BULK 4*f* STATES OF CORRELATED Ce SYSTEMS REVEALED FOR THE FIRST TIME BY UNPRECEDENTED HIGH-RESOLUTION RESONANCE PHOTOEMISSION SPECTROSCOPY

Photoemission spectroscopy (PES) is a direct means to probe the electronic structures of new functional materials which exhibit high-transition-temperature superconductivity, heavy fermion behavior, metal-to-insulator transitions and other unique properties. High-resolution PES is required to investigate their detailed electronic states near the Fermi level (E_F). However, conventional high-resolution PES using low-energy excitation is limited by its surface-sensitivity due to a short photoelectron mean free path. For example, previously reported 4*f* spectra (hν≤120 eV, with surface contribution of ~55%) for correlated Ce compounds have similar shapes, when they should be quite different due to effects of hybridization with other valence bands. This problem is solved by the bulk-sensitive high-resolution 3*d*-4*f* resonance photoemission (surface contribution of ~15 %), which has became available recently at beamline **BL25SU** [1]. Here we present unprecedented high-resolution Ce 3*d*-4*f* resonance photoemission spectra of moderately and strongly hybridized Ce compounds ($CeRu_2Si_2$ and $CeRu_2$), in which the genuine bulk Ce 4*f* states are revealed. $CeRu_2Si_2$ is a typical heavy fermion system with a Kondo temperature (T_K) of ~20 K. This material is thought to reside at a boundary between the localized and itinerant Ce 4*f* states. $CeRu_2$ is a typical material of strongly valence-fluctuating 4*f* systems, for which T_K is of the order of 1000 K.

High-resolution Ce 3*d*-4*f* resonance PES was performed on single crystal samples with an overall energy resolution of about 100 meV at hν~880 eV. The samples were cooled down to 20 K and cleaned *in situ* by scraping or cleavage. Surface cleanliness was confirmed before and after the measurements.

The Ce 4*f* spectra of $CeRu_2Si_2$ and $CeRu_2$ in a wide energy region for the Ce 3*d*-4*f* resonances are shown in Fig. 1. In the $CeRu_2Si_2$ spectrum, there is a sharp peak near E_F and a broad tail ranging from −1 to −5 eV. According to the single impurity Anderson model (SIAM), the former corresponds to the contribution of both the tail of the Kondo peak ($f_{5/2}^1$) and its spin-orbit partner ($f_{7/2}^1$). The latter is ascribed to the f^0 final state. Such a f^0 broad tail is greatly suppressed in the spectrum of $CeRu_2$ compared with that of $CeRu_2Si_2$, indicating that the bulk Ce 4*f* states are considerably hybridized in $CeRu_2$.

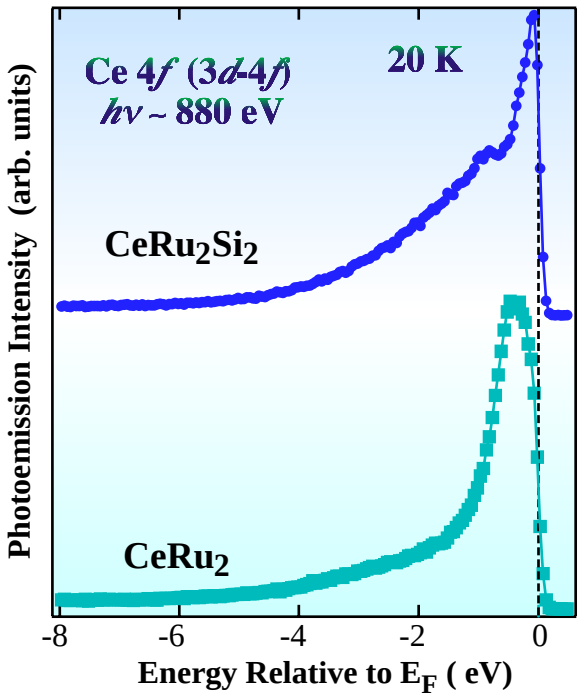

Fig. 1. Ce 4f spectra of $CeRu_2Si_2$ and $CeRu_2$ obtained from Ce 3d-4f resonance photoemission with an energy resolution of 200 meV. The Ce 4f contributions are estimated by subtracting the off-resonance spectra taken at hν = 875 eV from the on-resonance spectra at hν = 882.6 eV. The baseline photoemission level is shifted for clarity.

