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X-ray absorption spectroscopy and novel electronic properties in heavy fermion compounds YbT$_2$Zn$_{20}$ (T: Rh and Ir)

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Abstract. YbT$_2$Zn$_{20}$ (T: Rh and Ir), which crystallizes in the cubic CeCr$_2$Al$_{20}$-type structure, is a member of the well-known heavy fermion compounds, indicating a huge electronic specific heat coefficient $\gamma \simeq 500$ mJ/(K^2·mol). We have measured temperature and magnetic field dependences of Yb valence in YbT$_2$Zn$_{20}$(T: Rh and Ir) at ambient pressure by the L$_{\rm III}$ edge x-ray absorption spectroscopy in order to investigate the valence state of Yb 4f electrons in these compounds. It is revealed that the Yb valence in both compounds significantly decreases with temperature below about 100 K and increases with increasing magnetic field at low temperatures in contrast to the case of YbCo$_2$Zn$_{20}$.

1. Introduction

The quantum critical phenomena have attracted a great interest in f-electron systems of rare-earth and actinide compounds. The electronic properties of f-electron systems are well illustrated by the Doniach phase diagram, where the ground state is realized as a result of the competition between the Ruderman - Kittel - Kasuya - Yosida (RKKY) interaction and the Kondo effect. The tuning parameter of these effects are magnetic field, pressure, chemical substitution and so forth. For example, when pressure P is applied to an antiferromagnetic Ce-compound, the Néel temperature $T_{\rm N}$ decreases with increasing pressure and becomes zero, where the magnetic fluctuation becomes dominant, at a critical pressure $P_{\rm c}$: $T_{\rm N} \to 0$ K for $P \to P_{\rm c}$. Meanwhile, as the mechanism of the occurrence of the quantum critical phenomena, not only a magnetic fluctuation but also a critical valence fluctuation is discussed theoretically [1, 2]. Moreover, several Ce- and Yb-compounds are suggested as candidates for the critical valence fluctuation mediated quantum critical materials. For example, in the cases of CeCu$_2$Si$_2$ [3] and

CeCu$_2$Ge$_2$ [4], superconducting transition temperature T_{sc} is largely enhanced at the valence crossover pressure P_v compared to the critical pressure P_c of the antiferromagnetic quantum critical point (QCP).

