Crystal-field excitation in $Pr(Ru_{1-x}Rh_x)_4P_{12}$

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Higher-rank multipolar ordering of $4f$ electron state of Pr-filled skutterudite has been a fascinating topic. $PrRu_4P_{12}$ is a typical system, in which the antiferro-type hexadecapolar (rank-4 multipolar) ordering occurs below the metal-nonmetal transition at 63 K. This phase transition is interpreted as a new type of charge density wave formation due to the Fermi surface nesting property strongly coupled with the modulated $4f$ multipolar arrangement. p-f hybridization between $4f$ and conduction electrons plays an essential role in the ordering mechanism. However, some properties have remained unsolved. The low-temperature resistivity does not diverge with decrease of temperature, and the non-metallic state is easily suppressed by magnetic field of 1 T. The weak nonmetallic state is expected to originate from other mechanism in addition to p-f hybridization. The substitution of Rh to Ru gives rise to rapid suppress of metal-nonmetal transition; the electrical resistivity at low temperature in the Rh 10% system becomes the same magnitude at around 60 K. Thus, the doping effect is a key to understand the weak nonmetallic phase of $PrRu_4P_{12}$. Then we have carried out inelastic scattering experiments to measure crystal-field excitation of $Pr(Ru_{1-x}Rh_x)_4P_{12}$.

We performed the experiments using the triple-axis spectrometers HER (C1-1) and TOPAN (6G) for polycrystalline sample of $Pr(Ru_{1-x}Rh_x)_4P_{12}$ ($x = 0, 0.03, 0.05, 0.10$ and 0.15). Figure 1 depicts the low-energy spectra of the samples of x=0 and 0.03 at 7 K. The peak at 3.2 meV of the $x = 0$ sample corresponds to the exitation from the ground state triplet $\Gamma_4^{(2)}$ to the first excited state singlet Γ_1, which exhibit strong temperature dependence below the transition temperature. On the other hand, a strong peak at 2.4 meV in the data of the $x = 0.03$ sample appears, which is not observed in the $x = 0$ sample. The 2.4 meV peak is common for the Rh doped samples, and the excitation energy does not depend on temperature. Moreover, the integrated intensity increases with the increment of the Rh concentration. The Rh doping modifies the Pr $4f$ crystal-field levels not to shift against temperature variation, then the hexadecapolar ordering with the level shifts is suppressed. An additional $4d$ electron of Rh is expected to be doped in the conduction band. The metal-nonmetal transition of $PrRu_4P_{12}$ is sensitive not only to the p state originating from the pnictogen P but also to the $4d$ state by the transition-metal site.

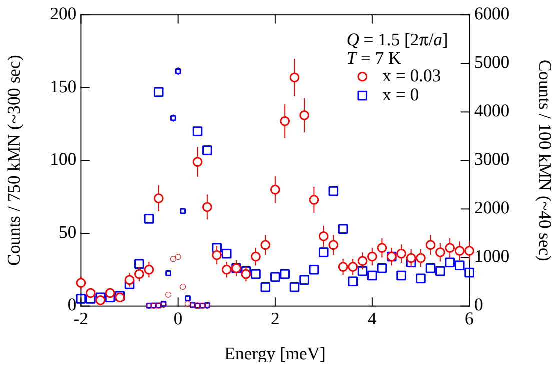

Fig. 1. Crystal-field excitations of $PrRu_4P_{12}$ and $Pr(Ru_{0.97}Rh_{0.03})_4P_{12}$ at 7 K.