MLF Experimental Report	提出日 Date of Report 2015.12.10
課題番号 Project No. 2015A0060 実験課題名 Title of experiment Magnetic excitations in heavy-electron ferromagnets NdT_4P_{12} (T = Fe and Ru) 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 2015.10.28 – 2015.11.5

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.

$NdFe_4P_{12}$ single crystal

$NdRu_4P_{12}$ powder

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 compounds, $NdFe_4P_{12}$ (L. Keller et al., J. Alloys and Comp. **323-324**, 516 (2001)) and $NdRu_4P_{12}$ (H. Sugawara and C. Sekine, private communication, S. Masaki et al., PRB **78**, 094414 (2008)) exhibit ferromagnetic ordering at 1.9 and 1.6 K, respectively. In addition, the electrical resistivity data of $NdFe_4P_{12}$ show an increment with decreasing temperature below approximately 20 K (H. Sato et al., PRB **62**, 15125 (2000)). This behavior is considered to be caused by the Kondo effect because the observed phenomenon of resistivity is reproduced by a function of temperature, $-\log T$. On the other hand, a less significant anomaly in the resistivity was found for $NdRu_4P_{12}$ (T. Utsumi et al., Muroran Inst. Tech. Journal **49**, 93 (1999) (in Japanese)). In order to investigate electronic correlation owing to hybridization between rare-earth 4*f* electrons and conduction electrons in these compounds, we performed inelastic neutron scattering (INS) experiments to identify magnetic excitation spectra at low-temperature region. As described below, we observed magnetic excitation spectra for both compounds. In addition, we attempt to measure INS spectra below 1 K by using the ^{3}He closed-cycle refrigerator and the oscillating collimator, which are installed to HRC (BL12).

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

Single crystalline sample of $NdFe_4P_{12}$ and that of polycrystalline sample of $NdRu_4P_{12}$ were synthesized by the Sn-flux method. The INS measurement for $NdFe_4P_{12}$ was carried out using the ^{3}He closed-cycle refrigerator, which was rotated by a goniometer, and the oscillating collimator. For the measurement for $NdRu_4P_{12}$, the GM cryostat was used without the oscillating collimator.

Figure 1 shows a measured INS spectrum of $NdFe_4P_{12}$ (open squares), together with background data for an empty sample cell (open triangles). Close circles are results of subtraction of the background data from the sample data, which give an actual excitation spectrum of $NdFe_4P_{12}$. A horizontal bar located between 20–27 meV indicates the instrumental resolution. The excitation spectrum is remarkably broader than the resolution.

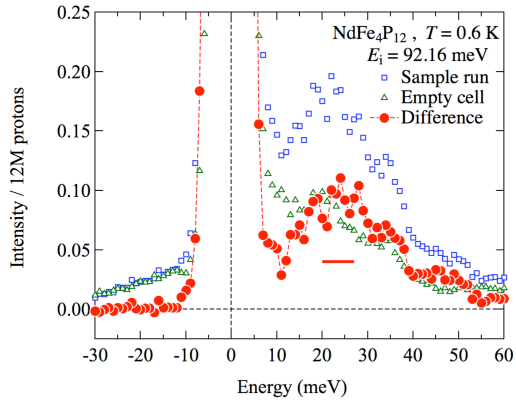

Fig. 1. INS spectra of $NdFe_4P_{12}$. Squares and triangles are data of the sample run and the empty-cell run, respectively. The difference shown by filled circles corresponds to the excitation in $NdFe_4P_{12}$. A horizontal bar located at 20–27 meV indicates the instrumental resolution width (FWHM) for E_i = 92.16 meV.

Figure 2 shows a result of INS measurement for $NdRu_4P_{12}$. Although we had no time to measure background data, we identified a sharp peak located at 27 meV, whose spectral width is almost the same as that of the resolution. We also detected a peak at 152 meV (not shown here). These two peaks can be attributed to excitations between three crystalline-electric-field (CEF) levels, which appear under the cubic point group at the Nd-ion site of this compound. Intensity above 30 meV seen in Fig. 2 does not depend on temperature, so that this is not ascribed to excitations of the sample.

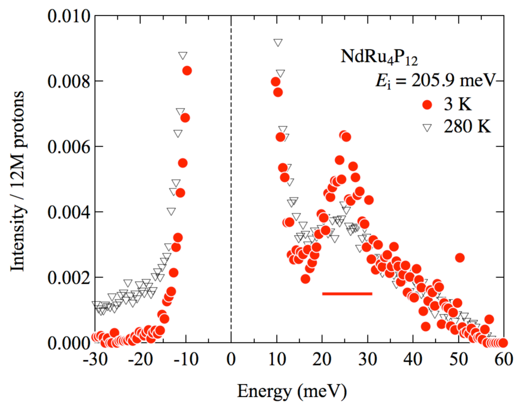

Fig. 2. INS spectra of $NdRu_4P_{12}$. Circles and triangles are data measured at 3 and 280 K, respectively. A horizontal bar located at 20–31 meV indicates the instrumental resolution width (FWHM) for E_i = 205.9 meV.

The same excitation could be expected also for $NdFe_4P_{12}$. Therefore, the broadening of INS spectrum of $NdFe_4P_{12}$ is suggestive of coupling of the 4f electrons with other degrees of freedom. As for the heavy-fermion system, hybridization between the 4f and conduction electrons is a candidate explanation to the spectral broadening. However, the momentum-transfer dependence of the INS intensity is not simply reproduced by the magnetic form factor of Nd^{3+} ion state. Another mechanism for the spectral broadening is coupling between the CEF excitation and the phonon modes.

In order to investing further the effect of phonon mode on the CEF excitation as well as the ferromagnetic heavy-electron behavior, it is necessary to measure temperature dependence of the broad spectrum.