Spin-glass Behavior in Ternary Uranium Compound U_2AuGa_3

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Abstract

The magnetic properties of the intermetallic compound U_2AuGa_3 were investigated on well-annealed polycrystalline samples by DC magnetization, specific heat and magnetic relaxation measurements. The temperature dependence of magnetization shows irreversibility and the frozen-in magnetic moment was observed below a characteristic temperature T_f=23 K. The temperature dependence of specific heat does not show visible singularity indicating the absence of long-range magnetic order around T_f. Furthermore, more than 50% of the remnant magnetization is observed even after 3 hours due to long-time magnetic relaxation effect. These results clearly indicate that U_2AuGa_3 undergoes spin-glass transition at T_f.

Key words: U_2AuGa_3; spin-glass; magnetic susceptibility; specific heat

Uranium ternary compounds U_2XSi_3 with X=Pd, Pt and Au have been reported to show the spin-glass behavior at low temperatures [1-3]. The formation of the spin-glass state in these compounds is attributed to the randomly frustrated U-U interaction owing to the intrinsic atomic disorder of AlB_2-type lattice. Recently, several ternary compounds U_2XGa_3 with $CeCu_2$-type structure have been studied. The compounds with X=Ru, Rh and Ir were found to show ferromagnetic orders [4], whereas the compounds with X=Pd and Pt are antiferromagnets with a ferromagnetic transition at T_m=80 K for the latter compound [5]. The magnetic properties of the U_2XGa_3 are not well understood so far, especially for X =Au. In this study the formation of spin glass state in U_2AuGa_3 is found on the basis of the magnetization and the specific heat measurements.

A polycrystalline sample of U_2AuGa_3 was synthesized by melting the stoichiometric amounts of the constituent elements using an arc furnace. The purities of the starting materials are 3N for U, 4N for Au and 6N for Ga. Weight loss in the melting process is smaller than 0.2 wt%. The sample was then annealed at 800°C for a week. Powder X-ray diffraction analysis of the U_2AuGa_3 sample shows a single phase of the orthorhombic $CeCu_2$-type structure with lattice parameters of a= 4.435 Å, b= 7.079 Å and c= 7.799 Å.

Figure 1 shows the temperature dependence of the ratio of the magnetization to the magnetic field ($\chi = M/H$, hereafter called as the susceptibility) of U_2AuGa_3. The susceptibility curve varies remarkably depending on the zero-field cooling (ZFC) and the field cooling (FC) conditions as shown in Fig.1. In an applied field of 0.3 T an apparent maximum appears at T_f= 23 K and irreversibility starts just above the temperature, *i.e.* 26 K. The difference between FC and ZFC curves is much significant below T_f. As shown in the inset of Fig.1, with increasing the magnetic field, the peak in $\chi_{ZFC}(T)$ broadens and the peak temperature decreases. The broad maximum under H= 0.5 T is observed around 22.0 K. These features are characteristic to spin glass systems with metastable states below the freezing temperature T_f.

The reciprocal susceptibility in the range 110-300 K (not shown here) is well described by the modified Curie-Weiss law $\chi = \chi_0 + \frac{N_A \mu_{eff}^2}{3k_B(T-\Theta_p)}$, where χ_0 denotes the temperature independent term, Θ_p the para-

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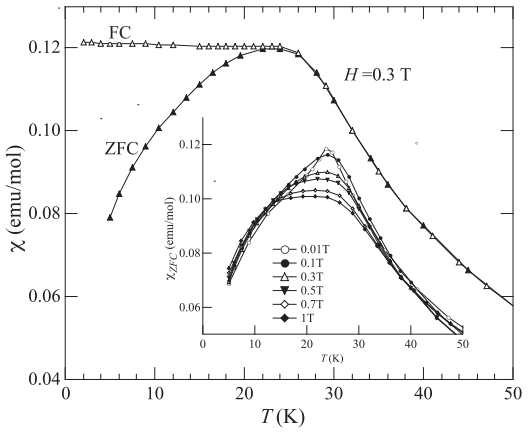

Fig. 1. Temperature dependence of susceptibility for U_2AuGa_3 after zero-field cooling (ZFC) and field cooling (FC) in a magnetic field of 0.3 T and susceptibility curves in ZFC condition under various DC magnetic fields (inset).

