Ferromagnetic cluster glass behavior in U_2IrSi_3

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Measurements of the ac susceptibility, dc magnetization, magnetic relaxation, specific heat, and electrical resistivity of U_2IrSi_3 are reported. The investigated compound crystallizes in an AlB_2-derived hexagonal structure and undergoes a ferromagnetic cluster glass transition at $T_f = 10.5$ K. Typical features of the ferromagnetic cluster glass state include (i) a frequency-dependent cusp in the ac susceptibility, (ii) an irreversibility in the temperature dependence of the dc magnetization, (iii) a very slow decay of the remanence, (iv) a small jump in the initial magnetization curve, and (v) a small specific heat anomaly, which contains a very small amount of magnetic entropy. The specific heat data also demonstrate a large value of the linear specific heat coefficient and the absence of true long-range magnetic order. Furthermore, a Kondo-effect-like behavior is observed in the temperature dependence of the electrical resistivity. It seems that both cluster glass freezing and Kondo-lattice effects make an impact on the low-temperature physical properties of U_2IrSi_3.

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Nonmagnetic atom disorder (NMAD) uranium compounds U_2XSi_3 (X = transition metal) form a very interesting family among the ternary intermetallic compounds with composition 2:1:3. Most of them crystallize in a disordered derivative of the hexagonal AlB_2-type structure, where X and Si atoms are randomly distributed into the trigonal prisms of a primitive hexagonal array of uranium atoms and U atoms within one layer form triangles of nearest neighbors.[1] Such a topologic disorder structure could induce the frustration of magnetic interactions in the case of antiferromagnetic (AF) coupling between nearest neighbors in one U layer, which have indeed been observed in the past few years and caused some controversies in physical explanation. Magnetic properties of U_2PtSi_3 were first measured by Geibel *et al.*[2] and later measured by Sato *et al.*[3] They classify this compound as either a spin glass (SG) or a weak itinerant ferromagnet. U_2XSi_3 with X = Fe, Ru, and Os display a spin fluctuation behavior at low temperatures, whereas for X = Co, Ni, Rh, Pd, Pt, and Au they order magnetically.[1] On the other hand, ac susceptibility data[4] also suggest the existence of a SG phase in compounds with X = Co, Ni, and Cu. Recently, our basic physical property measurements have given new evidence for the formation of SG state in U_2XSi_3 (X = Pd,Pt,Au,Rh).[5–7] As for the magnetic properties of U_2IrSi_3, to the best of our knowledge, only the field-cooled (FC) dc magnetization (in a field of 1 kOe down to 4.2 K), the ac susceptibility (may be at a frequency of 125 Hz), and the magnetic field dependence of magnetization (at 4.2 K up to 20 kOe) have been reported.[1] It is well known that the magnetic properties of a metastable system are sensitive to an applied magnetic field and frequency. In order to gain a completely physical picture of U_2IrSi_3 and as a continuation of our studies of the U_2XSi_3 family, we have systematically measured the basic physical properties of U_2IrSi_3 including the temperature dependence of ac and dc susceptibilities at different frequencies and in various magnetic fields, the magnetization and magnetic relaxation at different temperature, and the electrical resistivity and the specific heat on a well-annealed polycrystalline sample prepared by using an arc furnace under purified argon atmosphere.[5] The ac and dc susceptibilities, low-field magnetization, and magnetic relaxation were measured using a Quantum Design superconducting quantum interference device (SQUID) magnetometer. High-field magnetization in a steady magnetic field up to 230 kOe was measured at 4.2 K with an induction method. The adiabatic heat pulse method and a standard four-terminal dc method were employed for specific heat and electrical resistivity measurements, respectively.