YbT$_2$Zn$_{20}$ (T: Co, Rh, and Ir), which crystallizes in the CeCr$_2$Al$_{20}$-type structure (F$d\bar{3}m$), are heavy fermion compounds with an electronic specific heat coefficient $\gamma = 500{\sim}8000$ mJ/(K^2·mol) [5]. Due to a large Yb-Yb inter-atomic distance about 6 Å, the magnetic exchange interaction between Yb atoms is weak, preventing magnetic ordering in YbT$_2$Zn$_{20}$ compounds. Recently, we have revealed that the electronic state of YbIr$_2$Zn$_{20}$ [6] and YbRh$_2$Zn$_{20}$ [7] approaches to the quantum critical point at critical pressure $P_c \simeq 5$ GPa and they indicate one of the heaviest electronic states with the electronic specific heat coefficient γ, exceeding $\gamma = 10$ J/(K^2·mol) at P_c. It is also found that the residual resistivity values are drastically enhanced at P_c in both cases. It is suggested that the huge enhancement of the residual resistivity can be attributed to the critical valence fluctuation. In order to elucidate the valence state of Yb in these systems at ambient pressure, we have measured temperature and magnetic field dependences of Yb valence by means of the x-ray absorption spectroscopy (XAS).
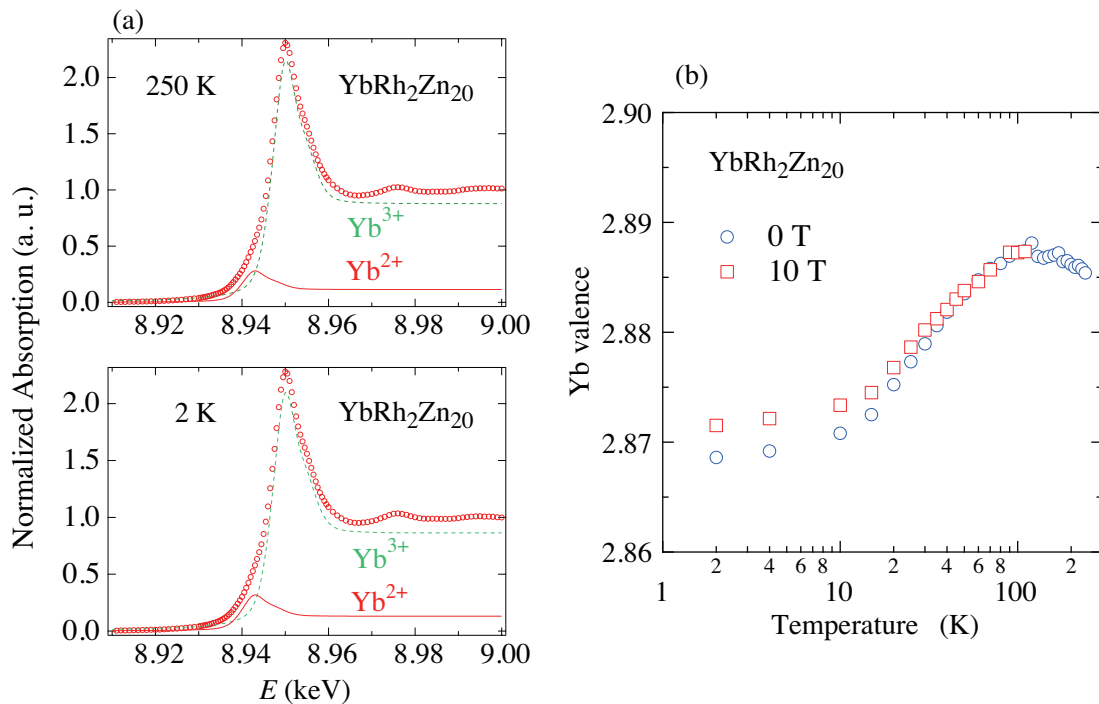
Figure 1. (a) X-ray absorption spectra of YbRh$_2$Zn$_{20}$ obtained at 250 and 2 K. Solid and dotted lines are fitted results for the absorption contribution from Yb^{2+} and Yb^{3+}, respectively. (b) Temperature dependence of the Yb valence at 0 and 10 T in YbRh$_2$Zn$_{20}$.

2. Experimental Procedure

Single crystals of YbT$_2$Zn$_{20}$ (T = Rh and Ir), grown by the Zn-self flux method [7,8], were crashed and powdered gently, and were made into pellets by mixing boron nitride (BN) powder uniformly. The sample was placed inside the ^{4}He cryostat with a 10 T superconducting magnet at BL39XU at SPring-8 [9]. Sample temperature was measured by a standard Cernox thermometer. Yb L_{III} edge XAS was measured by means of a direct transmission method. Scan in the X-Y plane was done to measure the same spot on the sample before each absorption measurements

in order to reduce experimental error caused by inhomogeneity of the pellet. In order to get homogenous temperature for the pellets, we waited enough time for temperature stabilization before starting each experiment.

3. Experimental results and discussion

Figure 1(a) shows the normalized x-ray absorption intensities at the L_{III} edge of Yb as a function of photon energy at 2 and 250 K in YbRh$_2$Zn$_{20}$ in zero magnetic field after subtracting linear back ground by Victoreen function [10]. The main peak at 8.95 keV indicates absorption by a trivalent Yb (Yb^{3+}) and a shoulder like trail around 8.94 keV represents absorption by a divalent Yb (Yb^{2+}). Fitted results using double Lorentzian/Gaussian and arctangent functions are also plotted with solid and dotted lines for the contribution of Yb^{2+} and Yb^{3+}, respectively. Detailed process for analyzing the XAS intensities are described elsewhere [11]. It is clear that the peak height of the main peak from Yb^{3+} decreases and, on the contrary, the one from Yb^{2+} increases with decreasing temperature.

Temperature dependence of the estimated Yb valence of YbRh$_2$Zn$_{20}$ in zero magnetic field is plotted in Fig. 1(b) together with the data obtained under the magnetic field of 10 T, which is described later. With decreasing temperature from room temperature down to 100 K, Yb valence is almost constant with the value of 2.887. Note that the absolute value of valence estimated by XAS is in the error of about 10% of obtained value and, on the other hand, the error for the relative change of the valence is in the range from 1-2% [11, 12]. Yb valence starts to decrease significantly below 100 K and reaches about 2.870 around 20 K. Thus, the substantial valence change $\Delta_{valence}$ is $\Delta_{valence} \simeq 0.017$ in YbRh$_2$Zn$_{20}$. The valence change becomes less prominent below 20 K, which can correspond to the Kondo temperature of YbRh$_2$Zn$_{20}$ $T_K = 16$ K obtained by Torikachvili et al. [5]. The similar behavior was also observed in heavy fermion Yb compounds YbRh$_2$Si$_2$ and β-YbAlB$_4$, where the change of the Yb valence becomes almost constant below $T_K = 20$ K for YbRh$_2$Si$_2$ [11] and $T_K = 200$ K for β-YbAlB$_4$ [13].

