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Spin-glass behaviour of $Ce_{0.69}Lu_{0.31}B_2C_2$

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Abstract

We have measured the magnetization and ac susceptibility of single-crystalline $Ce_{0.69}Lu_{0.31}B_2C_2$. Some features of a spin-glass state were found in the irreversibility between the zero-field-cooled and the field-cooled magnetizations under magnetic fields between 5 and 55 mT in the temperature range from 2 to 10 K and the frequency dependence of the real and imaginary parts of the ac susceptibility (χ' and χ''). The results indicate the occurrence of a spin-glass transition in $Ce_{0.69}Lu_{0.31}B_2C_2$. We suppose the spin-glass behaviour of $Ce_{0.69}Lu_{0.31}B_2C_2$ to originate from frustration of magnetic interactions in this compound.

1. Introduction

In the RB_2C_2 (R = rare earth) system with the tetragonal LaB_2C_2-type crystal structure (space group $P4/mbm$), there are three materials showing attractive antiferroquadrupolar (AFQ) ordering: DyB_2C_2 [1], HoB_2C_2 [2] and TbB_2C_2 [3]. DyB_2C_2 is the first compound with a tetragonal symmetry found to show AFQ order. The ordering temperature of $T_Q = 24.7$ K is about ten times higher than those of the AFQ ordering materials found to date. DyB_2C_2 also shows an antiferromagnetic (AFM) transition at $T_N = 15.3$ K. The magnetic structure below T_N can be described in terms of competitive coexistence of the AFQ and AFM orders. In HoB_2C_2 the AFQ ordering transition occurs below the AFM transition temperature ($T_Q = 4.5$ K and $T_N = 5.9$ K). TbB_2C_2 is an antiferromagnet with $T_N = 21.7$ K; that is, there is no AFQ phase. However, the AFQ ordering phases are induced by applied magnetic fields [3].

CeB_2C_2 is a unique compound in the RB_2C_2 systems from the point of view of magnetic coupling. CeB_2C_2 is an antiferromagnet with $T_N = 7.3$ K accompanied with an order–order transition at $T_t = 6.5$ K [4]. The moments lie in the c-plane. From neutron diffraction experiments, the ground state has a $[\delta\ \delta\ \delta']$-type long-periodic magnetic structure, where $\delta = 0.16$, $\delta' = 0.10$ [5]. The magnetic coupling in the c-plane in CeB_2C_2 is basically ferromagnetic, though the couplings in the other RB_2C_2 compounds are AFM.

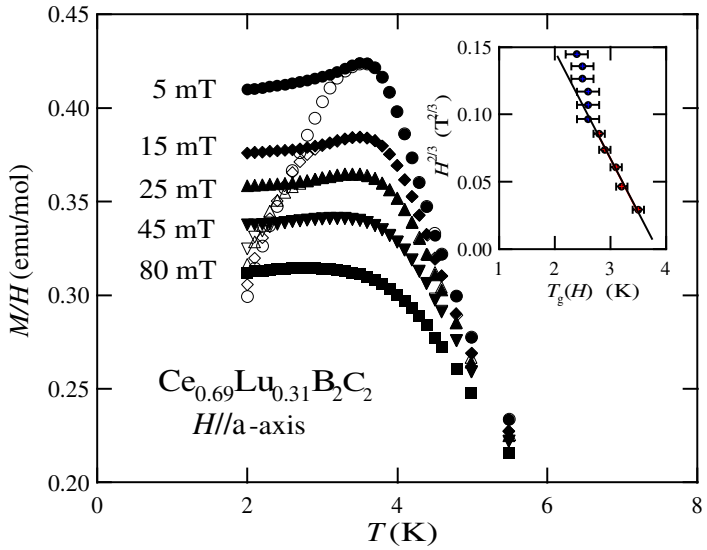

Figure 1. The temperature dependence of the magnetization M/H of $Ce_{0.69}Lu_{0.31}B_2C_2$ along the [100] direction. Open and closed symbols stand for the data obtained under ZFC and FC conditions, respectively. The inset shows the $H^{2/3}$–$T_g(H)$ plot of $Ce_{0.69}Lu_{0.31}B_2C_2$.

(This figure is in colour only in the electronic version)

Recently, substitution of Lu for Ce in CeB_2C_2 revealed that the characteristics of the magnetic transition at T_N change from first order to second order [6]. The sharp shape of the specific heat at T_N and the cusp at T_t disappear upon Lu substitution. There is only a slightly broadened peak at around 7 K. Furthermore, the susceptibility of $Ce_{0.69}Lu_{0.31}B_2C_2$ shows a cusp at 3.7 K, while the specific heat shows no anomaly at this temperature.

In order to study more precisely the magnetic properties of $Ce_{0.69}Lu_{0.31}B_2C_2$, we have carried out magnetization and ac magnetic susceptibility measurements on a single-crystalline $Ce_{0.69}Lu_{0.31}B_2C_2$ sample.

2. Experimental details

The starting materials were Ce of 99.9% purity, B of 99.7% purity and C of 99.999% purity. The polycrystalline ingot was prepared by arc melting in an argon atmosphere. The single crystal was grown by the Czochralski method using a tri-arc furnace. No second phase was recognized in crushed fractions of the single-crystalline samples by x-ray diffraction examinations. The magnetization and ac susceptibility were measured using a SQUID magnetometer in the temperature range from 2 to 10 K. The ac frequencies were 1, 10 and 100 Hz.