Figure 1 displays the in-phase component $\chi'_{ac}(T,\omega)$ and the out-of-phase component $\chi''_{ac}(T,\omega)$ of the ac susceptibility versus temperature for U_2IrSi_3 between 7 and 15 K in the frequency range $0.1 \leqslant \omega/2\pi \leqslant 1000$ Hz. Both χ'_{ac} and χ''_{ac}

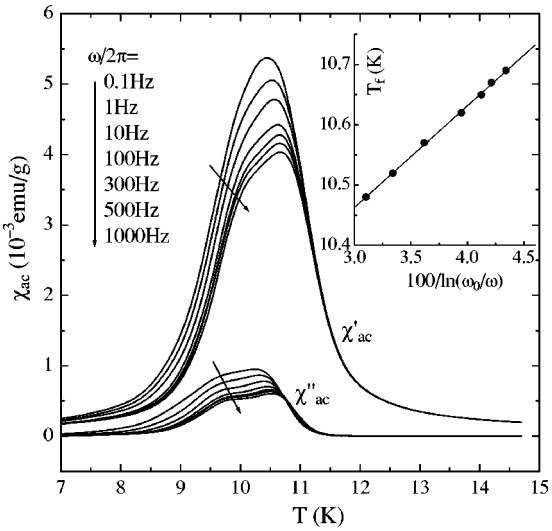

FIG. 1. Real (χ'_{ac}) and imaginary (χ''_{ac}) components of the ac susceptibility of U_2IrSi_3 vs temperature between 7 and 15 K at frequencies $0.1 \leqslant \omega/2\pi \leqslant 1000$ Hz in an applied ac field of 1 Oe. The inset shows the plot of T_f vs $100/\ln(\omega_0/\omega)$ with $\omega_0/2\pi = 10^{13}$ Hz.

curves show evident maxima with amplitudes and positions depending on the frequency ω of the applied ac magnetic field. The peak position shifts to high temperature and the peak amplitude decreases with increasing ω. These features are reminiscent of the frustrated magnetic state of a SG material. Since the magnetization curve shows ferromagnetic (FM) cluster behavior and no long-range magnetic phase transition can be detected in the specific heat measurement (see the following), the maxima in $\chi'_{ac}(T,\omega)$ and $\chi''_{ac}(T,\omega)$ are considered to signal a FM cluster glass transition. The spin freezing temperature defined as the maximum in $\chi'_{ac}(\omega)$ is $T_f = 10.5$ K at $\omega/2\pi = 0.1$ Hz. For the $\chi''_{ac}(T)$ curve and high-frequency $\chi'_{ac}(T)$ curve, a small shoulder appears at a temperature slightly lower than the peak temperature. It seems that an inhomogeneous magnetic structure exists in the sample to a certain extent. In spin glasses, a criterion—namely, $\delta T_f = \Delta T_f/(T_f \Delta \ln\omega)$—has often been used for comparing the ω dependence of T_f in different systems. We find $\delta T_f = 0.005$ for U_2IrSi_3. This value is comparable to those reported for some canonical spin glasses [e.g., CuMn: 0.005, AuMn: 0.0045 (Ref. 8)] and an extended short-range FM ordering system U_2RhSi_3 with SG behavior [$\delta T_f = 0.008$ (Ref. 6)], but is evidently smaller than those of concentrated NMAD "simple" SG systems [e.g., URh_2Ge_2: 0.025 (Ref. 9), Ce_2AgIn_3: 0.022 (Ref. 10), U_2PdSi_3: 0.020 (Ref. 7)]. As shown by a solid line in the inset of Fig. 1, our experimental data for U_2IrSi_3 could be fitted well using the empirical Vogel-Fulcher law[8] $\omega = \omega_0 \exp[-E_a/k_B(T_f - T_0)]$, with three fitting parameters: characteristic frequency ω_0, activation energy E_a (k_B is the Boltzmann constant), and Vogel-Fulcher temperature T_0, which is a measure of intercluster interaction strength. When $\omega_0/2\pi$ is 10^{13} Hz typically taken in the SG systems,[7,11] the best fit of this equation to the experimental data yields the values of fitting parameters $E_a/k_B = 16.8$ K and $T_0 = 10$ K. Clearly, the activation energy in U_2IrSi_3 determined from the peak temperature T_f in $\chi'(T)$ and $\omega_0/2\pi = 10^{13}$ Hz is $E_a \approx 1.6 k_B T_f$. Taken together, these data provide unambiguous evidence for the formation of a FM cluster glass state in U_2IrSi_3 below T_f.

