moments of the $4f$ electrons. The enhanced FL behavior with increasing field is probably attributed to large suppression of spin fluctuations under the field.

4. Conclusions

In summary, we have performed the XAS measurements on the HF compound $YbCo_2Zn_{20}$ up to 10 T in order to investigate the possibility that the application of magnetic field gives rise to changes in the Yb valence in the vicinity of the magnetic–nonmagnetic transition. Although the application of field enhances the FL behavior in $4f$ electron systems, we obtained no evidence for changes in the Yb valence within experimental error. Our results indicate that field induced modifications in

magnetic properties are mainly attributed to large suppression of spin fluctuations.

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