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High energy photoemission of Pr-based skutterudites

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Bulk sensitive photoemission by means of high energy photons, which is now available with high resolution, has been utilized for the study of the electronic states of Pr-based skutterudites.

Resonant photoemission at the Pr $3d \rightarrow 4f$ absorption region was performed to extract the Pr $4f$ excitation spectrum. A very strong spectral intensity is observed just below the Fermi level in the Pr $4f$ excitation spectrum of the heavy-fermion system $PrFe_4P_{12}$. The increase of its intensity at lower temperatures is observed. These suggest that there is a Kondo resonance (KR) in the $4f$ photoemission spectrum of $PrFe_4P_12$. The intensity of the KR increases as the temperature is lowered below 200 K, which is qualitatively consistent with the Kondo temperature of about 10 K. Such a tendency has been experimentally observed and also theoretically reproduced for Kondo resonance in the Yb compounds where the dominant $|f^{13}\rangle$ is mixed with $|f^{14}\rangle$ of increased electron number and Kondo resonance is directly observed by photoemission. On the other hand the temperature dependence for Kondo Ce compounds is that it decreases when the temperature is lowered, which is interpreted theoretically with assuming that the dominant $|f^1\rangle$ is mixed with $|f^0\rangle$ of decreased electron number and the tail of KR is observed in photoemission. Therefore the observed temperature dependence suggests that the Kondo state is composed of the dominant $|f^2\rangle$ mixed with $|f^3\rangle$. The origin of the KR is considered to be the Kondo effect caused by the strong hybridization between the Pr $4f$ and P $3p$ states in the vicinity of E_F[1].

On the other hand, the Pr $4f$ excitation spectrum of the heavy-fermion superconductor $PrOs_4Sb_{12}$ does not exhibit strong intensity near E_F but instead its spectral feature near E_F is quite similar to that of $PrRu_4Sb_{12}$. Namely, Kondo resonance was not observed in $PrOs_4Sb_{12}$. This seems to suggest that the Kondo temperature of $PrOs_4Sb_{12}$ is much lower than that of $PrFe_4P_{12}$.

Another means to study Pr $4f$ electronic states is Pr $3d$ core level photoemission (XPS) in which Pr $4f$ state is reflected through intraatomic Pr $3d-4f$ interaction on the spectral structures. Because of its high binding energy of about 900 eV, higher photon enegy is needed for bulk sensitive measurements. Therefore, measurements were performed by means of X-rays of ~ 2.5 and ~ 5.5 keV. The observed Pr $3d$ XPS was quite different for different compounds. The satellite structures were found in the lower binding energy side of the main peak in $PrFe_4P_{12}$ and $PrSn_3$, which reflects the $|3d^9 4f^3\rangle$ final state of photoemission. Cluster model calculation taking into account the atomic multiplet splittings due to the intraatomic electron-electron interaction qualitatively reproduced the observed spectra. The Pr $4f$ electron number estimated through this was 2.07, 2.03, and 2.02 for $PrFe_4P_{12}$, $PrSn_3$, and Pr metal, respectively. This supports the strong hybridization between Pr $4f$ and the conduction band in $PrFe_4P_{12}$.

[1] A. Yamasaki, *et al.* Phys. Rev. B in print.