The temperature dependence of the dc magnetization $M(T)$ of U_2IrSi_3 is measured in the FC mode and in the zero-field-cooled (ZFC) mode in various applied fields. A part of the results is shown in Fig. 2. For convenience, hereafter we call M/H the dc susceptibility and note it as χ. The cluster glass behavior in U_2IrSi_3 can also be observed by a peak in the $\chi_{ZFC}(T)$ curve at a strongly field-dependent temperature $T_m(H)$, as well as the appearance of irreversibility behaving as the difference between χ_{ZFC} and χ_{FC} below a characteristic temperature T_{ir}. In a low field, $\chi_{ZFC}(T)$ shows a sharp peak and T_{ir} closes to T_f, the peak temperature in the low-frequency χ'_{ac} curve. With increasing H, this peak becomes broader and its height decreases, while $T_{ir}(H)$ shifts toward lower temperatures. The inset of Fig. 2 shows the de Almeida–Thouless (AT) line for U_2IrSi_3. $T_{ir}(H)$ follows an $H^{2/3}$ law, which has also been observed in SG systems of UCuSi,[12] U_2PdSi_3,[5] and Nd_2AgIn_3,[13] and has also been predicted by the mean-field SG model.[14] It is unlike

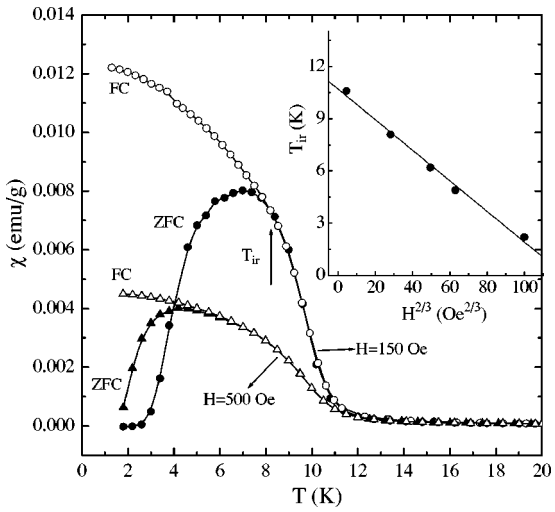

FIG. 2. The temperature dependence of dc susceptibility χ ($=M/H$) for U_2IrSi_3 measured in the FC (●) mode and in the ZFC (○) mode in magnetic fields of 150 and 500 Oe. The inset shows the de Almeida–Thouless line, plotted as T_{ir} vs $H^{2/3}$.

what happens in a simple SG system: in a low field both $\chi_{FC}(T)$ and $\chi_{ZFC}(T)$ show a much more rapid increase near $T_f = 10.5$ K, and the value of χ_{ZFC} at T_m (and χ'_{ac} at T_f) is about one order of amplitude larger than that of simple spin glasses U_2XSi_3 [X = Pd, Pt, and Au (Refs. 5 and 7)], signaling the formation of FM clusters.

The formation of a cluster glass state in U_2IrSi_3 is further confirmed by a specific heat measurement. Figure 3 shows the temperature dependence of the specific heat, $C(T)$, of this sample between 1.7 and 30 K. Different from a long-range magnetically ordered system, only a small anomaly is observed in the $C(T)$ curve near the freezing temperature $T_f = 10.5$ K. Such a specific heat anomaly contains a very small amount of magnetic entropy ($S_{mag}/R \ll \ln 2$ per U atom), which seems to be much too small for the development of the usual long-range FM order. This finding excludes

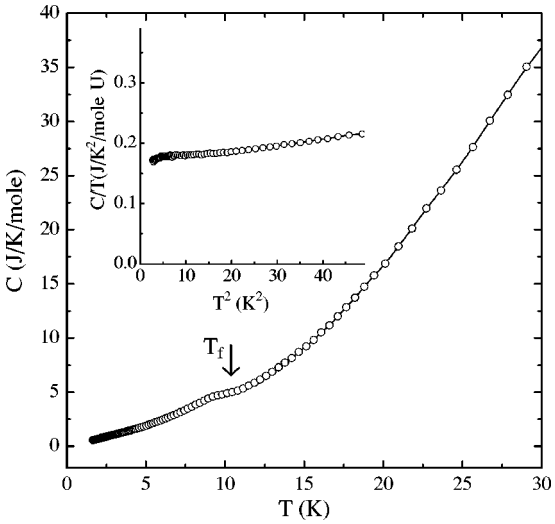

FIG. 3. Temperature dependence of the specific heat $C(T)$ of U_2IrSi_3. The inset shows the plot of C/T vs T^2.

