



Resonant inverse photoemission study on CeRh₃ at Ce 4d absorption edge

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Abstract

The resonant inverse photoemission study (RIPES) of CeRh₃ has been carried out at Ce 4d → 4f absorption edge. The 4f cross section increases drastically when the excitation energy is tuned to absorption edge. The line shape of 4f² state changes by the resonance effect because of its multiplet structure. The RIPES of CeRh₃ which is in the valence fluctuation (VF) region shows the strong f¹ intensity. The energy position of f¹ peak of CeRh₃ is about 1 eV, which reflects the extremely high Kondo temperature of CeRh₃. © 1999 Elsevier Science B.V. All rights reserved.

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Resonant inverse photoemission spectroscopy (RIPES) is a new technique to investigate the unoccupied state of solid. Recently, the RIPES study has been reported on several Ce compounds near the Ce M₅ absorption [1,5,6] and Ce N_{4,5} absorption [2] edge. The great ability to study the 4f electronic structure by resonant enhancement of the 4f signal was testified. The 4f cross section increases when the excitation energy tuned to Ce 3d or Ce 4d absorption edge and the Ce -4f contribution can be extracted. It is found that the normal fluorescence is not so serious and the resonance effect is stronger near the Ce 4d → 4f threshold as compared with the 3d → 4f threshold.

Measurements were performed in an ultra high vacuum chamber where the base pressure is under 1 × 10⁻¹⁰ Torr. Clean surface was obtained by scraping the sample with a diamond file in vacuum every 30 min. The temperature at the sample is about 30 K. A BaO-cathode-type electron gun was used for the excitation source.

The kinetic energy, E_k , of the electron was calibrated by the electron energy analyzer. The IPES was measured by the soft X-ray emission spectrometer of the Institute for Solid State Physics [3], which is Rowland mount type. The Fermi level position and a resolution of the system was determined by referring to the Fermi edge in the IPES spectra of Au which was evaporated on the sample holder. Resolution of this system is about 0.4 eV at $E_k = 80$ eV.

Fig. 1 shows the RIPES spectra of CeRh₃ as the excitation energy is tuned to the Ce 4d → 4d absorption edge, where the abscissa is the emission energy. The fluorescence spectra of 4d and 5p states are found with the same emission energies. The RIPES spectra shift in accord with the excitation energy. Fig. 2 shows the RIPES spectra of CeRh₃ whose abscissa is the energy above the Fermi level which was calibrated by the Fermi edge of gold. One can find two clear structures above the Fermi level. Both the structures have remarkable resonant enhancement. The peak at just above Fermi level is known to be the f¹ state which mainly comes from f⁰ initial state. The resonance enhancement is evident at 121 eV. The structure around 5 eV is the f² states which comes from f¹ initial states and the resonance is clear at

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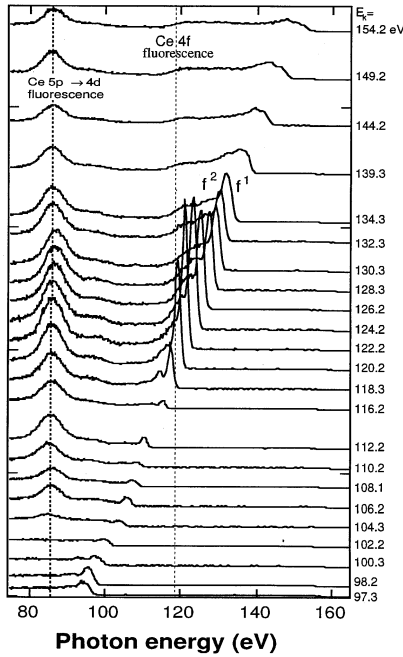


Fig. 1. The RIPES spectra of CeRh₃ near the Ce 4d → 4f edge, where the abscissa is the emission energy. The numbers written on the side of the right axis represent the kinetic energy, E_k of the electron.

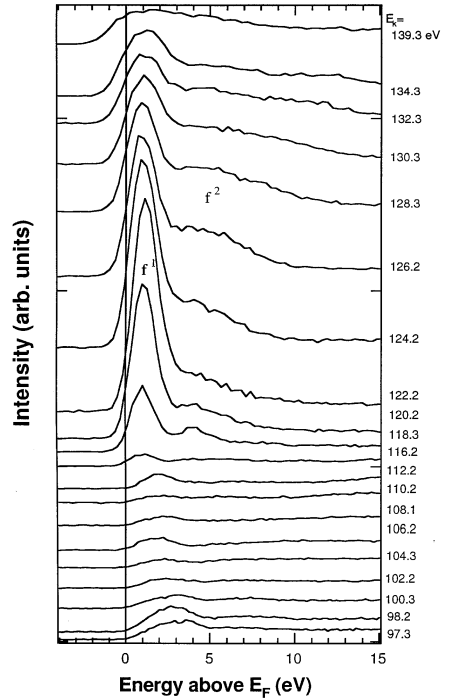


Fig. 2. The RIPES spectra of CeRh₃ near the Ce 4d → 4f edge, where the abscissa is the energy above Fermi level ($E_F = 0$ eV).

about 124 eV. Grioni et al. measured the RIPES spectra of CeRh₃ by Ce 3d resonance which is similar to these spectra.

The energy position of f^1 peak of CeRd₃ is about 1 eV, which reflects the extremely high Kondo temperature of CeRh₃. The extremely high T_K of CeRh₃ is mainly caused by this strong hybridization between the 4f state and the conduction band. The large T_K of several thousands is in agreement with the other experiments.

The normal IPES process is represented for the transition to Ce 4f states as follows:

$$|4d^{10}4f^n\rangle + e^- \rightarrow |4d^{10}4f^{n+1}\rangle + hv. \quad (1)$$

Here, n is the configuration number of the 4f electrons in the ground state. For the incident energy range of the present experiment, the contribution from non-f conduction bands also coexists with the f contribution in the normal IPES. In the RIPES experiment, on the other hand, the resonant process is expressed by the following processes:

$$|4d^{10}4f^n\rangle + e^- \rightarrow |4d^94f^{n+2}\rangle \rightarrow |4d^{10}4f^{n+1}\rangle + hv. \quad (2)$$

Since the initial and final states are the same in these two processes, they have an interference with each other. Therefore, the 4f cross section increases when the excita-

tion energy is tuned to Ce 4d → 4f absorption edge and we can extract the Ce 4f contribution.

Since f^1 and f^2 states have strong resonant enhancements, they are reliably 4f states and cannot be elucidated by the band effect. It is found that f^2 states have broad multiplet structures. Two multiplet structures become clear below the resonance excitation energy. On the other hand, only a sharp structure is enhanced at the resonance. These spectra are consistent with the calculated RIPES spectra that have been carried out by Tanaka et al. for several Ce compounds [4,7]. These facts show that the RIPES spectra have clear selection rule at the resonance. On the other hand, IPES spectra at off resonance have less selection rule, so that they have broad structures.

The resonance enhancement of the f^1 state is drastic around 126 eV, while the enhancement of f^2 state is caused in a wider energy range. This is because of the wide multiplet structures of f^2 state in the intermediate state.

In conclusion, the RIPES spectra of CeRh₃ have been measured. The CeRh₃ is in the valence fluctuation (VF) region, where the 4f electron has an itinerant character because of the strong hybridization with the conduction band. The energy position of f^1 peak of CeRd₃ is about 1 eV, which reflects the extremely high Kondo temperature of CeRh₃. The extremely high T_K of CeRh₃ is

mainly caused by this strong hybridization between the 4f state and the conduction band. This result is in agreement with the other experiments.

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