Figure 2(a) shows the normalized x-ray absorption intensities at the L_{III} edge of Yb as a function of photon energy at different temperatures in YbIr$_2$Zn$_{20}$ in zero magnetic field. Here, the gradual development of Yb^{2+} component and suppression of Yb^{3+} component with decreasing temperature were also observed. The oscillation like behavior in the high energy region is originated from the extended x-ray absorption fine structure (EXAFS) signal, where we neglect as a background for the fitting. Solid and dotted lines are for the contribution from Yb^{2+} and Yb^{3+}, respectively. Normalized absorption of XAS signal obtained at 2 K under the magnetic field of 0 and 10 T was shown in Fig. 2(b). It is clear that the Yb^{3+} component significantly increases under magnetic field. It should be noted that YbIr$_2$Zn$_{20}$ shows the typical heavy fermion metamagnetism around 10 T below the temperature which the magnetic susceptibility shows a broad maximum $T_{\chi max} = 7.4$ K [8]. Estimated Yb valence as a function of temperature under 0 and 10 T are plotted in Fig. 2(c). The valence is almost constant within an experimental accuracy above 100 K and decreases drastically below 90 K. We recall that the Yb valence becomes almost unchanged below about 20 K, which is close to $T_K = 21$ K of YbIr$_2$Zn$_{20}$.