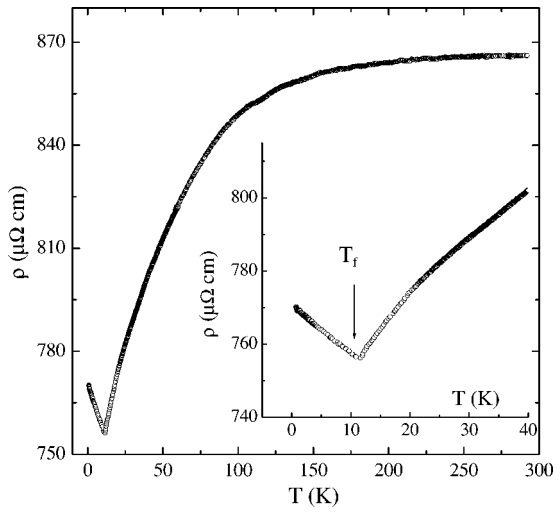

FIG. 4. Temperature dependence of the electrical resistivity of U_2IrSi_3. The inset shows the low-temperature part in an expanded scale.

the existence of long-range spatial magnetic ordering at T_f for U_2IrSi_3, and the small anomaly in $C(T)$ can originate only from the formation of the magnetic cluster state. Moreover, at low temperatures the C/T vs T^2 plot shown in the inset of this figure yields for $T \rightarrow 0$ K a large γ value (the specific heat coefficient of the T-linear term) of 170 mJ $(\text{mol U})^{-1}$ K^{-2}, Which can be considered to originate from both cluster glass freezing[8] and the formation of a moderate heavy-fermion state.[9] A coexistence of SG freezing and heavy-fermion behavior has been reported for amorphous $(U_{0.25}Pt_{0.75})_{1-x}Si_x$ alloys[15] and for the NMAD compound URh_2Ge_2.[9]

The temperature dependence of the electrical resistivity $\rho(T)$ between 0.5 and 290 K is shown in Fig. 4 for U_2IrSi_3. Three aspects of the data are noted. First, there are a large value of the residual resistivity $RR = \rho(T = 0.5\ \text{K}) = 770\ \mu\Omega\,\text{cm}$ and a small residual resistivity ratio $RRR = \rho(T = 290\ \text{K})/\rho(T = 0.5\ \text{K}) = 1.1$, which can be attributed to scattering at structural disorder as is usually observed in metallic spin glasses. Second, the $\rho(T)$ curve shows a flat slope at high temperatures, and a broad bend appears between 80 and 130 K. It is imaginable that a broad peak in $\rho_m(T)$, the *magnetic part* of the resistivity, should exist in this temperature range. This is a common feature of uranium compounds and usually attributed to the transition from a single Kondo-ion-like scattering at high temperature to a coherent Kondo state at low temperature. The pronounced decrease of $\rho(T)$ with temperature below about 80 K is a hallmark for the Kondo-lattice effect. And third, a deep minimum in $\rho(T)$ appears at low temperature. A minimum in $\rho(T)$ is the hallmark of the diluted Kondo effect and is very common in all Kondo systems showing some kind of disorder. In fact, we have observed a similar phenomenon of $\rho(T)$ minimum for most members of the U_2XSi_3 family. However, the minimum observed in U_2IrSi_3 is extremely sharp, as expected for a transition, in contrast to the quite broad minima observed in the other U_2XSi_3 systems, suggesting that this minimum has a different origin. Moreover, the observed

