Antiferroquadrupole ordering in PrFe$_4$P$_{12}$ studied by resonant x-ray scattering

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Introduction

During the last two decades many experiments using the resonant x-ray scattering (RXS) technique have been performed to observe an ordering of electronic degrees of freedom. RXS is a combined technique of diffraction and spectroscopy, and can elucidate a spatially ordered electronic states, such as charge, magnetic, and orbital order. We applied RXS to a filled skutterudite PrFe$_4$P$_{12}$, in which an antiferroquadrupole ordering of Pr 4$f$ electrons were proposed. The phase transition at $T_A = 6.5$ K is confirmed by the specific heat measurement [1], but no magnetic reflection was observed in neutron powder diffraction [2]. On the other hand, lattice distortion and magnetic field induced antiferromagnetic moment were observed at the same modulation wave vector [3, 4]. These facts strongly suggest that the phase transition at $T_A$ is accompanied by an antiferroquadrupole ordering. Last year we have reported the RXS study, in which we observed a resonant signal at the Pr-L$_{III}$ absorption edge below $T_A$ [5]. The result suggests that two Pr atoms in the bcc unit cell are in different electronic state below $T_A$, which is probably an antiferroquadrupole ordering. We proceeded the experiment furthermore to understand the nature of RXS signal in PrFe$_4$P$_{12}$.

Experimental

Single crystals of PrFe$_4$P$_{12}$ were grown by a tin-flux method. X-ray scattering experiments were carried out at beamline 4C and 16A2 at Photon Factory, KEK.

Results and Discussion

Figure 1 shows the temperature dependence of the intensity of (111) reflection at resonant (Pr-L$_{III}$ absorption edge) and non-resonant energies. The intensity at both energies increases gradually with decreasing temperature below $T_A$. The intensity of the reflection contains in general both components of lattice distortion and ordered electronic state. However, the latter is strongly enhanced in the intensity at the resonant energy due to the anomalous scattering. Quite similar temperature dependence at two energies indicates that the lattice distortion and ordering of electronic state of Pr occur and evolve simultaneously.

![Figure 1: Temperature dependence of the intensity of (111) reflection at resonant ($E = 5.962$ keV) and non-resonant ($E = 5.940$ keV) x-ray energies.](image)

References


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