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Abstract. We studied the electronic structure of $YbCdCu_4$, which is known as a heavy fermion compound, by means of photoemission spectroscopy. An energy shift and an intensity enhancement of the Yb^{2+} $4f_{7/2}$ peak are clearly observed with decreasing temperature. The characteristic energy scale of the Kondo temperature is estimated as ∼ 26.5 meV, which is in agreement with the Kondo temperature obtained by the magnetic susceptibility measurements. It suggests that the observed $4f_{7/2}$ state is associated with the heavy fermion behavior of $YbCdCu_4$.

INTRODUCTION

In rare-earth compounds, the hybridization effect between conduction and localized-4f electrons (c-f hybridization) leads to the formation of heavy electrons, and it is characterized by a renormalized band structure near the Fermi level (E_F) [1]. According to the single-impurity Anderson model, a sharp resonance peak in the electronic excitation spectrum appears at the energy scales of the Kondo temperature T_K, which is a measure of the hybridization strength [2]. The intensity of the Kondo resonance exhibits a strong temperature dependence that reflects magnitudes of the c-f hybridization [2, 3]. Hence, the temperature dependence of the Kondo resonance peak near E_F will provide important insights for understanding the heavy fermion state.

$YbCdCu_4$ with C15b-type structure is known as a heavy fermion compound with the electronic specific heat coefficient $\gamma \sim 175$ mJ/K^{2}mol [4, 5, 6]. The Kondo temperature is estimated to be $T_K \sim 220$ K from the magnetic susceptibility measurements [5, 7]. The research on $YbCdCu_4$ is expected to provide an informative insight into the electronic states of $YbInCu_4$ with the same structure. By replacing Cd with In atom in Yb$X$$Cu_4$, the valence transition appears at $T_V = 42$ K, and concomitantly the hybridization strength ($T_K \sim 400$ K) increases in the low-temperature phase [8, 9, 10]. The nominal valence configurations of the Cd and In atoms are $5s^2 5p^0$ and $5s^2 5p^1$, respectively. It is considered that the number of $5p$ electrons is critical in promoting the valence transition phenomenon. Therefore, the experimental findings of the electronic structure in $YbCdCu_4$ lead to a better understanding of not only the heavy fermion state but the valence transition as well.

In this study, we have investigated the electronic structure of $YbCdCu_4$ obtained by photoemission spectroscopy with synchrotron radiation. An obvious temperature dependence of the Yb^{2+} $4f_{7/2}$ peak is observed, thus allowing us to extract the characteristic energy scale of the Kondo temperature at the zero temperature limit. We discuss possible scenarios for the heavy fermion state and the valence transition phenomenon.

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EXPERIMENTAL DETAILS

High-quality single crystals of $YbCdCu_4$ were grown by the flux method with CdCu flux [4, 6]. The photoemission spectra shown in Figs. 1 and 2(a) were obtained with $h\nu = 50$ eV photons at BL-1 of the Hiroshima Synchrotron Radiation Center (HSRC) [11]. The total energy resolution was 37 meV. Figure 2(b) includes the data obtained using $h\nu = 24$ eV photons at BL-7U of the UVSOR facility [12]. The energy resolution was 16 meV. The samples were cleaved *in situ* along the (111) plane and maintained under an ultrahigh vacuum (8×10^{-9} Pa) during the measurements.

RESULTS AND DISCUSSION

Figure 1(a) shows the valence-band photoemission spectra of $YbCdCu_4$ at 15 K. The broad peak at $|\omega| \sim 3$ eV is derived from the Cu $3d$ state. Two peaks at $\sim$ 10.3 and 11.0 eV originate from the spin-orbit split Cd $4d$ states. The Yb^{3+} $4f$ multiplet structures are observed at 5 – 12 eV. These assignments are consistent with those of previous studies [13, 14, 15].

In Fig. 1(b), we show an enlarged view of the valence-band spectra near E_F. Two peaks at $|\omega| \sim 0.02$ and 1.3 eV are the bulk-derived Yb^{2+} $4f_{7/2}$ and Yb^{2+} $4f_{5/2}$ states, respectively [14, 15, 16]. The observability of the $4f_{7/2}$ peak is improved by slightly increasing the photon energy from those used in previous studies, $h\nu < 22$ eV [13, 16]. A small and broad peak is observed at $\sim$ 0.9 eV, which can be assigned to the surface contribution of the topmost divalent Yb ions [17].

