MLF Experimental Report	提出日 Date of Report 2012.12.27
課題番号 Project No. 2012A0065 実験課題名 Title of experiment Magnetic excitation in the electron - doped heavy- electron system $Pr(Fe_{1-x}Co_x)_4P_{12}$ 実験責任者名 Name of principal investigator Hao Lijie 所属 Affiliation China Institute of Atomic Energy	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 2012.5.18-2012.5.21 2012.10.27-2012.10.31

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.

Filled Skuterudite Compound

$Pr(Fe_{0.97}Co_{0.03})_4P_{12}$

$Pr(Fe_{0.90}Co_{0.10})_4P_{12}$

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.

$PrFe_4P_{12}$ exhibits an anomalous phase transition from a heavy electron state(HF) to a non-magnetic ordered state below T_A = 6.5 K. Numerous experimental results and theoretical calculations suggested that the ordered state is a new type one in the Kondo system. Recently, a theoretical study reveals that the HF state of $PrFe_4P_{12}$ attributes to the Kondo effect with the low-energy quasi-quartet crystal-field-splitting (CF) of 4*f* electrons. The ordering state below T_A was explained as a staggered order of the Kondo-singlet sites and localized CF-singlet sites. [S.Hoshino, et al.: J. Phys. Soc. Jpn. 80 (2011) 033703. Y. Kuramoto, et al. : J. Phys. Soc. Jpn. 80 (2011) SA018]. In this theory, the two Fermi surfaces with quarter filling is necessary for explaining the ordering in $PrFe_4P_{12}$. Since the density of states of Fe-3*d* election has a very sharp peak close to the Fermi level according to the band calculation study, we expect that close to the Fermi level according to the band calculation study, we expect that *d*-electron dope will influence the HF state and the non-magnetic ordered state of $PrFe_4P_{12}$. Therefore, we performed inelastic neutron scattering experiment by the cold-neutron disk-chopper spectrometer AMATERAS at BL14, to observe the magnetic excitation of $Pr(Fe_{1-x}Co_x)_4P_{12}$.

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

The measurements were preformed by multi Ei method where the incident neutron energies were 6.39, 11.66, and 27.60 meV. The sample enclosed in a cylindrical aluminum container was installed in a helium-gas close refrigerator. Inelastic spectra were measured between 6 and 60 K.

We measured two polycrystalline sample of $Pr(Fe_{1-x}Co_x)_4P_{12}$ (x=0.03 and 0.10). The Figure 1 shows a typical spectrum of x = 0.03 as integration of intensity in the reciprocal-space range Q = 1.75 - 1.85 $Å^{-1}$ at 5.5 K. Blue triangles represent the difference between the sample measurement (black circles) and the empty cell one (red squares). The sharp peaks were observed around 2.5 and 10 meV. Figure 2 shows temperature dependence of spectrum. With increasing temperature the intensities of peaks become weak and almost disappear around 60 K. The data for Co 10% shows similar spectra.

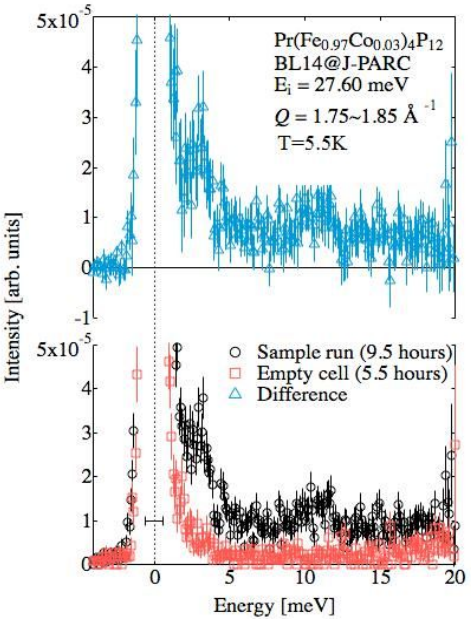

Figure 1. Inelastic neutron scattering spectra of $Pr(Fe_{0.97}Co_{0.03})_4P_{12}$. Excitation peaks around 2.5 meV and 10meV.

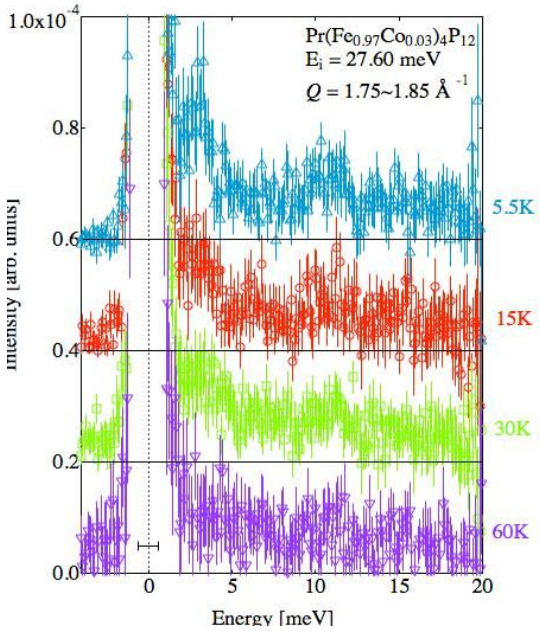

Figure 2. Temperature dependence of the spectrum at Ei=27.60meV.

In our previous inelastic neutron studies for $PrFe_4P_{12}$, we observed broad quasielastic spectra above T_A indicating the HF state arising from hybridization between 4f and conduction electrons. Below T_A, the spectrum is composed of sharp peaks around 1.5 and 3.0 meV.[K. Iwasa, et al.: Acta Physica Polonica B 34 (2003) 1117, J. Phys. Soc. Jpn. 81 (2012) 094711.] . Based on the theoretical study as described above, the sharp peak at 1.5meV can be reproduced by the low-energy CF-splitting excitation between Γ_1 CF ground state and $\Gamma_4^{(1)}$ excited state, while the peak at 3 meV is a broad response expressed by a Lorentzian-like form. By comparing these previous results with the present result for the Co-doped compounds, the different positions and temperature dependence of the peaks indicate distinct variation of 4f-electron state, which is also validated from behaviors in magnetic susceptibility measurements. It would be concluded that the Co doping suppresses the HF state, and enhances the localized nature of Pr 4f electrons which is not identical with that in the ordered phase of pure $PrFe_4P_{12}$.