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Antiferroquadrupole ordering in $PrFe_4P_{12}$ 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 $PrFe_4P_{12}$, 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 $PrFe_4P_{12}$.

Experimental

Single crystals of $PrFe_4P_{12}$ 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.

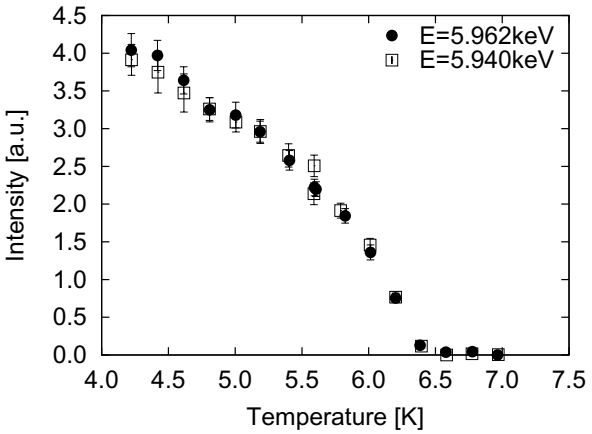

Figure 1: Temperature dependence of the intensity of (111) reflection at resonant (E = 5.962 keV) and non-resonant (E = 5.940 keV) x-ray energies.

We have tried to measure the azimuthal angle dependence to know the local symmetry of Pr sites in the ordered phase. We selected the resonant energy of 5.962 keV and measured (111), (300), and (210) reflections. Polarization of scattering x-rays were analyzed using Al (220) crystal for (300) and (210) reflections. The observed intensity was almost independent of the azimuthal angle for three reflections. Even though the existence of the domains in the crystal, which appear below T_A, smear out a part of anisotropy, the isotropic part of scattering tensor is much larger than the anisotropic part. It might be related to the fact that the 4*f* electrons of Pr is close to an itinerant state as evidenced by a heavy-fermion-like behavior under magnetic field, and the spread of the wave function is fairly isotropic.

References

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