Bulk Pr 4f electronic state of heavy fermion Pr filled skutterudites

S. Imada,^a A. Yamasaki,^a T. Nanba^b, A. Sekiyama,^a H. Sugawara,^c H. Sato,^c C. Sekine,^d I. Shirotani,^d H. Harima,^e and S. Suga^a

^aGrad. Sch. Eng. Sci., Osaka Univ., ^bGrad. Sch. Sci., Kobe Univ., ^cGrad. Sch. Sci., Tokyo Metropolitan Univ., ^dMuroran Inst. Tech., ^eISIR, Osaka Univ

In order to obtain the bulk electronic states of Pr filled skutterudites, photoemission experiments have been performed by utilizing soft x-rays as incident photons. In particular, information of Pr 4f electronic states was obtained from the spectrum taken by tuning the photon energy to the Pr 3d-4f excitation energy (on-resonance spectrum). On the other hand, the spectrum taken by using the photon energy well below it (off-resonance spectrum), yields the information about the transition metal d electronic states was obtained. The systems which have been studied are $PrFe_4P_{12}$, $PrRu_4P_{12}$, $PrRu_4Sb_{12}$, and $PrOs_4Sb_{12}$.

The off-resonance spectrum depended very much upon the system. The intensity at the Fermi level (E_F) is the strongest for $PrFe_4P_{12}$ and the next was $PrOs_4Sb_{12}$. We should note that these are the systems which shows heavy-fermion like behavior. The system dependence of the off resonance spectrum was found to be qualitatively consistent with the valence band structure obtained by the band structure calculation.

The on-resonance spectrum also depended very much upon the system. The on-resonance spectra were qualitatively different from the Pr 4f density of states obtained by the band structure calculation, which is due to the localized character of the Pr 4f electrons. The most characteristic feature was the very strong intensity near E_F found for $PrFe_4P_{12}$ whereas the other systems showed more or less similar spectra. The strong near- E_F structure found in the spectrum of $PrFe_4P_{12}$ is interpreted as the Kondo resonance. The intensity of the structure has further been found to depend upon temperature, namely it increased as the temperature was lowered. This temperature dependence has been interpreted to suggest that the 4f electronic states that enter the Kondo state are $4f^2$ and $4f^3$ rather than $4f^1$. On the other hand, the weak intensity near E_F for $PrOs_4Sb_{12}$ might appear to be strange. However, this can be interpreted by the quite low Kondo temperature (T_K) of $PrOs_4Sb_{12}$.