Magnetic Behavior in Nonmagnetic Atom Disorder System Ce_2CuSi_3

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Abstract

Results of magnetic susceptibility, specific heat and electrical resistivity measurements on a polyceystalline Ce_2CuSi_3 sample are reported. This compound shows the spin-glass behavior below T_f=2.7 K with extended short-range magnetic order. The spin glass state is considered to originate from the randomly distributed RKKY-type exchange interactions due to the disordering of Cu and Si atoms on the crystallographic sites.

Key words: Ce_2CuSi_3; susceptibility; specific heat; spin glass

Nonmagnetic atom disorder compounds R_2CuSi_3 (R= rare earth element), which crystallize in a hexagonal AlB_2-type structure with random distribution of Cu and Si atoms on the B positions in AlB_2, is a very interesting family among the ternary intermetallic compounds with composition 2:1:3. Among them complex magnetic orderings exhibit in Nd_2CuSi_3 [1] and Pr_2CuSi_3 [2] with antiferromagnetic (AF) transition at T_N=5 K and ferromagnetic (F) transition at T_C=10 K, respectively. At much lower temperatures, both them show spin glass (SG) behavior. For Ce_2CuSi_3, there is a peak in the low field dc susceptibility curve near 2.1 K and the temperature dependence of specific heat measured down to 4.5 K shows a mass enhancement behavior [3]. These unusual features motivated us to investigate R_2CuSi_3compounds systematically. In this paper, we present the results of magnetic susceptibility, specific heat and electrical resistivity measurements on a well-annealed polycrystalline Ce_2CuSi_3 sample.

The sample was prepared in an arc furnace and annealed at 950 $^\circ$C for 20 days. X-ray measurement showed only a single phase with AlB_2-type structure (space group *P6/mmm*). The lattice constants are a=4.058(2) Å and c=4.288(2) Å. The ac susceptibility χ_{ac} and dc magnetization $M(T)$ were measured between 1.8 and 300 K using a SQUID magnetometer. Adiabatic heat pulse method and four-terminal dc method were used for specific heat $C(T)$ and electrical resistivity $\rho(T)$ measurements, respectively.

Figure 1 shows the temperature dependence of the real component χ_{ac}' of the ac susceptibility of Ce_2CuSi_3 between 2 and 4 K at the frequency range $0.1 \leq \nu \leq 1000$ Hz. Typical SG behaviors are observed: χ_{ac}' exhibits a pronounced maximum around $T_f(v)$, and an upward-shift of $T_f(v)$ with increasing v is clearly observed. The initial frequency shift δT_f was calculated as $\delta T_f = \Delta T_f/(T_f \Delta \log\nu) = 0.013$. This value is comparable to those reported for typical metallic SG systems, for example, 0.010 for AuFe [4], 0.025 for URh_2Ge_2[5] and 0.008-0.022 for some other 2:1:3 SG systems [6-9]. The formation of SG states in Ce_2CuSi_3 is also confirmed by dc susceptibility χ ($=M/H$) measurements performed in the zero-field cooling (ZFC) mode and in the field-cooling (FC) mode. As shown in the inset of Fig. 1, $\chi_{ZFC}(T)$ measured in a field of 100 Oe is irreversible and shows a peak at 2.4 K. In contrast, $\chi_{FC}(T)$ is reversible and increases monotonously down to 1.8 K resulting in the

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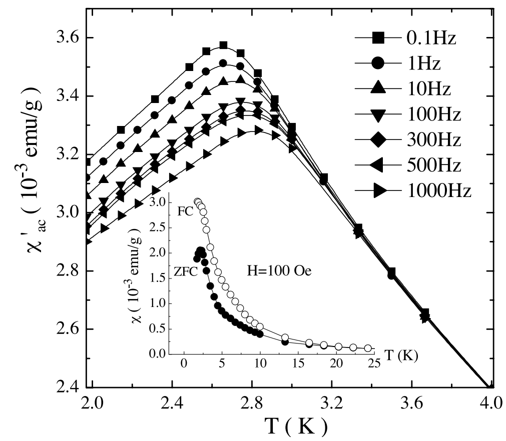

Fig. 1. Real component χ_{ac}' of the ac susceptibility of Ce_2CuSi_3 versus temperature between 2 and 4 K at various frequencies ranging from 0.1 to 1000 Hz. The inset shows the difference between FC and ZFC dc susceptibility.