The detailed high-resolution spectra near E_F are shown in Fig. 2. The spectra of the two compounds are drastically different from one another. For $CeRu_2Si_2$, there are a prominent peak in the vicinity of E_F and a weak shoulder near –0.3 eV. These prominent and weak structures originate from the bulk $f_{5/2}^1$ and $f_{7/2}^1$ final states; that is, from the tail of the Kondo peak and its spin-orbit partner as predicted from SIAM. Such a strong tail of the bulk Kondo peak has clearly been observed for the first time owing to the bulk-sensitive 3*d*-4*f* resonance PES with an unprecedented high-resolution of 100 meV. The spectral shape near E_F of $CeRu_2$ is, however, rather surprising since no structure is seen except for a broad feature centered at –0.5 eV. One would have expected that the tail of the Kondo peak should be much stronger in the bulk-sensitive 3*d*-4*f* resonance PES spectrum for such a strongly hybridized system like $CeRu_2$, as predicted from SIAM. The fact that the spectrum demonstrates a rather conventional Fermi cut-off together with the broad peak at –0.5 eV cannot be explained by SIAM. Thus, the bulk 4*f* spectrum of $CeRu_2$ is interpreted to represent the itinerant 4*f*-band due to the very strong hybridization effect, which is beyond the scope of SIAM. This behavior, which has not been revealed by any previous 4*d*-4*f* resonance photoemission studies, reflects the real T_K and bulk properties of these materials. Further detailed studies, including a comparison between these 3*d*-4*f* spectra and surface-sensitive 4*d*-4*f* resonance PES spectra of the same compounds are described elsewhere [2].

In summary, a new photoemission technique using high-resolution, high-hν excitation has revealed the bulk electronic states of high- and low-T_K Ce compounds, which were inaccessible by conventional high-resolution PES. SIAM is applicable to the low-T_K system, but not to high-T_K system, which demonstrates significant itinerant 4*f* character. This technique can be applied as well to systems such as strongly correlated transition metal compounds.

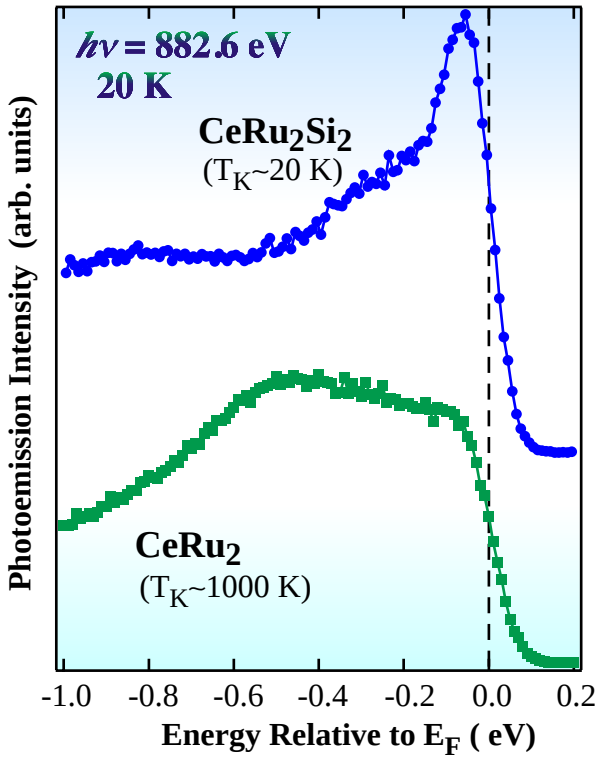

Fig. 2. High-resolution Ce 3d-4f on resonance spectra with an resolution of 100 meV of $CeRu_2Si_2$ and $CeRu_2$.

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References

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