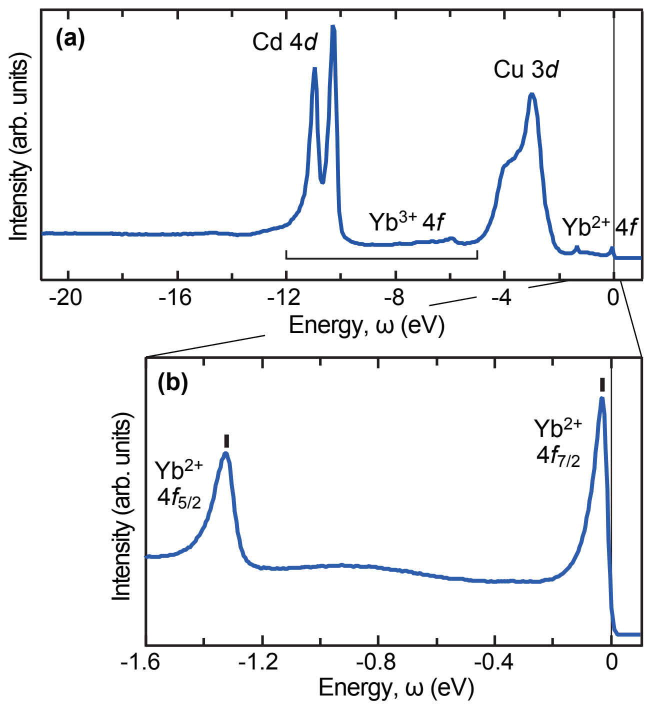

FIGURE 1. Photoemission spectra of $YbCdCu_4$ obtained with $h\nu = 50$ eV photons at 15 K. (a) Valence-band spectra over a wide energy region. (b) Enlarged view of the valence-band spectra near E_F. Vertical bars represent the peak position for the $4f_{7/2}$ and $4f_{5/2}$ states.

Figure 2(a) shows the temperature dependence of the Yb^{2+} $4f_{7/2}$ peak. We found an energy shift and an intensity enhancement of the $4f_{7/2}$ peak with decreasing temperature. It is noteworthy that integrated spectral intensity of the peak is not conserved, and this behavior is incompatible with the simple thermal broadening of the metallic density of states. In contrast, the temperature evolution of the $4f_{7/2}$ peak is consistent with the prediction of the single-impurity Anderson model [2]. The observed $4f_{7/2}$ peak near E_F can therefore be assigned to the Kondo resonance peak in $YbCdCu_4$.

The sharpness of the $4f_{7/2}$ peak is sufficient even at 180 K and thus allows for precise determination of the peak energy. Figure 2(b) shows the energy of the Kondo resonance peak as a function of temperature, together with the data obtained in the UVSOR facility. The results coincide with each other within the error bars, indicating the consistency of our data. The peak energies decrease linearly with decreasing temperature. The appearance of the Kondo resonance at $k_B T_K$ in the limit of zero temperature represents the formation of the singlet ground state [2]. Thus, we have estimated the energy scale of the Kondo temperature at $T = 0$ K. As shown by the dashed line in Fig. 2(b), the energy extrapolated to $T = 0$ K is estimated as $\sim$ 26.5 meV, which corresponds to $T_K \sim 308$ K. Its value is approximately consistent with T_K obtained by the magnetic susceptibility measurements [5, 7]. This consistency demonstrates that the observed $4f_{7/2}$ state in this study is indeed responsible for the heavy fermion behavior of $YbCdCu_4$.

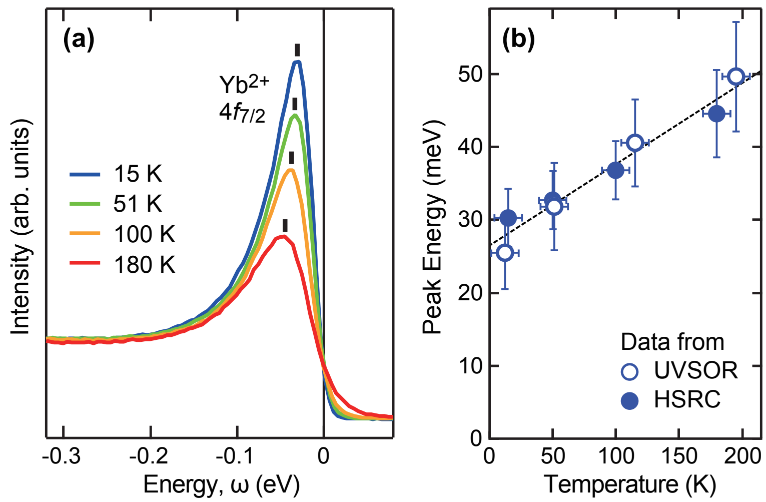

FIGURE 2. (a) Temperature dependence of photoemission spectra near E_F. Vertical bars indicate the peak positions of Yb^{2+} $4f_{7/2}$ state. (b) The peak energies as a function of temperature, together with the data obtained in the UVSOR facility. Dashed line represents the result of the linear fits.

It has been reported that replacing Cd with In in YbXCu$_4$ shifts the Fermi edge to the minimum in the density of states and consequently depresses the electrical conductivity [18]. This reduction in the conductivity changes the long-range interactions in $YbCdCu_4$ to the short-range interactions in $YbInCu_4$ [19]. Therefore, a change in the hybridization strength is relevant to the emergence of the valence transition. The band dispersions of $YbCdCu_4$ have not been reported hitherto, whereas the hybridization strength between the conduction and $4f$-derived bands in $YbInCu_4$ has been quantified from angle-resolved photoemission spectroscopy [20]. Further investigations on $YbCdCu_4$ for the momentum, temperature, and band dependences of electronic structures are required to understand the mechanism of the valence transition phenomenon.

CONCLUSION

We investigated the temperature dependence of the $4f_{7/2}$ state of $YbCdCu_4$ by photoemission spectroscopy. With decreasing temperature, the energy shift and the intensity enhancement in the $4f_{7/2}$ peak are observed. The peak energy at $T = 0$ K is estimated to be $\sim$ 26.5 meV, which corresponds approximately to the scale of T_K. Our results therefore demonstrate that the observed $4f_{7/2}$ state is responsible for the heavy fermion behavior of $YbCdCu_4$.

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