Elastic Properties of the Kondo Compounds $Ce_xLa_{1-x}B_6$ ($x = 0.75$ and 0.70)

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Abstract

We have made thermal expansion measurements to examine the characters of an ordered phase IV in the dense Kondo compounds $Ce_xLa_{1-x}B_6$ (x = 0.75, 0.70). For both compounds, expansion along the [001] direction and shrinkage along [111] with decreasing temperature have been observed in phase IV. These results indicate the lattice distortion from cubic to trigonal structure. A ferro-quadrupole ordering of O_{yz}, O_{zx}, O_{xy} with Γ_5 symmetry is a probable candidate for the order parameter of the phase IV.

Key words: $Ce_xLa_{1-x}B_6$; quadrupole ordering; thermal expansion; Kondo compound

The dense Kondo compound CeB_6 and diluted compound $Ce_xLa_{1-x}B_6$ with a cubic CaB_6-type crystal structure has a Γ_8 ground state and a Γ_7 excited state at $\Delta \simeq 540$ K. Because the Γ_8 quartet possesses spin and orbital degrees of freedom, it is expected that magnetic interactions, quadrupole interactions and the Kondo effect compete each other in CeB_6. CeB_6 exhibits successive phase transitions of antiferro-quadrupole (AFQ) ordered phase II at T_Q = 3.3 K and antiferro-magnetic (AFM) phase II at T_N = 2.3 K. It is now established that the AFQ ordering in phase II characterized by O_{xy}-type quadrupole at $\boldsymbol{k}$ = [1/2 1/2 1/2] [1]. It has been pointed out that magnetic octupole moment T_{xyz} plays an important role to stabilize the AFQ phase II in magnetic fields [2].

In the case of diluted compound $Ce_{0.75}La_{0.25}B_6$, an ordered phase IV has been found between a paramagnetic phase I and AFM phase III [3,4]. Specific heat shows double sharp peaks at 1.6 K and 1.1 K corresponding to the I-IV and IV-III phase transitions [3]. Magnetization measurement indicates isotropic nature in phase IV [4]. μSR measurement does not detect the internal magnetic field due to magnetic ordering in phase IV [5]. It should be noted that a considerable softening of 30% in C_{44} has been observed in phase IV of $Ce_{0.75}La_{0.25}B_6$ [3]. In the case of $Ce_{0.70}La_{0.30}B_6$, the phase IV is stable down to T = 20 mK [6]. Although a possibility of octupole ordering in phase IV is discussed [7], the order parameter of phase IV has not been identified yet.

In the present study, we have measured thermal expansion in $Ce_xLa_{1-x}B_6$ for x = 0.75 and 0.70 by using three-terminal capacitance method. The capacitance cell is built in a handmade ^{3}He-cryostat with a superconducting magnet up to 14 T. Single crystals were grown by a floating zone method. Plane parallel surfaces of [001] and [111] in both compounds were prepared for the thermal expansion measurements. The present thermal expansion apparatus based on the capacitance method detects lattice distortion as high as $\Delta L/L \simeq 10^{-8}$.

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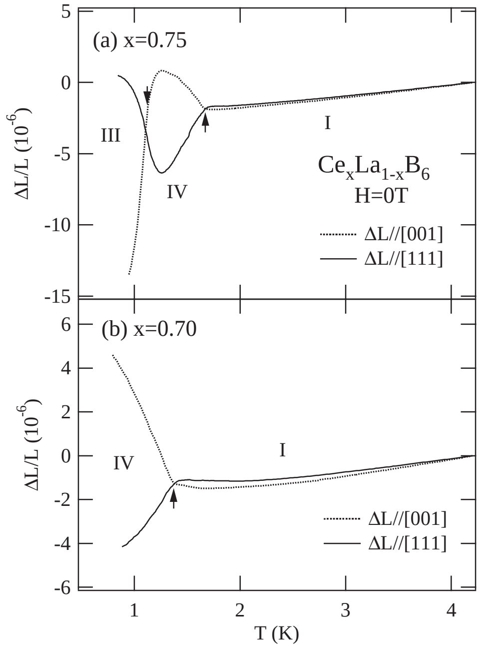

Fig. 1. The thermal expansion of $Ce_xLa_{1-x}B_6$ for (a) $x = 0.75$ and (b) $x = 0.70$ along the [001] and [111] directions in zero magnetic field. The arrows are indications of phase transition points.

