MLF Experimental Report	提出日 Date of Report 2010.7.1
課題番号 Project No. 2009B0003 実験課題名 Title of experiment Actual Ground State of Higher-Rank Multipolar Ordered State in Pr-filled Skutterudite 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 Kenji Nakajima 装置名 Name of Instrument/(BL No.) BL 14 実施日 Date of Experiment 2010.5.24 – 5.27

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.

6 g of a polycrystalline sample of filled skutterudite $PrRu_4P_{12}$.

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.

A filled skutterudite compound $PrRu_4P_{12}$ undergoes a metal–nonmetal transition that is expected to originate from the high-rank multipole ordering of Pr 4*f* electrons (C. Sekine *et al.*: Phys. Rev. Lett. **79** (1997) 3218, T. Takimoto: J. Phys. Soc. Jpn. **75** (2006) 034714). It accompanies the shift of crystal-field-splitting levels (K. Iwasa *et al.*: Phys. Rev. B **72** (2005) 024414). In particular, the excited triplet level above the transition temperature (63 K) is lowered with decrease of temperature, and it becomes a ground state below around 30 K. It has remains unsolved how the triplet ground state degeneracy would be lift to release the 4*f*-electron entropy. This issue is important for understanding the multipole order parameter and the weak nonmetallic state without diverging electrical resistivity down to the lowest temperature. The specific heat data indicates that the triplet splits by energy of 1 K. We performed inelastic neutron scattering experiment by the high-resolution chopper spectrometer AMATERAS at BL14, in order to resolve such ground state splitting.

The sample enclosed in a cylindrical aluminum container was installed in a helium-gas close refrigerator of BL14. Inelastic spectra were measured between 6 and 70 K. We used three conditions of incident neutron energies E_i (energy resolution): (1) 3.13 meV (35 μeV), (2) 1.69 meV (20 μeV), (3) 1.05 meV (12 μeV), as the multi E_i -mode by the chopper frequency of 300 Hz for CH01 and 02 and the window width of 10 mm for CH02.

Figure 1 shows a measured intensity contour map in the Q–E space under the measurement condition (1). In addition to the intensities at E = 0 dominated by incoherent scattering, two inelastic signal lines showing

quadratic dependences on Q appears on both Stokes and anti-Stokes sides. The similar lines were observed also under the condition (2), at all temperatures. Although the origin of the inelastic lines were unclear during the experiment, it was later suggested by the J-PARC instrumental teams that the lines were due to scattering from radiation shield chambers of the refrigerator.

Figure 2 depicts energy profiles at Q = 1.9 Å^{-1} under the condition (1). The sharp peaks at +0.21 and −0.24 meV independent of temperature are attributed to the scattering from the radiation shield. The peaks at +0.12 and −0.13 meV obtained under the condition (2) shown in Fig. 3 also arise from the same scattering process. Except these peaks, we found increase of intensities in the range between −0.2 and +0.2 meV at 6 K compared to 70 K, as shown by solid squares for the spectral difference. It implies the energy spectrum change through the transition temperature. We can assign the increasing intensity at lower temperatures to the excitations between the splitting levels form the triplet state in the ordered phase.

We will continue the study for clarifying the observed promising intensity composed of well-defined excited states or quasi-elastic component that may arise from hybridization of 4f electrons with a low-density carrier in the weak nonmetallic phase. For this purposes, we should pursue success of much higher-resolution measurement under the condition (3) that did not give clear signals at present, as well as diminishing the backgrounds from the refrigerator.

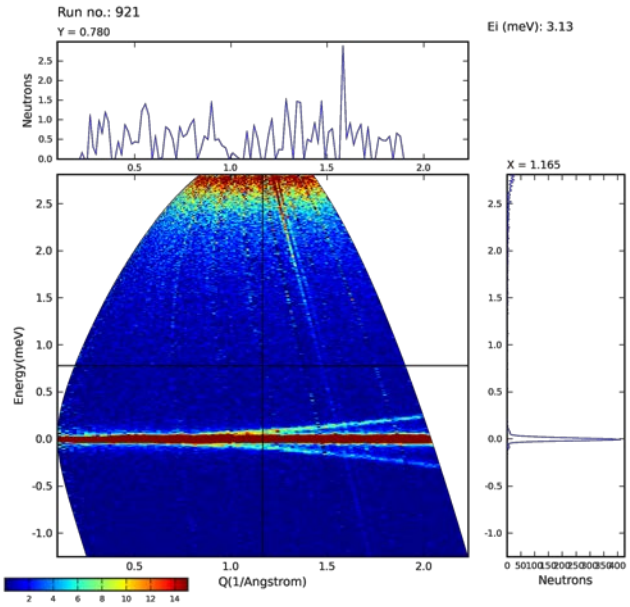

Fig. 1 An intensity contour map at 6 K in the Q-E space under the measurement condition (1) (E_i = 3.13 meV).

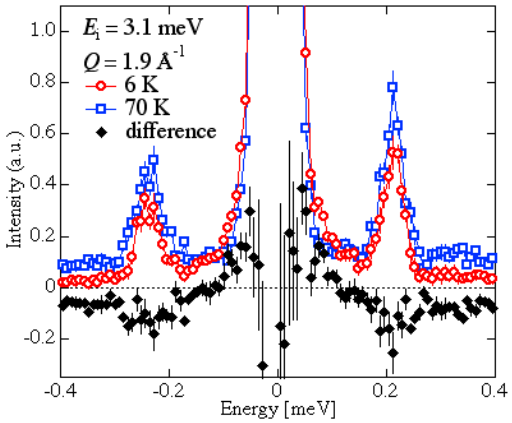

Fig.2 Energy spectra at Q = 1.9 Å^{-1} under the measurement condition (1).

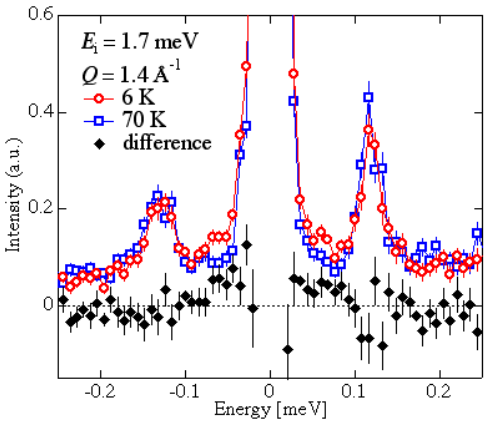

Fig.3 Energy spectra at Q = 1.4 Å^{-1} under the measurement condition (2).