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Multipolar ordering phase transition of Pr-filled skutterudite

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Neutron diffraction experiments of $PrFe_4P_{12}$ revealed antiferromagnetic (AFM) structures composed of two different magnetic moments induced along the magnetic fields along the [1, 0, 0] and $[1, \bar{1}, 0]$ axes in the ordered phase (L. Hao *et al.*: Acta Physica Polonica B **34** (2003) 1113, Physica B **359-361** (2005) 871). This result has been thought to be consistent with the antiferro-type ordering of a quadrupole O_2^0 accompanied by the Fe displacement whose coordinate is $(1/4+\delta, 1/4+\delta, 1/4-2\delta)$ with the ordering wave vector $\mathbf{q} = (1, 0, 0)$. However, the overall physical features including the magnetic-field induced AFM moment as well as the phase diagram cannot be explained simply by the quadrupolar degree of freedom. Recently, in the metal-nonmetal transition of $PrRu_4P_{12}$, a hexadecapole moment becomes finite and plays a crucial role in the transition (K. Iwasa *et al.*: PRB **72** (2005) 024414, JPSJ **74** (2005) 1930, Y. Kuramoto *et al.*: Prog. Theor. Phys. Suppl. **160** (2005) 134, T. Takimoto: JPSJ **75** (2006) 034714). Therefore, we have to reconsider the order parameter of the low-temperature phase of $PrFe_4P_{12}$. We examined the Γ_1-type Fe displacement (the coordinate is $(1/4+\delta, 1/4+\delta, 1/4+\delta)$) without breaking the local cubic symmetry at the Pr-ion sites, as in the nonmetal phase of $PrRu_4P_{12}$. This model was found to be able to reproduce the observed intensities of polarized neutron diffractions at the superlattice points, as shown in Figure 1. This result demonstrate that not only the quadrupole O_2^0 but also others keeping the cubic symmetry is a candidate of the order parameter in $PrFe_4P_{12}$.

In this talk, we will present also the recent data of magnetic excitation spectra below and above the transition temperature of $PrFe_4P_{12}$ and La-substituted systems.

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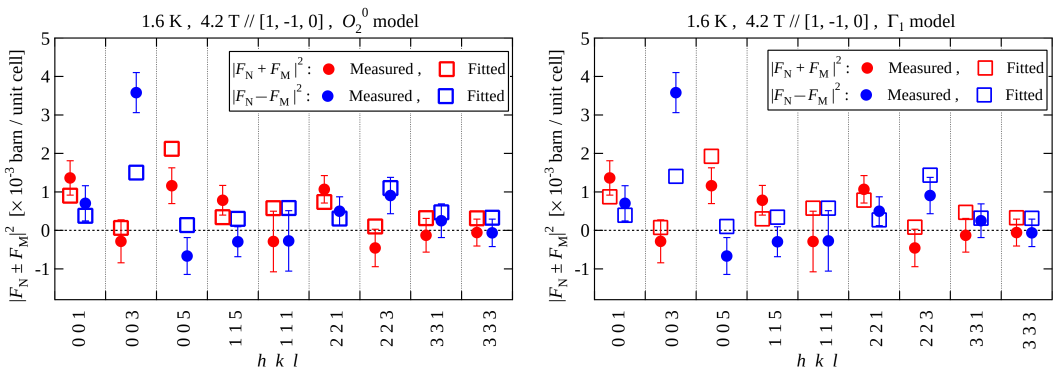

Figure 1: Solid symbols represent the measured $|F_N + F_M|^2$ and $|F_N - F_M|^2$ at the superlattice points under the magnetic field of 4.2 K parallel to the $[1, \bar{1}, 0]$ axis, where F_N and F_M are the nuclear and magnetic structure factors, respectively. The lefthand and righthand figures contain the calculated values based on the O_2^0 ordering with the Fe displacement of $(1/4+\delta, 1/4+\delta, 1/4-2\delta)$ and the Γ_1-type ordering with $(1/4+\delta, 1/4+\delta, 1/4+\delta)$, respectively. Both results give the similar agreement with the experimental data.