Neutron-Diffraction Study of the Unusual Ordered Phases of $Ce_{0.75}La_{0.25}B_6$

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Abstract

$Ce_{0.75}La_{0.25}B_6$ shows paramagnetic phase (phase I), 4f-electron quadrupolar ordered phase (II), magnetically ordered phase (III), and an unknown phase IV. In order to investigate order parameters in phase III and IV, neutron diffraction experiments were performed down to 74 mK. Magnetic diffraction pattern measured in phase III is reproduced by a model in which spin arrangement is modified from the previously suggested structure for CeB_6. No magnetic reflections were observed in phase IV. Thus, the order parameter in phase IV is expected to be 4f-electron multipoles rather than magnetic dipoles.

Key words: $Ce_{0.75}La_{0.25}B_6$; electron multipole effect; neutron diffraction

1. Introduction

CeB_6 crystallizing in a Pm$\bar{3}$m cubic structure shows paramagnetic phase I with typical dense-Kondo-like behaviors and two ordered phases below $T_Q = 3.3$ K and below $T_N = 2.3$ K at zero magnetic field [1]. In phase III below T_N, an antiferromagnetic (AF) ordered structure appears, which is characterized by two components of propagation vectors of $\mathbf{k_1} = (1/4, 1/4, 0)$ and $\mathbf{k_2} = (1/4, 1/4, 1/2)$ [2]. In phase II ($T_N < T < T_Q$), a neutron diffraction experiment revealed a magnetic-field-induced AF structure with a propagation vector $\mathbf{k} = (1/2, 1/2, 1/2)$. The experimental fact was interpreted as an evidence for the quadrupole ordering in phase II, which was recently supported by a theory taking into account the 4f-electron multipole interactions [3].

Unusual ordered phases in diluted systems by substituting non-magnetic La ions for Ce ions have been studied extensively. For the case of $Ce_{0.75}La_{0.25}B_6$, a new phase (phase IV) appears in a small region of temperature ($1.2 < T < 1.6$ K) and magnetic field ($H < 0.7$ T) surrounded by other phases in the phase diagram [4]. A neutron scattering experiment revealed the AF ordering in phase III below 1.2 K, which is represented by the same propagation vectors as for CeB_6 [2]. Magnetization behaves isotropic with a cusp at 1.7 K, in contrast to the anisotropic larger magnetization in phase III [5]. Strong softening of elastic constant C_{44} in phase IV indicates an important role of the orbital degeneracy [4]. Recently, to explain these complicated physical properties, an octupole moment is suggested to be a principal order parameter of phase IV [5]. We performed a neutron diffraction experiment on $Ce_{0.75}La_{0.25}B_6$ in order to elucidate magnetic properties in phase IV and to determine the magnetic structure in phase III.

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2. Experimental procedure

Powder sample of $Ce_{0.75}La_{0.25}B_6$ was synthesized with 98.5% enriched ^{11}B to reduce neutron absorption. Magnetization of this sample was confirmed to be similar to that reported previously [5]. A neutron diffraction experiment with wave length of 2.42 Å was performed at a diffractometer D20 of ILL, Grenoble, France. Sample temperatures were controlled by a dilution refrigerator. Prior to this experiment, preliminary experiments with similar conditions were carried out at JAERI, Tokai, Japan and PSI, Villigen, Switzerland.

3. Experimental result and discussion

The upper pattern in Fig. 1 shows an intensity difference between 74 mK and 3.5 K. Distinct magnetic

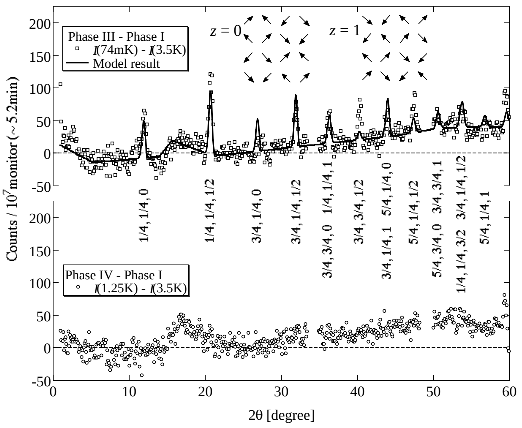

Fig. 1. An upper figure represents the intensity difference between 74 mK and 3.5 K. The present magnetic-structure model is depicted by arrows corresponding to Ce-ion magnetic moments, and z means the position of the (001) layer. A lower part shows the intensity difference between 1.25 K and 3.5 K.

reflections are indexed by $\mathbf{k_1}$ and $\mathbf{k_2}$ as in the previous study [2]. Gradual change of background intensity may be due to liquid helium inserted into the sample container to cool down the sample. It is notable that the observed pattern is not reproduced well by the magnetic-structure model for CeB_6 suggested by Effantin *et al* [2]. Their model gives intensity at (1/4, 1/4, 0) larger than that at (1/4, 1/4, 1/2), but the present experimental result is vise versa. Recently, Fischer also pointed out the similar discrepancy between the model and the measurement of pure CeB_6 [7]. A profile fitting procedure of FullProf [8] was adopted to examine several models, and we found a plausible structure shown in the inset. Like in the Effantin's model, we assumed all Ce-ion magnetic moments with a same magnitude aligning along the [110] direction. But the spin arrangement is different. A fitted result, indicated by a solid line, with $0.42\mu_B$/Ce and $\chi^2 = 1.41$ which is smaller than 1.70 by the fit based on the Effantin's model is in good agreement with the experimental data. Fischer proposed another model based on two kinds of magnetic moments by considering a structural symmetry in the ordered phases. It also reproduces the experimental data of $Ce_{0.75}La_{0.25}B_6$ and CeB_6 [7].

The lower pattern of the figure represents difference between intensity at 1.25 K in phase IV and that at 3.5 K in phase I. Any signal due to magnetic ordering was not observed within the present experimental accuracy. We conclude that a magnetic dipole is not a principal order parameter in phase IV. It is consistent with the suggested ordering of quadrupoles or octupoles without spontaneous magnetic dipoles in phase IV.

4. Concluding remarks

No clear magnetic dipolar ordering exists in phase IV and we proposes the more plausible magnetic-structure model in phase III. These facts are keys to understand the 4f-electron multipole effect in $Ce_{0.75}La_{0.25}B_6$.

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