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Antiferro-quadrupolar ordering of $PrFe_4P_{12}$ studied by polarized neutron diffraction

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$PrFe_4P_{12}$ has attracted much attentions due to its very heavy electron mass of $81m_0$ and unusual phase transition at T_A=6.5 K. Below T_A, X-ray superlattice reflections characterized by the wave vector q=[1,0,0] were observed and the scattering pattern were fully explained by the formation of crystal structure modulation mainly with Fe-ion displacements. In our previous unpolarized neutron diffraction study, the magnetic-field induced antiferromagnetic(AFM) reflections were observed below T_A, demonstrating that the $4f$ electrons of Pr ions undergo antiferro-quadrupolar(AFQ) ordering. In order to investigate the order parameters of AFQ phase, we carried out polarized neutron diffraction experiments to observe interference effect of scattering amplitudes due to the Fe-ion displacement and the field-induced magnetic structure.

Below T_A, scattering intensities clearly depend on the spin direction of the incident neutrons. Fig.1 shows the obtained $|F_N + F_M|^2$ and $|F_N - F_M|^2$ under magnetic fields along the [0,0,1] and [1,-1,0] directions by circles, where F_N and F_M are structure factors for nuclear and magnetic scattering, respectively. Because the superlattice reflection patterns under magnetic field applied along the [0,0,1] and [1,-1,0] directions were explained well by sum of the reflections due to induced AFM components along the field directions and Fe-ion displacements in the previous unpolarized neutron diffraction, the quadrupoles O_2^0 and O_2^2 are thought to be possible order parameters under T_h crystal-structure symmetry. If O_2^0=$(3J_z^2$-J(J+1))/2 is the dominant order parameter at [0,0,1] field direction, the Fe-ion displacement is naturally expressed as the atomic coordinate (1/4,1/4,1/4)+$(\delta, \delta, \delta')$. Using this model, the fitted $|F_N + F_M|^2$ and $|F_N - F_M|^2$ reproduces well the observed values as shown in Fig.1(a) by bars. On the other hand, if the $O_2^2 = \sqrt{3}(J_x^2 - J_y^2)/2$ were assumed as dominant order parameter, the Fe-ion displacement is expressed as $(\delta, -\delta, 0)$ and no satisfactory fit to the data is obtained. Therefore, O_2^0 is the dominant order parameter at 1.6K and 3.6T along the [0,0,1] direction. Under the [1,-1,0] field direction, the clear interference effect cannot be explained by only O_2^2 order parameter because the reflection due to the coupled Fe-ion displacement with $(\delta, -\delta, 0)$ are expected to be too small to be observed at the measured reciprocal-lattice points. On the other hand, considering only O_2^0 order, the data are reproduced well as shown in fig.1(b) by bars. Therefore, the quadrupole O_2^0 is considered to exist also under the magnetic field along the [1,-1,0] axis.

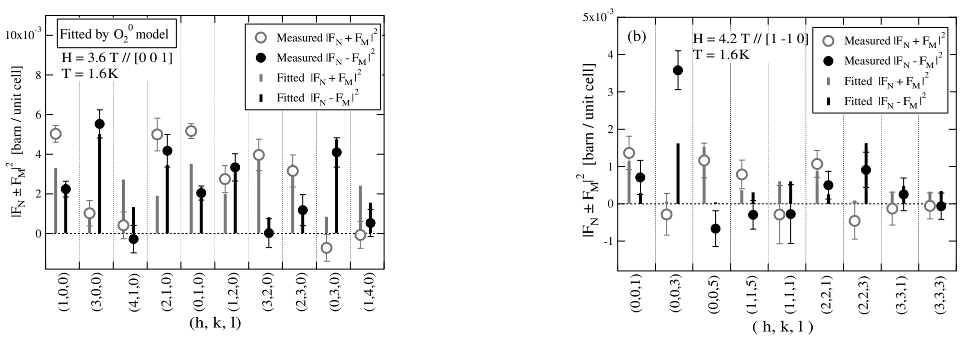

Figure 1: Circles are experimentally determined $|F_N+F_M|^2$ and $|F_N-F_M|^2$ for the various superlattice reflections along the [0,0,1](a) and the [1,-1,0](b) magnetic field directions. The bars are fitted results.