

 MLF Experimental Report		提出日 Date of Report
課題番号 Project No. 2012A0014	装置責任者 Name of responsible person Shin-ichi Itoh	
実験課題名 Title of experiment Successive Metal–Nonmetal Transitions with Totally–Symmetric Electron Ordering in $(\text{Pr}_{1-x}\text{Ce}_x)\text{Ru}_4\text{P}_{12}$	装置名 Name of Instrument/(BL No.) HRC (BL12)	
実験責任者名 Name of principal investigator Kazuaki Iwasa	実施日 Date of Experiment 2012.5.17 – 5.19	
所属 Affiliation Department of Physics, Tohoku University	2012.10.24 – 10.29	

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)

Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.
Polycrystalline samples of filled skutterudite $(\text{Pr}_{0.95}\text{Ce}_{0.05})\text{Ru}_4\text{P}_{12}$ $(\text{Pr}_{0.85}\text{Ce}_{0.15})\text{Ru}_4\text{P}_{12}$

2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。) Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.
A filled skutterudite $\text{PrRu}_4\text{P}_{12}$ undergoes a metal–nonmetal transition at 63 K. The nonmetallic phase is characterized by antiferro-type ordering of a Pr 4f-electron high-rank multipole (C. Sekine <i>et al.</i> : Phys. Rev. Lett. 79 (1997) 3218, T. Takimoto: J. Phys. Soc. Jpn. 75 (2006) 034714). The shift of crystal-field-splitting (CF) levels and the alignment of two inequivalent level schemes are signatures of ordering of f-electron multipole (K. Iwasa <i>et al.</i> : Phys. Rev. B 72 (2005) 024414). At half of the Pr ions, the triplet level is lowered with decrease in temperature from an excited state to a ground state below 30 K, while the rest Pr ions conserve a singlet ground state. On the other hand, $\text{Pr}_{1-x}\text{Ce}_x\text{Ru}_4\text{P}_{12}$ exhibit reentrant-type metal–nonmetal transition (C. Sekine <i>et al.</i> : JPSJ 80 (2011) SA024). The systems of $x = 0.10 – 0.15$ undergo phase transition to a metallic phase below approximately 10 K. Magnetic susceptibilities are suppressed by the Ce doping. This phenomenon indicates uniform nonmagnetic singlet ground state at low temperature, in contrast to the magnetic triplet ground state at the half of Pr sites in the pure $\text{PrRu}_4\text{P}_{12}$. The aim of present study is to investigate the CF levels in the Ce-doped system, and to unveil the 4f-electronic state responsible for the reentrant transition. We performed inelastic neutron scattering experiment on the high-resolution chopper spectrometer HRC at BL12 of MLF. The sample enclosed in a cylindrical aluminum container was installed in a closed-cycle helium refrigerator. Inelastic spectra were measured between 7.5 and 70 K. We used incident neutron energy of 30.5 meV with energy resolution of 1.3 meV at the elastic scattering position.

2. 実験方法及び結果(つづき) Experimental method and results (continued)

Figure 1 shows inelastic neutron scattering spectra of Ce5% system measured at 10 and 44 K. In addition to the intensities at $E = 0$ dominated by incoherent scattering, strong inelastic signals are observed at 9 and 13 meV at 10 K, and these seem to shift to 8.5 and 11 meV at 44 K. The spectral evolution is very similar to that for pure $\text{PrRu}_4\text{P}_{12}$ in the nonmetallic phase. This result indicates that the sample with light Ce-doping of 5% does not influence dramatically on the metal–nonmetal transition, which is consistent with no clear reentrant transition was found in electrical resistivity data of this doping range. Figure 2 shows the results of Ce15% sample, which shows reentrant-type two phase transitions at 45 and 7.5 K. The spectrum measured at 9.4 K of Ce15% is similar to that at 44 K of Ce5% shown in Fig. 1. This means that the ground state CF level does not switch from singlet to triplet in Ce15% system. In addition, the spectrum of Ce15% drastically changes at 7.5 K just the temperature of reentrant transition. This spectrum does not identical also with that in the disordered metallic phase above 45 K. The reentrant metallic phase at the lowest temperature accompanies reconstruction of the 4f-electron CF state. The doping of Ce ions to $\text{PrRu}_4\text{P}_{12}$ suppresses the shift of CF levels in the ordering process, and modifies the high-rank multipole order parameter of Pr 4f electrons. From these experimental results, we conclude that the reentrant-type metal–nonmetal transition of the Ce-doped $\text{PrRu}_4\text{P}_{12}$ occurs by tuning the shift of CF levels corresponding to the magnitude of multipole order parameter.

We have a plan to continue this study using much higher Ce-doping system, which shows higher transition temperature for the reentrant metallic phase.

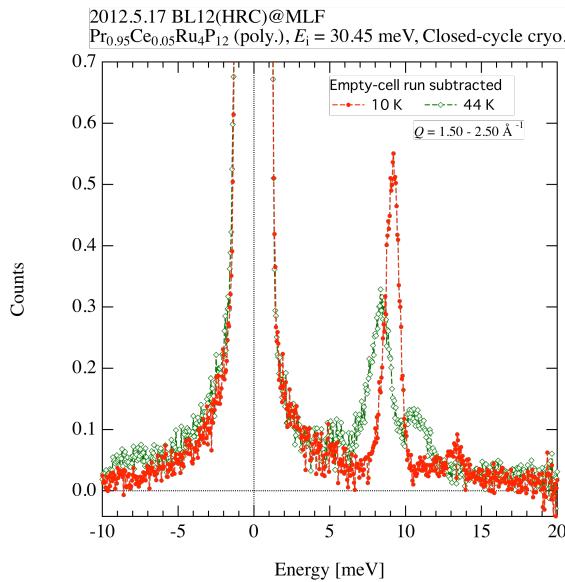


Fig. 1 INS spectra of Ce5% sample.

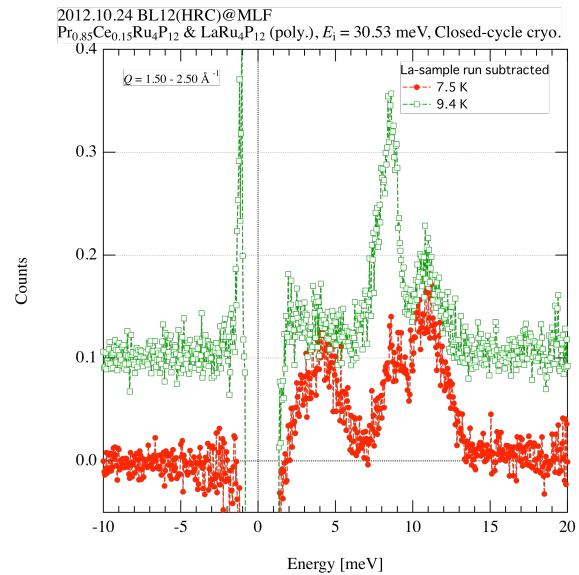


Fig. 2 INS spectra of Ce15% sample.