Magnetic Behavior in Nonmagnetic Atom Disorder System

\[ \text{Ce}_2\text{CuSi}_3 \]

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Abstract

Results of magnetic susceptibility, specific heat and electrical resistivity measurements on a polycrystalline \[ \text{Ce}_2\text{CuSi}_3 \] sample are reported. This compound shows the spin-glass behavior below 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: \[ \text{Ce}_2\text{CuSi}_3 \]; susceptibility; specific heat; spin glass

Nonmagnetic atom disorder compounds \[ R_2\text{CuSi}_3 \] (\( R \) = rare earth element), which crystallize in a hexagonal AlB\textsubscript{2}-type structure with random distribution of Cu and Si atoms on the B positions in AlB\textsubscript{2}, is a very interesting family among the ternary intermetallic compounds with composition 2:1:3. Among them complex magnetic orderings exhibit in \[ \text{Nd}_2\text{CuSi}_3 \] \cite{1} and \[ \text{Pr}_2\text{CuSi}_3 \] \cite{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 \[ \text{Ce}_2\text{CuSi}_3 \], 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 \cite{3}. These unusual features motivated us to investigate \[ R_2\text{CuSi}_3 \] compounds systematically. In this paper, we present the results of magnetic susceptibility, specific heat and electrical resistivity measurements on a well-annealed polycrystalline \[ \text{Ce}_2\text{CuSi}_3 \] sample.

The sample was prepared in an arc furnace and annealed at 950 °C for 20 days. X-ray measurement showed only a single phase with AlB\textsubscript{2}-type structure (space group \textit{P6/mmm}). The lattice constants are \( a = 4.058(2) \) Å and \( c = 4.288(2) \) Å. The ac susceptibility \( \chi_{ac} \) 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 \( \chi_{ac}' \) of the ac susceptibility of \[ \text{Ce}_2\text{CuSi}_3 \] between 2 and 4 K at the frequency range \( 0.1 \leq \nu \leq 1000 \) Hz. Typical SG behaviors are observed: \( \chi_{ac}' \) exhibits a pronounced maximum around \( T_f(\nu) \), and an upward-shift of \( T_f(\nu) \) with increasing \( \nu \) is clearly observed. The initial frequency shift \( \delta T_f \) 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 \textit{AuFe} \cite{4}, 0.025 for \textit{URh}_2\textit{Ge}_2 \cite{5} and 0.008-0.022 for some other 2:1:3 SG systems \cite{6-9}. The formation of SG states in \[ \text{Ce}_2\text{CuSi}_3 \] is also confirmed by dc susceptibility \( \chi = \Delta M/H \) 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...
Fig. 1. Real component $\chi_{ac}'$ of the ac susceptibility of Ce$_2$CuSi$_3$ 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$_2$CuSi$_3$ between 1.7 and 12 K. The inset displays the electrical resistivity data of Ce$_2$CuSi$_3$ 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$_2$CuSi$_3$ as the peak temperature in $\chi_{ac}'(T)$ curve. It is interesting to note that different from canonical SG system, the deviation between $\chi_{ZFC}$ and $\chi_{FC}$ of Ce$_2$CuSi$_3$ 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$_2$CuSi$_3$ shows a slow rise starting at $T \approx 10$ 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 bend 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$_2$PtSi$_3$ 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$_2$CuSi$_3$ from magnetic, transport and thermal properties measurements. The origin of SG behavior for Ce$_2$CuSi$_3$ 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$_2$CuSi$_3$.

References