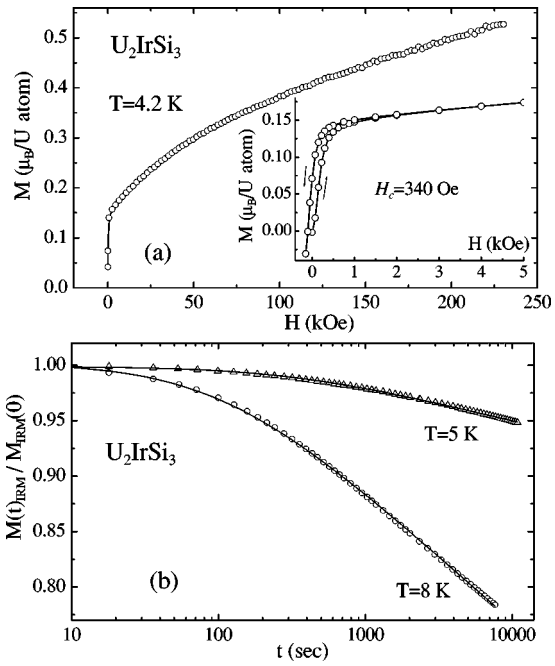

FIG. 5. (a) High-field magnetization $M(H)$ up to 230 kOe for U_2IrSi_3 measured at 4.2 K. The inset displays the hysteresis loop measured by using a SQUID magnetometer below 5 kOe. (b) Decay of isothermal remanent magnetization $M_{IRM}(t)$ as a function of time t at 5 and 8 K for U_2IrSi_3, plotted as $M_{IRM}(t)/M_{IRM}(0)$ vs t in a semilogarithmic scale. The sample was first cooled in zero field from 80 K to the desired temperature; then a magnetic field of 30 Oe was applied for 5 min and switched off at $t = 0$. The solid lines represent least-squares fits using the expression $M_{IRM}(t) = M_0(T) - S(T)\ln(t + t_0)$.

minimum temperature $T_{\min}$ (= 11.4 K) is very close to the cluster freezing temperature T_f. This is also different from that observed in U_2PdSi_3 [$T_{\min} \sim 2T_f$ (Ref. 5)] and in U_2RhSi_3 [$T_{\min} \sim T_f/3$ (Ref. 6)]. The sharpness of the minimum in U_2IrSi_3 and its temperature being almost identical with T_f are very strong hints that the formation of the ferromagnetic cluster glass state and the rapid increase of $\rho(T)$ are related.

Magnetization $M(H)$ and magnetic relaxation $M_{IRM}(t)$ measurements shown in Fig. 5 for U_2IrSi_3 also reveal the typical features of FM cluster glass: the $M(H)$ curve at 4.2 K shows a evident jump below 500 Oe and then increases monotonously up to 230 kOe [Fig. 5(a)]. The inset of Fig. 5(a) illustrates the hysteresis loop carefully measured by using the SQUID magnetometer up to 5 kOe. From this curve the coercive field H_C is determined to be 340 Oe at 4. 2 K. As shown on the semilogarithmic plots in Fig. 5(b), the evident isothermal remanent magnetization M_{IRM} is observed for U_2IrSi_3 at low temperatures, and the decay of $M_{IRM}(t)$ is remarkably slow. It is well known that in literature various functions, e.g., the *simple* logarithmic time dependence $M_{IRM}(t) \propto \ln(t/t_0)$ (Refs. 16 and 17) and the power law $M_{IRM}(t) \propto t^{-\alpha(t,H)}$ (Ref. 18) have been used to fit the observed magnetic relaxation data for different SG (or SG-like) systems. In the present case of U_2IrSi_3, however, both of them cannot independently fit the data well over the full time

range studied, although the former and latter seem to be better for fitting the data at long times (>200 s) and at short times, respectively. As shown by the solid lines in Fig. 5(b), a better representation of our data is given by the logarithmic form of $M_{IRM}(t)=M_0(T)-S(T)\ln(t+t_0)$, with the fitting parameters $M_0=0.184$ and 0.379 emu/g, $S=2.39\times10^{-3}$ and 1.58×10^{-3} emu/g, and $t_0=317.4$ and 111.4 s for $T=5$ and 8 K, respectively.

Summarizing the various data, the NMAD system U_2IrSi_3 shows the typical features of FM cluster glass behavior. Although a perfect explanation for the mechanism of the cluster glass state formed in this compound is impossible at present, it can be qualitatively considered that the statistical distribution of Ir and Si atoms in the crystal lattice could vary the electronic environment around the U atoms and introduce the formation of magnetic clusters with a randomly frustrated exchange interaction between them necessary for the cluster glass state. This mechanism may also be operating in some other 2:1:3 systems. Moreover, a Kondo-lattice-like behavior is observed in the temperature dependence of the electrical resistivity. It seems likely that the large (T-linear) specific heat coefficient γ perhaps does not result from cluster freezing alone, and U_2IrSi_3 can be considered as a FM cluster glass system showing Kondo-lattice-like effects.

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