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

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Spontaneous ordering of higher-rank multipoles of $4f$ electrons has been detected in various rare-earth based materials. $PrRu_4P_{12}$ is one of typical systems, exhibiting the antiferro-type hexadecapolar (rank-4 multipolar) ordering below the metal-nonmetal transition at 63 K (T. Takimoto: J. Phys. Soc. Jpn. 75 (2006) 034714). The phase transition is a new type of charge density wave formation originating from the coupling of the Fermi surface nesting property to the modulated $4f$ multipolar arrangement. This coupling is due to $p$-$f$ hybridization between $4f$ and conduction electrons. It should be notable that the ordering is characterized by the crystal field excitations exhibiting the strong temperature dependence in accordance with the evolution of the multipolar order parameter (K. Iwasa *et al.*: Phys. Rev. B 72 (2005) 024414). 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 (C. Sekine *et al.*: Physica B 378-380 (2006) 211). Thus, a study of the doping effect leads to understand the ordered 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 have performed experiments using the triple-axis spectrometers HER (C1-1) and TOPAN (6G) for polycrystalline sample of $Pr(Ru_{1-x}Rh_x)_4P_{12}$. These previous studies revealed that the Rh doping systems ($x$ = 0, 0.03, 0.05, 0.10 and 0.15) shows the crystal field excitations at 2.4 and 13 meV, whose peak positions do not show any temperature dependence, in addition to the strongly temperature dependent ones. The most recent measurement for $x$ = 0.01 performed at HER also shows the same excitation spectrum. The peaks appearing in the Rh doped systems indicate appearance of Pr ions without contribution to the ordering. In order to identify the level schemes produced by the Rh doping, we carried out the inelastic measurements under magnetic field for the $x$ = 0.05 system. Figure depicts the magnetic field dependence of the excitation at 2.4 meV measured at 1.6 K. The broadening and asymmetric shape of the peak under finite magnetic field indicate that the peak corresponds to excitation from singlet ground state to triplet state. On the phase transition to the antiferro-type hexadecapolar ordering in the pure system, the crystal field level scheme at one of the Pr ion sites switches from a singlet to a triplet state. Thus, the Rh doping is thought to force the Pr $4f$ electron state to remain at the singlet state, resulting into the suppression of the transition.

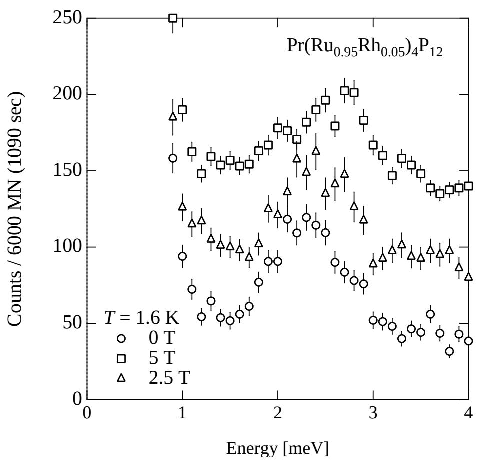


Fig. 1. Magnetic field dependence of crystal-field excitations of $Pr(Ru_{0.95}Rh_{0.05})_4P_{12}$ at 1.6 K.