Order Parameter of Phase IV in (Ce,La)B6

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Cerium hexaboride CeB6 undergoes successive phase transitions from paramagnetic phase (Phase I) to antiferroquadrupolar ordering with a wave vector k = [1/2,1/2,1/2] at TQ = 3.3 K (Phase II) and to antiferromagnetic ordering with 4-k magnetic structure at TN = 2.3 K (Phase III). This complex magnetic phase diagram comes from the multipolar degrees of freedom in a Gamma8 quartet ground state of Ce3+ ion. By substituting La for Ce in this material, one can change the strength of the multipolar interactions. In the La diluted compound CexLa1-xB6, with decreasing x, TQ rapidly decreases, while TN slightly decreases. Then, a new ordered state called "Phase IV" appears at x < ˜ 0.8. In Phase IV, various unusual properties such as the large softening of the c44 mode in the ultrasonic measurement, the isotropic cusp in the bulk magnetic susceptibility, the trigonal lattice distortion along the [111] direction, have been reported. Recently the resonant X-ray scattering experiment (Mannix et al, Phys.Rev.Lett. 95(2005)117206) and the theoretical analysis (Kusunose and Kuramoto, J.Phys.Soc.Jpn. 74(2005)3139) have reported that the octupolar ordering with a wave vector [1/2,1/2,1/2] occours in Phase IV. In this case, the superlattice reflections at (h/2,k/2,l/2) with high Q-vectors are expected to be detected by neutron scattering because the magnetic form factor of magnetic octopole moment exhibits a maximum at a high Q-vector. Therefore we have performed the neutron diffraction experiment on (Ce,La)B6 in Phase IV.

The large single crystal of B11 99.52% enriched Ce0.7La0.3B6 was grown by the floating zone method. In this sample of Ce 70%, Phase IV is below ˜ 1.5K. The sample was mounted in a 3He-4He dilution refrigerator with (hkk) as horizontal scattering plane. The neutron diffraction experiment was performed on the thermal neutron triple axis spectrometer TOPAN (6G) with the incident energy Ei = 41 meV.

We scanned in reciprocal space along the main symmetry directions at lowest temperature of 0.2 K and found the superlattice reflections at (h/2,h/2,k/2). Furthermore the intensity of the superlattice reflections have a tendency to increase as increasing Q-vectors. This result is consistent with the octupolar ordering.

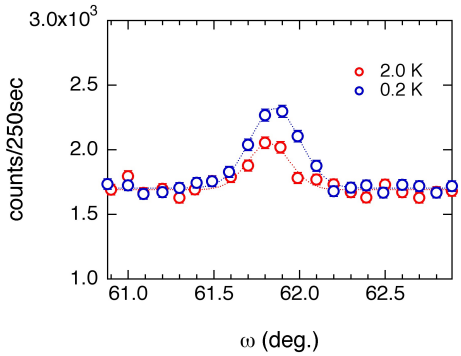

Fig. 1. Profile of Ce0.7La0.3B6 at (3/2,3/2,3/2) at 0.2 K and 2 K