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The concentration–temperature phase diagram of $Ce_xLa_{1-x}B_2C_2$

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Abstract
We have synthesized polycrystalline $Ce_xLa_{1-x}B_2C_2$ samples and measured the temperature dependence of the low-field magnetization, ac susceptibility and specific heat. As the Ce concentration x decreases, the Néel temperature T_N decreases linearly for $x \geqslant 0.80$. However, the dependence on the concentration x of the transition temperature changes slope at $x \sim 0.75$. Spin-glass transitions are observed for $0.4 \leqslant x \leqslant 0.75$. The origin of the spin-glass behaviour will be discussed.

1. Introduction

RB_2C_2 (R = rare earth) compounds, which have the tetragonal LaB_2C_2-type crystal structure have been attracting much interest. DyB_2C_2 is one of these compounds and it shows both an antiferroquadrupolar (AFQ) ordering and an antiferromagnetic (AFM) ordering, at $T_Q = 24.7$ K and $T_N = 15.3$ K, respectively. T_Q is almost ten times as high as those for other systems reported so far [1]. HoB_2C_2 also shows both an AFQ ordering and an AFM ordering, but the AFQ ordering transition occurs at $T_Q = 4.5$ K, below $T_N = 5.9$ K, which is unusual for quadrupolar ordering compounds [2, 3]. TbB_2C_2 shows an AFM transition at $T_N = 21.7$ K but no AFQ transition at zero field [4]. However, an AFQ ordered phase is induced by magnetic fields in TbB_2C_2.

Because of these magnetic properties of the RB_2C_2 compounds, has CeB_2C_2 attracted our attention. CeB_2C_2 is an antiferromagnet with $T_N = 7.3$ K [5]. It also shows a small anomaly at $T_t = 6.5$ K, which is presumed to indicate an order–order transition. Because the moment direction of CeB_2C_2 is in the c-plane like those of DyB_2C_2, HoB_2C_2 and TbB_2C_2, we expected that it might show stimulative magnetic behaviour as well, although no AFQ ordering has been observed in pure CeB_2C_2. We therefore tried changing the strength of the interactions in CeB_2C_2 by substituting nonmagnetic La^{3+} for Ce^{3+}.

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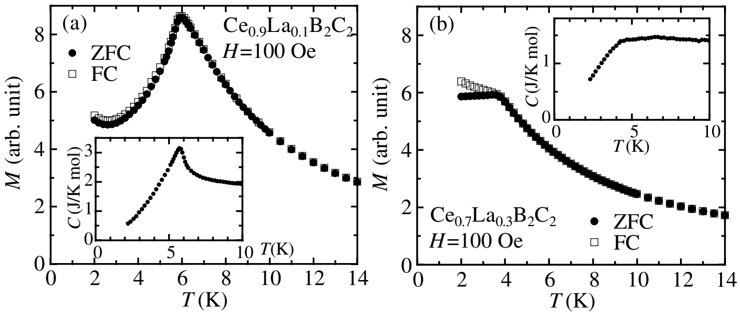

Figure 1. The temperature dependence of the magnetization of for $Ce_xLa_{1-x}B_2C_2$. (a) $x = 0.9$, (b) $x = 0.7$. The applied magnetic field is 100 Oe. The insets show the temperature dependence of the specific heat for each sample.

2. Experimental details

We synthesized the $Ce_xLa_{1-x}B_2C_2$ compounds by the conventional argon arc technique. The low-field magnetization M and ac susceptibility χ' of the polycrystalline samples were measured using a SQUID magnetometer (Quantum Design's MPMS). We measured M both under zero-field-cooled (ZFC) conditions and field-cooled (FC) conditions. The ZFC magnetization M_{ZFC} was measured on heating after the sample was cooled in zero field and then a measuring field was applied. The FC magnetization M_{FC} was measured on heating after the sample was cooled in a measuring field. We also measured the specific heat C in zero magnetic field using a Quantum Design PPMS.

3. Results

The magnetization M versus temperature T curves show a peak or a kink for the $0.4 \leqslant x \leqslant 0.9$ samples. In figure 1, we show the temperature dependence of the magnetization M of two $Ce_{1-x}La_xB_2C_2$ samples, with $x = 0.9$ and 0.7. The magnetization was measured with the magnetic field $H = 100$ Oe. The M_{ZFC}–T curve for $x = 0.9$ shows a peak at 5.9 K and that for $x = 0.7$ shows a kink at 3.7 K. Below those temperatures where the anomaly is observed, both samples show irreversibility between M_{ZFC} and M_{FC}, but otherwise the magnetic behaviour differs considerably between these samples.

