Resonant x-ray scattering study on the filled skutterudite $PrFe_4P_{12}$

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Introduction

The filled skutterudites RT_4X_{12} (R = rare earth, T = Fe, Ru, and Os, X = P, As, and Sb), which crystallize in a *bcc* structure, exhibit a wide variety of electronic properties, such as superconductivity, magnetic order, and metal-insulator transition. Among them, the interest in $PrFe_4P_{12}$ is an anomalous ordered state at low temperature. 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 which is characterized by the modulation wave vector of $\mathbf{q} = [1, 0, 0]$ was observed below T_A in recent X-ray diffraction study [3], and magnetic field induced antiferromagnetic moment with the same characteristic wave vector $\mathbf{q}$ as above was also observed at the temperatures below T_A by neutron diffraction experiment [4]. These facts strongly suggest that the phase transition at T_A is accompanied by an antiferroquadrupolar ordering. This ordered state is suppressed by magnetic field, and a heavy-fermion-like behavior appears, which is evidenced by large electronic specific heat coefficient and large cyclotron effective mass. Here we report the resonant x-ray scattering (RXS) experiments at the Pr-L_{III} absorption edge to investigate the ordered state of $PrFe_4P_{12}$. RXS is a combined technique of diffraction and spectroscopy, and can elucidate a spatially ordered electronic states, such as charge, magnetic, and orbital order.

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. We measured three reflections, (300), (111), and (210) which are forbidden in the *bcc* structure above T_A and become allowed below T_A due to the lattice distortion.

Results and Discussion

Figure 1 shows fluorescence and scattering intensity of (111) reflection near the Pr-L_{III} absorption edge. Two resonant features were observed around 5.965 eV and 6.000 eV. We also observed the resonance in (300) and (210) reflections. The scattering amplitude of the superlattice reflections at the absorption edge of Pr consists of two components, the lattice distortion and the resonant scattering of Pr. If both components are finite, interference term appears in the intensity. The assumption that Pr atoms do not change the position at the phase transition is reasonable in the structural model proposed experimentally [3] and theoretically [5]. Based on this assumption, resonant scattering term for the odd number of $h + k + l$ corresponds to the difference of anomalous scattering factor between two Pr atoms in the *bcc* unit cell. Anomalous scattering factor is closely related to the electronic states of resonating atom. Therefore our result suggests a spatial ordering of two different electronic states of Pr atom, which is probably an antiferroquadrupolar ordering, appears below T_A.

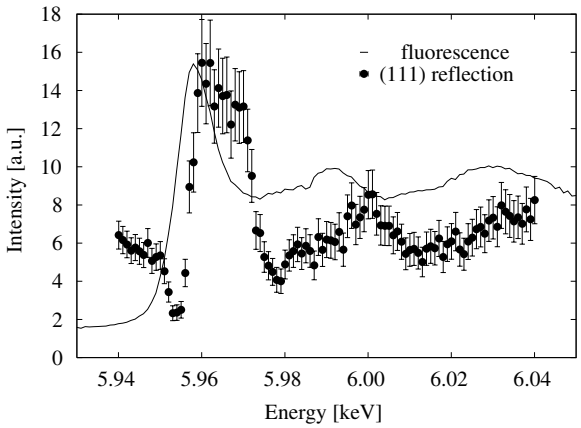

Figure 1: Energy dependence of fluorescence yield and (111) reflection

References

[1] T. D. Matsuda *et al*., Physica B **281-282**, 220 (2000).
[2] L. Keller *et al*., J. Alloys Compd. **323-324**, 516 (2001).
[3] K. Iwasa *et al*., Physica B **312-313**, 834 (2002).
[4] L. Hao *et al*., to be published .
[5] S. H. Curnoe *et al*., Physica B **312-313**, 837 (2002).

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