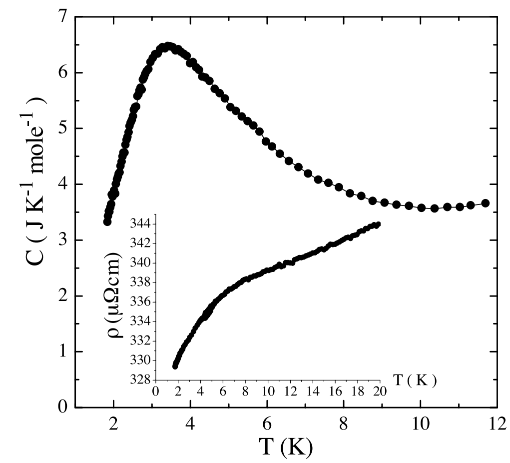

Fig. 2. Temperature dependence of specific heat of Ce_2CuSi_3 between 1.7 and 12 K. The inset displays the electrical resistivity data of Ce_2CuSi_3 at low temperatures.

evident bifurcation between the FC and ZFC curves at low temperatures. This irreversibility is also a usual characteristic for a spin glass. In this work, we define the spin freezing temperature T_f (=2.7 K at v=0.1 Hz) of Ce_2CuSi_3 as the peak temperature in χ_{ac}'(T) curve. It is interesting to note that different from canonical SG system, the deviation between χ_{ZFC} and χ_{FC} of Ce_2CuSi_3 starts at T_m (=17 K) much higher than T_f. It may be due to the formation of magnetic cluster causing a superparamagnetic contribution.

The specific heat and electric resistivity measurements give further evidences for above consequence. As illustrated in Fig. 2, $C(T)$ of Ce_2CuSi_3 shows a slow rise starting at T ≈10 K followed by a broad maximum around T=3.4 K (> T_f=2.7 K). The data do not show visible indication of a *long-range* magnetic phase transition. This particular behavior of $C(T)$ is a characteristic feature of a SG system with existence of magnetic clusters. It is clear from the inset of Fig. 2 that no anomaly (peak or sudden bend) is observed in $\rho(T)$ curve at T_f. This result suggests the absence of *long-range* spatial magnetic order near T_f in agreement with the $C(T)$ measurements. Between 1.5 and 10 K $\rho(T)$ curve manifests a broad bent with negative curvature. The formation of magnetic clusters may be responsible for this feature. The $C(T)$ and $\rho(T)$ behavior described above are different from those observed for general long-range magnetic order material such as isostructural Nd_2PtSi_3 that shows a rapid increase in $C(T)$ curve and a sudden bend in $\rho(T)$ curve at the phase transition temperature T_C [10].

In conclusion, we have observed the SG behavior with extended short-range magnetic order for Ce_2CuSi_3 from magnetic, transport and thermal properties measurements. The origin of SG behavior for Ce_2CuSi_3 is different from that in diluted metallic SG or uranium intermetallic compound. The frustrated magnetic moments may be originated from the competing between F and AF interactions in Ce-atom layer in which nearest neighbour Ce atoms form triangular structure. On the other hand, the statistical distribution of Cu and Si atoms could introduce the randomly distributed RKKY-type magnetic exchange interactions [8]. Thus satisfying the necessary conditions, frustration and randomness [4], for formation of spin-glass state is possible in Ce_2CuSi_3.

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