The insets of figures 1(a) and (b) show the temperature dependence of the specific heat C for each sample. For $x = 0.9$, the C–T curve shows a sharp peak. The temperature dependences of M and C are similar to those for CeB_2C_2, although no anomaly corresponding to T_t is observed. The ac susceptibility measurement shows no frequency dependence for $x = 0.9$. Consequently, the peak at 5.9 K comes from the AFM transition. On the other hand, the C–T curve for $x = 0.7$ shown in the inset of figure 1(b) shows no sharp peak around 3.7 K, the temperature at which the kink appears in the M–T curve. Only a broad rounded maximum is observed in the C–T curve. In figure 2, we show the temperature dependence of the linear part of the ac susceptibility χ' for $x = 0.7$. Below the kink temperature, χ' becomes smaller with increasing frequency. These features are consistent with those of a spin-glass system. We defined the peak temperature of M_{ZFC} as the spin-glass freezing temperature T_{SG}. The spin-glass transition is also observed for $0.4 \leqslant x \leqslant 0.75$.

In figure 3, we show the dependence on the Ce concentration x of the transition temperatures T_N, T_t and T_{SG} of $Ce_xLa_{1-x}B_2C_2$. As the Ce concentration x decreases, T_N

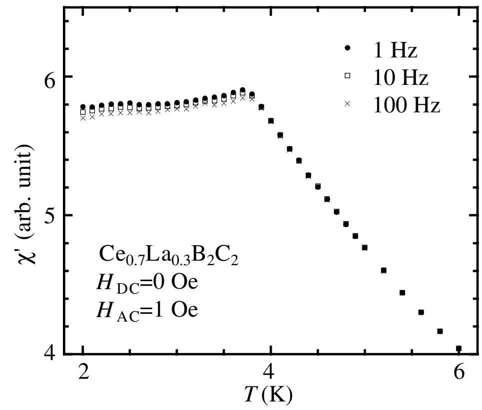

Figure 2. The temperature dependence of the ac susceptibility for the $Ce_{0.7}La_{0.3}B_2C_2$ sample at various frequencies in zero magnetic field. The applied ac field is 1 Oe.

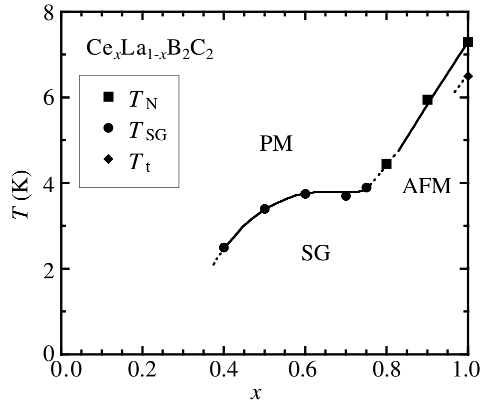

Figure 3. The magnetic phase diagram of $Ce_xLa_{1-x}B_2C_2$ in the temperature–concentration plane at $H \sim 0$. PM: paramagnetic phase; AFM: antiferromagnetic phase; SG: spin-glass phase. The solid curves are guides to the eye.

decreases linearly for $x \geqslant 0.8$. However, the concentration dependence of the transition temperature changes slope at $x \sim 0.75$. T_{SG} is almost concentration independent for $0.5 \leqslant x \leqslant 0.7$. As x decreases further, the spin-glass temperature decreases. For the $x = 0.3$ and 0.2 samples, no anomaly is observed down to 2 K.

4. Discussion

The spin-glass behaviour is also observed for the Lu-diluted $Ce_xLu_{1-x}B_2C_2$ system [7]. However, the other RB_2C_2 systems which have been studied so far, including $Dy_xY_{1-x}B_2C_2$ and $Ho_xY_{1-x}B_2C_2$, show no spin-glass behaviour [6, 8]. The magnetic structure of CeB_2C_2 in the AFM phase is a long-periodic one based on ferromagnetic coupling. The propagation vector which describes the structure is $[\delta\ \delta\ \delta']$, where $\delta = 0.16$ and $\delta' = 0.10$, while the magnetic coupling in the c-plane is basically AFM in the other RB_2C_2 compounds [9]. The particular magnetic character of CeB_2C_2 may cause latent frustrations between the AFM and ferromagnetic interaction. Thus it is possible that $Ce_xLa_{1-x}B_2C_2$ and $Ce_xLu_{1-x}B_2C_2$ show spin-glass freezing, because the Ce^{3+} moments randomly replaced by La^{3+} or Lu^{3+} are involved in the randomly distributed exchange interaction.

It is noted that similar x–T phase diagrams have been found for the heavy-fermion compounds $U_{1-x}Y_xPd_2Al_3$ and $U_{1-x}La_xPd_2Al_3$ [10], which exhibit non-Fermi-liquid (NFL) behaviour of the highly diluted samples where the spin-glass state disappears. Experiments on highly diluted $Ce_xLa_{1-x}B_2C_2$ samples are now in preparation—in order to examine the NFL behaviour, since CeB_2C_2 also shows Kondo character.

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