MLF Experimental Report	提出日 Date of Report
課題番号 Project No. 2014A0208 実験課題名 Title of experiment Magnetic Impurity Effect on the Metal-Nonmetal Transition with Totally-Symmetric Electron Multipole Ordering 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 2014.6.13 – 6.16 2014.6.20 – 6.23 2014.6.25 – 6.26

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

$(Pr_{0.85}Nd_{0.15})Ru_4P_{12}$

$PrRu_4P_{12}$

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.

Rare-earth filled skutterudite compound, $PrRu_4P_{12}$, undergoes a metal-nonmetal transition at 63 K (C. Sekine *et al.*: Phys. Rev. Lett. **79** (1997) 3218). The 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 shifts of crystal-field-splitting (CF) levels of the periodically aligned two inequivalent Pr sites with structural superlattice formation are signatures of ordering of *f*-electron multipole (K. Iwasa *et al.*: Phys. Rev. B **72** (2005) 024414, J. Phys. Soc. Jpn. **74** (2005) 1930). $Pr_{1-x}Ce_xRu_4P_{12}$ (x = 0.10 – 0.15) exhibits reentrant-type transition to a metallic phase below approximately 10 K (C. Sekine *et al.*: JPSJ **80** (2011) SA024). Magnetic susceptibilities are suppressed by the Ce doping, and the CF excitation spectra also exhibit drastic variation at 10 K (K. Saito *et al.*: Phys. Rev. B **89** (2014) 075131). 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 $PrRu_4P_{12}$. With respect to this previous investigation, the aim of present study is to investigate modification of the ordered phase by Nd doping as a magnetic impurity.

We performed inelastic neutron scattering experiment on the high-resolution chopper spectrometer HRC at BL12 of MLF. The samples of $Pr_{0.85}Nd_{0.15}Ru_4P_{12}$ and $PrRu_4P_{12}$ enclosed in a cylindrical aluminum container were installed in a closed-cycle helium refrigerator. Inelastic spectra were measured between 7 and 70 K. We used multi-E_i setup with of 8.82, 30.6, and 94.5 meV.

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 pure (top panel) and Nd15% system (bottom panel) measured at 7 and 43 K by using E_i = 30.6 meV. Strong and sharp inelastic signals are observed at 9 and 14 meV at 7 K, and these shift to 8.5 and 11 meV at 43 K, respectively. The spectral evolutions of these compounds are very similar to each other. This result indicates that the nonmetallic multipole ordered phase is robust against the Nd-doping of 15%, in contrast to the switch of the CF ground state in the Ce15%-doped material. Such robustness is consistent with the slightly reduced transition temperature from 63 K of the pure compound to 58 K of the Nd15% sample, as seen in the emergence of the superlattice structure.

On the other hand, we found difference in the energy spectra between the two compounds. In the Nd15% result, intensities near 2.5 meV at 7.8 K is slightly stronger than those at 43 K. In contrast, the spectra at 7 and 43 K of the pure system are almost identical in this energy range. In order to see behavior in the lower-energy range, the spectra was measured with E_i = 8.82 meV, as shown in Fig. 2. The intensity enhancement below 2 meV at lower temperature arises for the Nd15% system, while the spectrum of the pure system does not depend strongly on temperature. The enhancement of intensity indicates that a lower-energy excited level appears in the Nd15% system. Intensities at the neutron energy loss side (negative energy) is stronger at high temperature, which can be ascribed to transition from this low-energy excited level to the ground state together with phonon annihilation process. We emphasize that the excitation at approximately 2 meV is characteristic for the Nd-doped system. One can expect that the 2-meV peak corresponds to excitation between CF splitting levels of a Nd ion and quasi-degeneracy of the Nd-ion *f* electrons down to low temperatures. It may indicate that the Nd ions possess effective multipole degrees of freedom. If such multipole is compatible with that of the neighboring Pr ions, the impurity due to the Nd substitution does not destroy the non-metallic multipole ordered phase. We will analyze the obtained spectra more quantitatively, in order to reveal the Nd CF state and the possible multipole degrees of freedom.

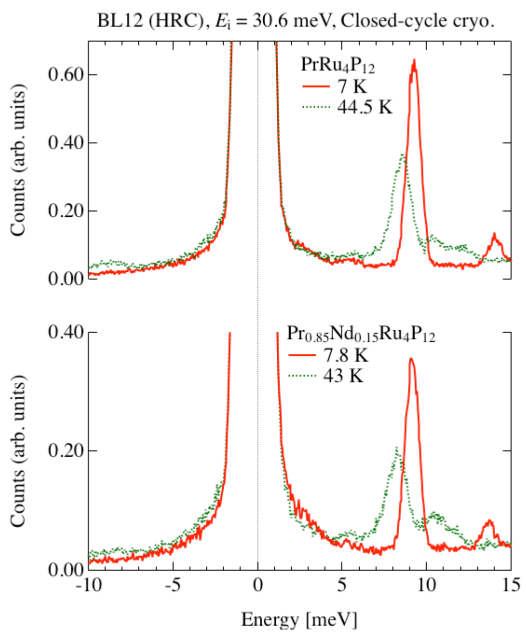

Fig. 1 INS spectra of the pure and Nd15% samples measured at 7 and 43 K. Incident neutron energy is E_i = 30.6 meV.

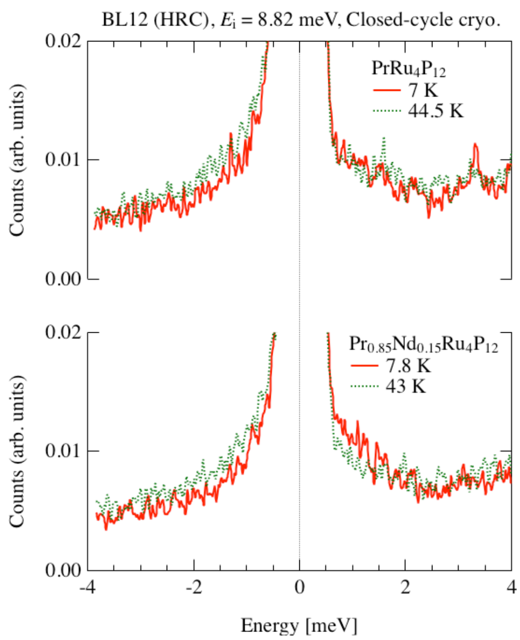

Fig. 2 INS spectra of the pure and Nd15% samples measured at 7 and 43 K. Incident neutron energy is E_i = 8.82 meV.