Here, we would like to remind that the XAS experiment on the isostructural compound YbCo$_2$Zn$_{20}$ was performed and no significant change of Yb valence has been observed with respect to temperature as well as magnetic field [14]. This can be connected with the lowest Kondo temperature, $T_K \simeq 1.5$ K in YbCo$_2$Zn$_{20}$ among the YbT$_2$Zn$_{20}$ system. In fact, the electronic state of YbCo$_2$Zn$_{20}$ is much closer to the quantum critical point compared to YbRh$_2$Zn$_{20}$ and YbIr$_2$Zn$_{20}$, implying that the valence fluctuation in YbCo$_2$Zn$_{20}$ occurs just at the lowest temperatures below about 1 K as is suggested by the temperature dependence of ν_Q in the ^{59}Co nuclear quadrupole resonance experiment [14]. From these results, it is suggested that T_K is a good measure for the $4f$ valence state of Yb compounds. To clarify the relation between valence state and T_K, further experiments such as XAS under pressure are indispensable.
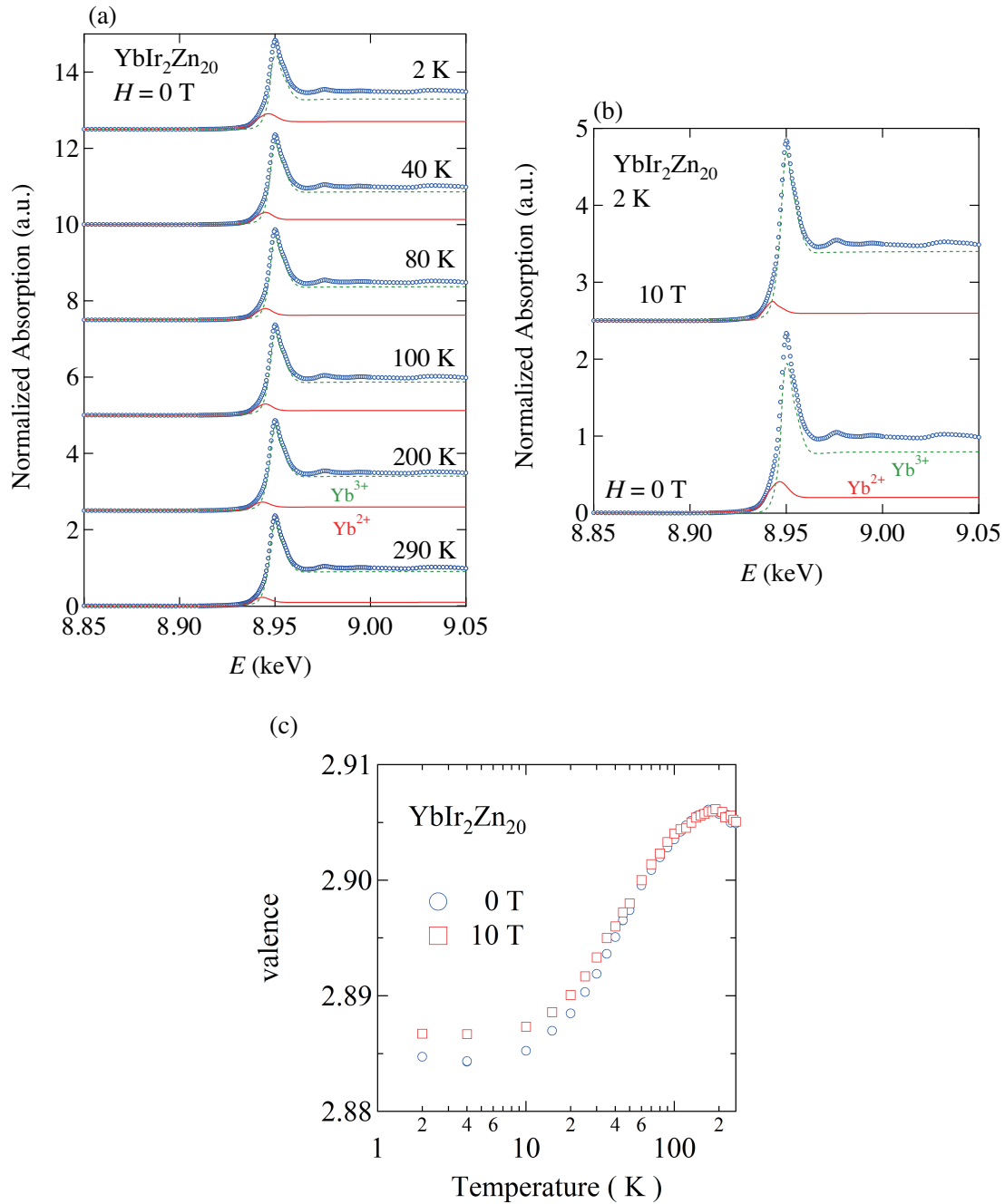
Figure 2. X-ray absorption spectra of $YbIr_2Zn_{20}$ (a) at several temperatures without magnetic field and (b) at 2 K in 0 and 10 T. Solid and dotted lines are fitted results for the absorption contribution from Yb^{2+} and Yb^{3+}, respectively. (c) Temperature dependence of the Yb valence at 0 and 10 T in $YbIr_2Zn_{20}$.

Relative valence change in YbT_2Zn_{20} was also estimated on the basis of the scaling of the products of the magnetic susceptibility and temperature χT, and precise thermal expansion results [15]. It should be noted that the substantial valence change $\Delta_{valence}$ between 0 and 100 K in YbT_2Zn_{20} (T: Rh and Ir) estimated from the χT scaling is $\Delta_{valence} \simeq 0.015$, which is in

good agreement with our direct measurements $\Delta_{\mathrm{valence}} \simeq 0.02$ obtained from the present x-ray absorption experiment.

Significant difference of the Yb valence between 0 and 10 T appears below about 80 K which is well above $T_{\chi\mathrm{max}}$ in both cases. It is not clear whether the valence change with respect to the magnetic field is directly connected with the heavy fermion metamagnetism or not. In order to clarify the relation between the heavy fermion metamagnetism and the critical valence fluctuation, further precise measurements of magnetic field and pressure dependence of the Yb valence in these systems are in progress.

4. Summary
We have carried out the Yb L_{III} edge x-ray absorption spectroscopy at low temperatures down to 2 K and under the magnetic fields of up to 10 T in $YbRh_2Zn_{20}$ and $YbIr_2Zn_{20}$. We have revealed significant changes of Yb valence with respect to temperature as well as the magnetic field in these compounds in contrast to those in $YbCo_2Zn_{20}$. The drastic changes of Yb valence with respect to the temperature and magnetic field can be connected with the strong correlation of electronic states in these systems. It is also found that the Yb valence becomes almost temperature independent below T_{K}.

Acknowledgement

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