Resonant Photoemission Spectroscopy of RFe₄P₁₂ (R= La, Ce, Pr)

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Introduction
Ternary intermetallic compounds RFe₄P₁₂ (R= rare earth element) with the filled skutterudite structure exhibit various interesting properties. According to the measurements on a high quality single crystal [1], CeFe₄P₁₂ shows the complex temperature dependence of resistivity unexpected for a simple single-gap semiconductor; PrFe₄P₁₂ exhibits the Kondo-like anomalies in the transport properties. We investigated the electronic states of RFe₄P₁₂ (R= La, Ce, Pr) by the high-resolution resonant photoemission spectroscopy. In this paper, we report the Pr 4f spectra of PrFe₄P₁₂.

Experimental
The photoemission experiments were performed using synchrotron radiation at the beam line BL-11D of the Photon Factory, High Energy Accelerator Research Organization (KEK). The instrumental resolution was about 65 meV. The samples were cooled to 20 K.

Results and Discussion
Figures 1 shows the resonant photoemission spectra of PrFe₄P₁₂ and the Pr 4f spectrum obtained by subtracting the resonance minimum spectrum (hν = 115 eV) from the resonance maximum spectrum (hν = 124 eV). In the 115 eV and 124 eV spectra, the intense band located at the binding energy of ~0.7 eV is mainly due to the Fe 3d band. In the 4f spectrum, the peaks located at ~4.6 eV and ~0.5 eV correspond to the f1 and f2 peaks, respectively. Furthermore, a weak additional structure was observed at 2.6 eV and the f2 peak shows the complicated multiplet structure. To assign these structures, we calculated the multiplet structure of the 4f² configuration using Cowan’s code [2]. The Vertical bars in Fig. 1 indicate the calculated multiplet components.

For Pr compounds, data for the high-resolution photoemission spectrum are sparse. We have measured the 4f spectra of PrSn₃ with a high Kondo temperature (T_K) and a weakly hybridization system PrAl₂ as the reference samples. Figure 2 shows the Pr 4f photoemission spectra in the vicinity of the Fermi energy. As can be seen from Fig. 2, the intensity of the f² peak for PrFe₄P₁₂ is stronger than those for the reference systems. This indicates the strong hybridization between the Pr 4f and the valence band states.

In all the spectra, the structures due to the ³H₄ state were observed at ~0.06 eV. For PrFe₄P₁₂ and PrAl₂, the ³H₄ peaks were observed as a shoulder. On the other hand, the spectrum of PrSn₃ with a high-T_K exhibits the ³H₄ peak as the peak structure. According to the experimental and theoretical results of the Ce 4f spectrum of a Kondo system [3], the spectral shape of the f5/2 peak ascribed to the tail of the Kondo resonance peak varies from a shoulder to a prominent peak with increasing T_K. The shoulder structure for PrFe₄P₁₂ is similar to the feature observed in the very low-T_K Ce system. Thus, we infer that PrFe₄P₁₂ is not a high-T_K system.

References

Figure 1: Resonant photoemission spectra of PrFe₄P₁₂. The solid line and vertical bars indicate the Pr 4f spectrum and the calculated multiplet components, respectively.

Figure 2: Pr 4f photoemission spectra of PrFe₄P₁₂, PrSn₃ and PrAl₂.

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