Crystal structure and Magnetic Property of $Pr_xFe_4Sb_{12}$

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Rare-earth filled skutterudite compounds have been studied for various phase transitions of $4f$ electron states. $Pr_xFe_4Sb_{12}$ has been considered to exhibit magnetic ordering at around 4 K. It is suggested that not only Pr $4f$ but also Fe $3d$ electrons give the ordered moment (N. P. Butch *et al.*: Phys. Rev. B 71 (2005) 214417), since the magnitude of effective magnetic moment estimated from the high temperature magnetic susceptibility is larger than that of Pr^{3+} free ion. The ordered structure of the two kinds of magnetic moments depending on the Pr filling has been unsolved yet. It is notable that the magnetic phase transition is reported to disappear in case of full occupation of the Pr-ion sites ($x = 1$). The effect of Pr filling to the magnetic ordering has also not been explained.

We performed neutron scattering experiment using the triple-axis spectrometer TOPAN (6G) in order to reveal Pr-ion crystal field levels by using powdered simple and the four-circle diffractometer FONDER (T2-2) to investigate the crystal and magnetic ordered structures by using a single crystalline sample. In this year, we measured crystal field levels in the high-pressure synthesis sample, in order to compare that in the unfilled sample by the so-called Sb-self method. The sample for FONDER was synthesized by the flux method, and the Pr concentration x is expected to be less than unity ($x = 0.7 - 0.9$), as was reported in the previous reports.

Figure shows inelastic spectra observed at TOPAN. We succeeded in observing two magnetic excitation peaks at 2.4 and 11 meV. The peak positions are almost equivalent with those of the x 1 one synthesized by K. Tanaka *et al.* using the high-pressure method (J. Phys. Soc. Jpn. 76 (2007) 103704) but also of the $x < 1$ sample reported by E. Bauer *et al.* (J. Magn. Magn. Mater. 310 (2007) 286) . Therefore, it is still under controversial that the magnetic nature of $4f$ electrons localized at Pr ions does not strongly depend on the Pr ion concentration or not. We are now trying to analyze the intensity to discuss the crystal field level schemes and their role on the magnetic ordering. In the Experiment at FONDER, we succeeded in observing many reflections that is expected to allow the crystal structure determination (Pr concentration). In addition, the increase of fundamental reflection intensities with decreasing temperature thorough the transition temperature. This results is consistent with the previous study, and the magnetic ordering pattern is composed of ferromagnetic component. However, the magnetic ordering signal depends on samples. We will carry out a subsequent measurement with longer wave length setup of FONDER to focus the low-Q magnetic intensities.

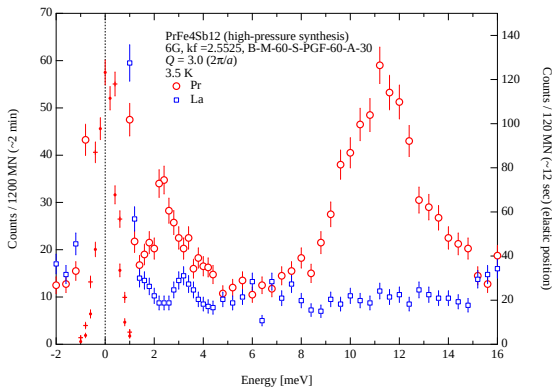

Fig. 1. Magnetic excitation spectra of $Pr_xFe_4Sb_{12}$ synthesized by the high-pressure method and $La_xFe_4Sb_{12}$ at 3.5 K.