High-resolution, low-temperature photoemission spectroscopy of Kondo semiconductor CeRhAs and related compounds

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Abstract

Ce $4f$ states of the Kondo semiconductor CeRhAs, semimetal CeRhSb, and metal CePtSn single crystals were observed directly by high-resolution resonant photoemission spectroscopy. A large gap and a pseudogap were found in CeRhAs and CeRhSb, respectively.

Key words: Kondo semiconductor;photoemission spectroscopy;CeRhAs;CeRhSb;

CeRhAs and CeRhSb, with the orthorhombic ϵ-TiNiSi-type structure, are Kondo semiconductor and Kondo semimetal, respectively.[1–3] They have attracted much interest for a small energy gap or pseudogap formation in the ground state without magnetic ordering.[1] In this paper, we report Ce $4f$ states in CeRhAs and CeRhSb single crystals by means of high-resolution, low-temperature resonant photoemission spectroscopy. We discuss unusual Ce $4f$ electronic states in these compounds comparing them with that of the isostructural Kondo metal CePtSn single crystal.[4]

CeRhAs and CeRhSb single crystals were grown by the Bridgman method,[2,3] and CePtSn single crystals were grown by the Czochralski method.[4] The Kondo temperatures (T_K's) for CeRhAs, CeRhSb, and CePtSn were estimated to be T_K ∼1500 K (∼130 meV), ∼360 K (∼30 meV), and ∼10 K (< 1 meV),[4] respectively. The former two temperatures were inferred by assuming the relation $T_K \sim 3\ T_m$.[5] The present measurements were performed on a high-resolution linear undulator beamline (BL-1) of a compact electron storage ring (HiSOR) located at Hiroshima Synchrotron Radiation Center (HSRC), Hiroshima University.[6] Photoemission experiments were done using SCIENTA SES200. The total instrumental energy resolution was set at 18-20 meV at $h\nu$ = 126 eV. The samples were cooled down to 10-12 K. To obtain clean surfaces, we fractured the single crystalline samples *in situ* in ultrahigh vacuum (3×10^{-10} Torr). The binding energy was calibrated using the Fermi edge of Au with the accuracy of ±2 meV. At photon energy of $h\nu$ = 126 eV, which is close to Ce $4d$-$4f$ on-resonance ($h\nu$ = 126 eV), the Ce $4f$ contribution dominates the spectra.

Figure1 shows the Ce $4f^1$ derived spectra near the Fermi level (E_F). The intensities are normalized to the spectral intensity at ∼300 meV. The Ce $4f^1$ spectra of CeRhSb and CePtSn are split by the spin-orbit interaction into two peaks at ∼300 meV and ∼E_F. On the other hand, there is no peak structure near E_F in the spectrum of CeRhAs.

To estimate the spectral density-of-states (SDOS), we divided the photoemission spectra (normalized at

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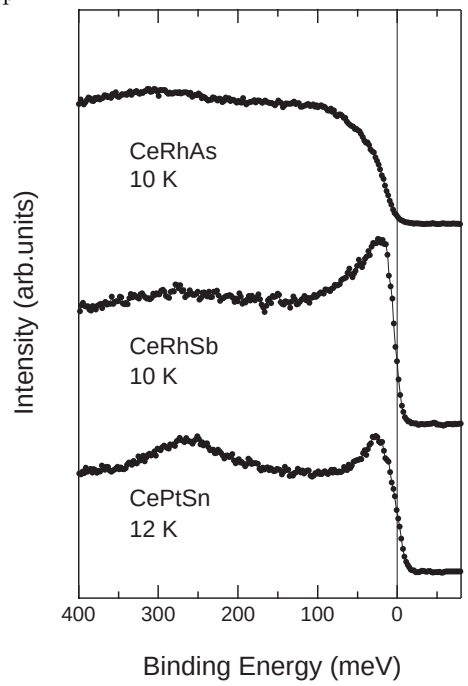

Fig. 1. High-resolution resonant photoemission spectra of CeRhAs, CeRhSb, and CePtSn taken at $h\nu = 126$ eV. These spectra reflect the Ce $4f$ electronic states near E_F.

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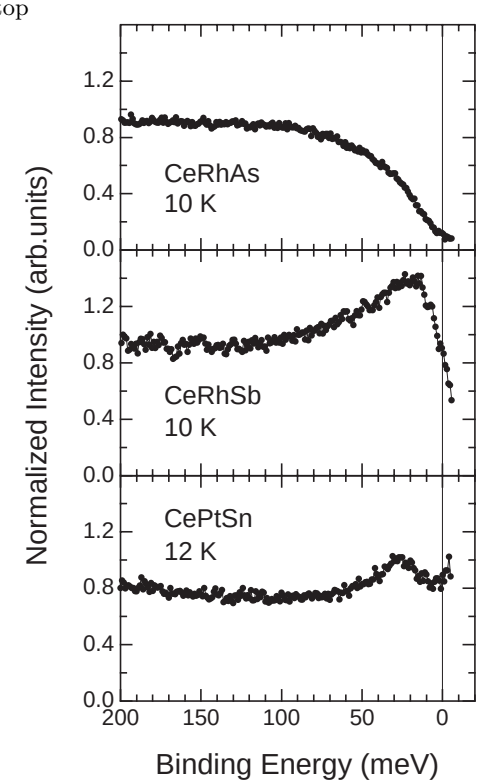

Fig. 2. Photoemission spectra of CeRhAs, CeRhSb, and CePtSn divided by broadened FDD. These spectra are assumed to reflect the SDOS broadened with the instrumental resolution.

$\sim$300 meV) by a Fermi-Dirac distribution function (FDD), convoluted with a Gaussian which represents the instrumental resolution,[7] as shown in Fig.2. The spectral intensity of CeRhAs decreases monotonically above $\sim$90 meV, forming a large gap structure. The energy gap of CeRhAs is very close to a fullgap rather than a pseudogap.

As shown in Fig.2, the spectral intensity of CeRhSb shows enhancement above $\sim$120 meV, which is similar to that of Kondo metals with high T_K.[8] However, above $\sim$13 meV the spectral intensity decreases steeply, which is an important feature different from that of CePtSn and other Kondo metals.[8,7] The rapid decrease in the spectral intensity strongly supports the existence of a narrow pseudogap.

The spectral intensity of CePtSn exhibits no remarkable enhancement near E_F, except for a peak structure at $\sim$27 meV. The peak is derived from crystal field splitting. Weak Kondo resonance is consistent with the low $k_B T_K < 1$meV.[8]

Based on the periodic Anderson model (PAM), Ikeda and Miyake,[9] and Moreno and Coleman[10] showed the semimetallic spectral density for CeNiSn or CeRhSb. The observed spectrum for CeRhSb can be qualitatively understood in terms of these models. However, the observed spectral feature of CeRhAs is significantly different from the spectral density given by the PAM. The absence of a peak structure near E_F is highly suggestive of much stronger c-f hybridization. More realistic energy band dispersions should be taken into account.

In summary, the Ce $4f$ derived electronic states of the isostructural Kondo semiconductor CeRhAs, semimetal CeRhSb and metal CePtSn single crystals have been investigated, by high-resolution, low-temperature resonant photoemission spectroscopy. A large gap and a pseudogap were found in CeRhAs and CeRhSb, respectively.

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