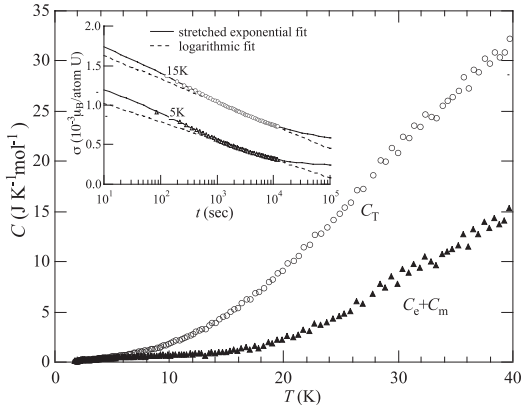

Fig. 2. Temperature dependence of total specific heat (C_T) of U_2AuGa_3 displayed with C_e+C_m curve obtained by subtracting the C_p from the C_T, where C_p is calculated using the Debye temperature Θ_D= 116.2 K. Relaxation of the remnant magnetization M_{IRM} at 5 K and 15 K (inset), where the solid lines represent the fitting based on the least-squared method.

magnetic Curie temperature, and μ_{eff} the effective magnetic moment. The fitting by the nonlinear least-squares method yields $\chi_0 = 1.03 \times 10^{-3}$ μ_B/atom U, Θ_p= 27.2 K, and μ_{eff}= 2.61 μ_B/atom U. The small kink in M/H curve observed at T_c= 110 K in a field of 0.01 T and the positive value of Θ_p suggest a weak ferromagnetic transition at T_c.

Figure 2 shows C_e+C_m curve obtained by subtracting the C_p from the C_T, where C_p, C_e and C_m are the phonon, electronic and magnetic contributions to the total specific heat C_T, respectively. C_p is calculated using the Debye temperature Θ_D= 116.2 K estimated from the low temperature data. The C_e+C_m curve indicates no singularity around T= 23 K where the magnetic susceptibility shows a sharp cusp, which excludes the existence of long-range ferromagnetic or antiferromagnetic phase transition. Although it has been pointed out that magnetic specific heat of spin glasses usually shows a broad peak at the temperature T_m 20-40% higher than T_f [6], such a broad peak is not seen in the C_e+C_m curve for U_2AuGa_3. The γ value is evaluated as 85.1 mJ mol U^{-1} K^{-2} in a plot of $C(T)/T = \gamma + \beta T^2$ at $T << \Theta_D$.

The remnant magnetization of U_2AuGa_3, measured by the method reported previously [2], decays slowly over more than three hours (the inset of Fig. 2). The M_{IRM} curves are satisfactorily fitted by the stretched exponential function $M_{IRM}(T,t) = M_0(T) + \alpha(T)exp[-(\frac{t}{\tau(T)})^{\beta(T)}]$ [7], where T denotes the temperature, t the elaplsed time, τ the relaxation time constant, M_0 and α the fitting parameters, β the positive index number less than the unity. The best fitting results are shown by the solid lines in the inset of Fig. 2 with $M_0 = 3.08 \times 10^{-4}$ and $7.21 \times 10^{-4} \mu_B$/atom U, $\tau = 9.18 \times 10^3$ and 1.43×10^4 s, $\alpha = 7.54 \times 10^{-4}$ and 7.61×10^{-4} μ_B/atom U, β=0.526 and 0.575 for 5 K and 15 K, respectively. This "slow dynamic" relaxation, which is not a simple exponential decay, is indicative of the "multi-valley" picture of spin glasses.

Conclusively, our magnetic and specific heat measurements reveal the spin-glass state in U_2AuGa_3 compound with orthorhombic $CeCu_2$-structure, where U atom occupies the Ce site and Au and Ga atoms randomly occupy the Cu site. The disordering of the Au and Ga atoms is expected to introduce some frustrations to U-U magnetic interactions mediated by $5f$(U)-$nd(X)$ hybridization as proposed for some spin glass compounds U_2XSi_3, where X and Si atoms are randomly distributed to boron site of hexagonal AlB_2-type structure.

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