3. Results and discussion

Figure 1 shows the temperature dependence of the magnetization M/H of $Ce_{0.69}Lu_{0.31}B_2C_2$ along the [100] direction in the temperature range from 2 to 6 K. The measurements were made under both zero-field-cooled (ZFC) and field-cooled (FC) conditions. There is a significant difference between the ZFC and FC magnetizations in the fields below 55 mT. The irreversibilities between the ZFC and FC magnetizations manifest an aspect of spin-glass behaviour.

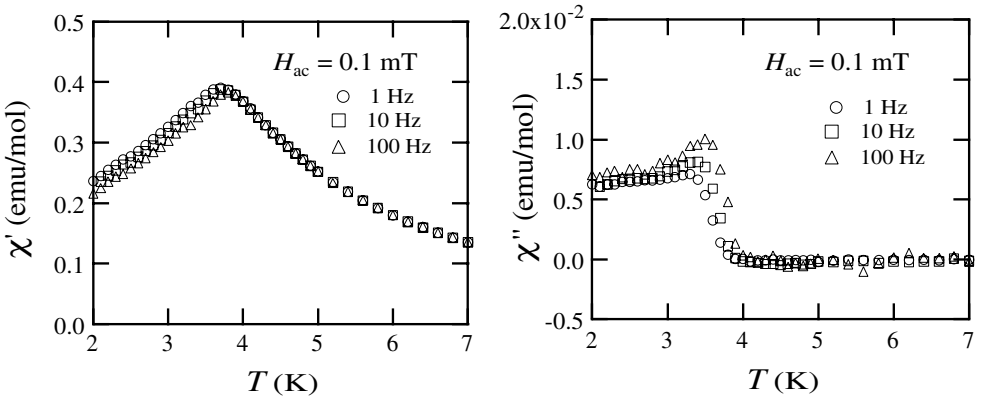

Figure 2. The frequency dependences of the real and imaginary parts of the ac susceptibility (χ' and χ'') along the [100] direction in the temperature range from 2 to 7 K.

The inset of figure 1 shows the $H^{2/3}$–$T_g(H)$ plot for $Ce_{0.69}Lu_{0.31}B_2C_2$ along the [100] direction. The critical temperatures $T_g(H)$ are determined from the onset temperature of irreversible magnetization, which is the difference between the FC and ZFC magnetizations. Linearity of $H^{2/3}$–$T_g(H)$ is exhibited for the data above 2.5 K (below 35 mT), although the data above 35 mT deviate from this line. A linear $H^{2/3}$–$T_g(H)$ relation has been observed in many spin-glass systems under weak magnetic fields [8, 10] and has been discussed as the critical H–T line which has been given by de Almeida–Thouless (AT) for the Ising model [7]. The AT line has the form

$$H = A\left(1 - \frac{T_g(H)}{T_g(0)}\right)^{3/2},$$

where $T_g(0)$ is the spin-glass temperature and A a coefficient depending on the material. From the magnetic measurements, however, $Ce_{0.69}Lu_{0.31}B_2C_2$ may be classified as an XY-type spin glass. According to the mean-field theory of Kotliar and Sompolinsky [9], even an XY-type spin glass possibly shows an AT line. The value of T_g (0) is estimated to be 3.8 ± 0.1 K. We also evaluated the experimental value of the coefficient A as 0.171 ± 0.003 T, which is about a tenth of the theoretical value [9]. This tendency of A has often been obtained in previous studies of other spin-glass systems [10].

Figure 2 shows the frequency dependences of the real and imaginary parts of the ac susceptibility (χ' and χ'') along the [100] direction in the temperature range from 2 to 7 K. The χ'–T curve exhibits a sharp peak at 3.7 K under 1 Hz and its peak position shifts to higher temperatures with increasing frequency. Inflection points in the χ''–T curves correspond to the peak temperature of χ'. The values of χ'' are nearly constant in the lower-temperature region. These behaviours of the ac susceptibility are also characteristic of spin glass. An anomaly around 2.5 K is observed in each χ'–T curve, although no anomaly appears in the χ''–T curves. This anomaly seems not to be related to spin-glass behaviour, although its origin remains unresolved. The present study reveals that $Ce_{0.69}Lu_{0.31}B_2C_2$ undergoes a spin-glass transition. We suppose that the spin-glass behaviour of $Ce_{0.69}Lu_{0.31}B_2C_2$ originates in the frustration of magnetic interactions. Precise measurements on the anomaly around 2.5 K must be performed in the future.

4. Summary

We performed dc magnetization and ac susceptibility measurements on a single-crystalline $Ce_{0.69}Lu_{0.31}B_2C_2$ compound. Clear irreversibility appears in the FC and ZFC magnetizations. The peak that appears in the ac susceptibilities depends on the ac frequency. The results indicate that a spin-glass transition occurs in $Ce_{0.69}Lu_{0.31}B_2C_2$. Measurements on compounds with high Lu concentrations are now in preparation.

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