Figs. 1 (a) and (b) show the thermal expansion along the fourfold [001] and body diagonal [111] directions on $Ce_{0.75}La_{0.25}B_6$ and $Ce_{0.70}La_{0.30}B_6$ in zero magnetic field, respectively. In the paramagnetic phase I, the length along both directions decreases monotonously with decreasing temperature for both compounds. It is remarkable that the length along the [001] direction expands, while the length along [111] shrinks in phase IV. Around IV-III phase boundary in $Ce_{0.75}La_{0.25}B_6$, the length shrinks rapidly along [001] and expands along [111] with decreasing temperature.

The expansion of $\Delta L/L$ along the [001] direction is described by using symmetry strain as $\Delta L/L_{[001]} = \varepsilon_B/3 + \varepsilon_u/\sqrt{3}$ and along [111] by $\Delta L/L_{[111]} = \varepsilon_B/3 + 2(\varepsilon_{yz} + \varepsilon_{zx} + \varepsilon_{xy})/3$. Where $\varepsilon_B = \varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}$ is a volume strain and $\varepsilon_u = (2\varepsilon_{zz} - \varepsilon_{xx} - \varepsilon_{yy})/\sqrt{3}$ is a tetragonal strain. The temperature dependence of $\varepsilon_{xy}(= \varepsilon_{yz} = \varepsilon_{zx})$ of $Ce_xLa_{1-x}B_6$ for $x = 0.75$ and 0.70 in Fig. 2 was obtained from the results of thermal expansion experiment in Figs. 1 (a) and (b). In phase I for both compounds, the strain ε_{xy} is zero, namely the system belongs to cubic. Below I-IV phase transition temperature indicated by arrows, ε_{xy} decreases monotonously in both compounds. These results show evidently a spontaneous lattice distortion from cubic to trigonal in phase IV. The macroscopic lattice distortion with symmetry breaking in phase IV favors a ferro-

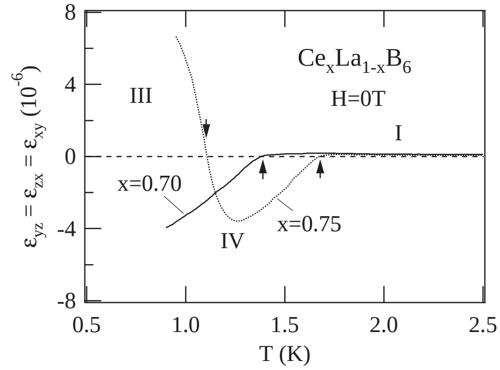

Fig. 2. The temperature dependence of $\varepsilon_{xy}(= \varepsilon_{yz} = \varepsilon_{zx})$ of $Ce_xLa_{1-x}B_6$ for $x = 0.75$ and 0.70 obtained from the results of thermal expansion experiment in Figs. 1 (a) and (b).

quadrupole (FQ) ordering. Taking into account the quadrupole-strain interaction $H_{QS} = -g_{\Gamma5}(O_{yz}\varepsilon_{yz} + O_{zx}\varepsilon_{zx} + O_{xy}\varepsilon_{xy})$, the spontaneous strain $\langle\varepsilon_{xy}\rangle \neq 0$ of the trigonal distortion is expected by the FQ ordering $\langle O_{xy}\rangle \neq 0$ as $\langle\varepsilon_{xy}\rangle = Ng_{\Gamma5}\langle O_{xy}\rangle/C^0_{44}$. N is the number of ion per unit volume, $g_{\Gamma5}$ is a coupling constant of the quadrupole-strain interaction. C^0_{44} is the elastic constant without the quadrupole-strain interaction. The magnitude of the distortion $\langle\varepsilon_{xy}\rangle \simeq 4 \times 10^{-6}$ of $Ce_xLa_{1-x}B_6$ in phase IV is considerably small as compared to that of typical FQ ordering phase, which shows distortion as large as $10^{-3} \simeq 10^{-4}$. We refer typical examples of FQ ordering in HoB_6, $Ce_3Pd_{20}Ge_6$ [8,9]. It has been proposed the possibility of antiferro-octupole ordering in phase IV. Because of the octupole does not couple to the elastic strain, the trigonal distortion in $Ce_xLa_{1-x}B_6$ ($x = 0.75$, 0.70) is free from octupole ordering. The absence of precession in μSR experiment in phase IV of $Ce_{0.70}La_{0.30}B_6$ does not contradict the present scenario of FQ ordering in phase IV [5]. In order to confirm the FQ ordering in phase IV, further experiments of the thermal expansion and ultrasonic attenuation at low temperatures are required.

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