MLF Experimental Report	提出日 Date of Report 2013.4.5
課題番号 Project No. 2012B0115 実験課題名 Title of experiment Successive Metal-Nonmetal Transitions with Totally-Symmetric Electron Ordering in $(Pr_{1-x}Ce_x)Ru_4P_{12}$ 実験責任者名 Name of principal investigator Kazuaki Iwasa 所属 Affiliation Department of Physics, Tohoku University	装置責任者 Name of responsible person Shin-ichi Itoh 装置名 Name of Instrument/(BL No.) HRC (BL12) 実施日 Date of Experiment 2013.3.10 – 3.16

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 $(Pr_{0.80}Ce_{0.20})Ru_4P_{12}$

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.

$PrRu_4P_{12}$ undergoes a metal–nonmetal transition at 63 K (C. Sekine *et al.*: Phys. Rev. Lett. **79** (1997) 3218). The low-temperature nonmetallic phase is characterized by antiferro-type ordering of a Pr 4*f*-electron high-rank multipole (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 in the Pr bcc sublattice are signatures of ordering of *f*-electron multipole (K. Iwasa *et al.*: Phys. Rev. B **72** (2005) 024414). Half of the Pr ions carry the triplet ground state, whose energy is lowered with decrease in temperature. This state becomes a ground state below 30 K, while the rest Pr ions conserve a singlet ground state. On the other hand, $Pr_{1-x}Ce_xRu_4P_{12}$ exhibits a reentrant-type metal–nonmetal transition (C. Sekine *et al.*: JPSJ 80 (2011) SA024). The systems of $x = 0.10 - 0.15$ (synthesized under high pressures) undergo transition to a metallic phase below approximately 10 K, and magnetic susceptibilities are suppressed. 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 $PrRu_4P_{12}$. The aim of present study is to investigate the CF levels in the Ce-doped system (20% synthesized by a flux technique), and to unveil the 4*f*-electronic state responsible for the reentrant transition.

We performed inelastic neutron scattering experiment on HRC at BL12 of MLF. The sample enclosed in a cylindrical aluminum container was installed in a closed-cycle helium refrigerator. 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 measured at 5.3, 17.0, and 52.4 K of Ce20% sample, together with the data at 44 K of Ce5% (measured in 2012A). In addition to the intensities at $E = 0$ dominated by incoherent scattering, strong inelastic signals are observed at 8 and 11 meV at 17.0 K. These peaks shift to 6 and 11.5 meV at 52.4 K corresponding to the high-temperature metallic phase. On the other hand, the spectrum of Ce20% at 5.3 K in the reentrant-like metallic phase is composed of peaks at 4, 7, and 11 meV. Therefore, the 4*f*-electron CF level schemes in these three phases are not equivalent each other. In particular, we insist that the reentrant metallic state at the lowest temperature range is not identical with that at high temperature disordered phase. It means that the electronic state in these two metallic phases are not equivalent, while the electrical resistivity behaves as reentrant type. Another important feature in the Ce20% system is that the spectrum at 17 K just above the reentrant transition temperature is similar to that of the dilute system of Ce5% at 44 K, where the order parameter of multipole does not evolved fully. It indicates that the doping of Ce ions to $PrRu_4P_{12}$ suppresses the shift of CF levels in the ordering process, and also suppresses the high-rank multipole order parameter of Pr 4*f* electrons. From these experimental results, we conclude that the reentrant-type metal-nonmetal transition of the Ce-doped $PrRu_4P_{12}$ occurs by tuning the shift of CF levels corresponding to the magnitude of multipole order parameter.

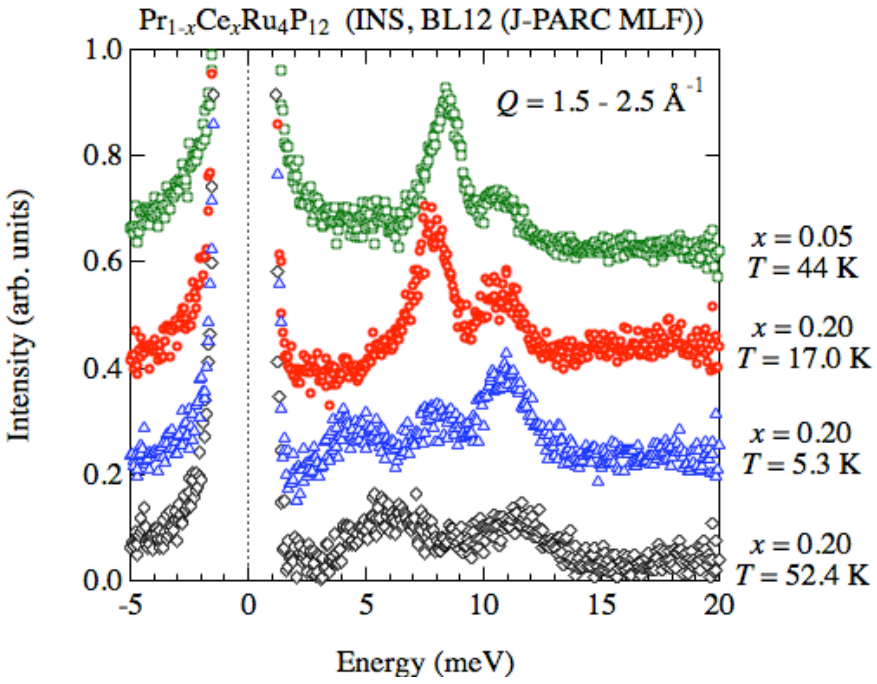

Fig. 1 INS spectra of Ce5% and 20% sample.