Re-entrant Spin Glass Behavior in U$_2$NiSi$_3$

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The results of DC magnetization, AC susceptibility, specific heat and magnetic relaxation measurements on a well-annealed polycrystalline U$_2$NiSi$_3$ sample are reported. The temperature dependencies of the DC magnetization and AC susceptibility show a cusp at a characteristic temperature $T_c$ and a weak ferromagnetic transition at $T_F$. The temperature $T_F$ strongly depends on the applied magnetic field and frequency. Below $T_F$, the magnetic relaxation measurements reveal a decay of the isothermal remnant magnetization versus time which is drastically slower than above $T_F$. No anomalies around $T_F$ are observed in the specific heat data which rules out the existence of usual long-range spatial magnetic order at $T_F$. These results can be considered as clear evidence for the formation of a re-entrant spin glass state in U$_2$NiSi$_3$. The necessary randomness in the U-U magnetic exchange interactions arises from a statistical distribution of Ni and Si atoms on the crystallographic sites of the U$_2$NiSi$_3$ crystal lattice.

**Key words:** U$_2$NiSi$_3$, re-entrant spin glass, DC magnetization, AC susceptibility, specific heat, magnetic relaxation

1. Introduction

Recently new ternary uranium compounds U$_7$TSi$_5$ ($T$=transition metals), crystallizing in \(\text{AlB}_2\)-derived hexagonal structure, have been prepared \(^{13}\). X-ray and electron diffraction studies show that \(T\) and Si atoms in these compounds, with the exception of U$_7$O$_5$Si$_5$ and U$_7$RuSi$_5$, are randomly distributed into the trigonal prisms of a primitive hexagonal array of uranium atoms \(^{13}\). The disordering of transition metal elements and silicon is expected to cause some anomalous magnetic properties. We have reported the formation of low-temperature spin glass state in U$_7$PdSi$_5$ \(^9\) and U$_7$PtSi$_5$ \(^9\) resulting from the random distribution of Pd (Pt) and Si atoms. In order to look for such effects in other compound and also as a continuation of our efforts to understand the magnetic behavior of this class of compounds, the investigation on U$_2$NiSi$_3$ has been undertaken.

2. Experimental details

Polycrystalline sample of U$_2$NiSi$_3$ was prepared by arc melting of the element components (with purity of 3N for U, 4N for Ni, and 6N for Si) in an argon atmosphere. The button was turned over and re-melted several times to ensure good homogeneity. The sample was sealed in a evacuated quartz tube and annealed at 800°C for 240h. The X-ray diffraction patterns show that the sample is a single phase of the hexagonal AlB$_2$-type structure with lattice parameters \(a=3.979\ \text{Å}\) and \(c=3.949\ \text{Å}\). The DC magnetization, AC susceptibility, magnetic hysteresis and magnetic relaxation measurements were performed between 5 and 40K in magnetic fields up to \(\pm 10000\text{G}\) using a superconducting quantum interference device (SQUID) magnetometer. The specific heat measurements were carried out with an adiabatic method at \(1.6<T<40\text{K}\) under zero fields.

3. Results and discussion

The temperature dependence of DC magnetization was measured under various magnetic fields, in two regimes of the sample cooling: with an applied field (FC) and without an applied field (ZFC). The temperature variation of the ZFC magnetization \(M_{\text{ZFC}}\) divided by the applied magnetic field \(H\), of U$_2$NiSi$_3$ is displayed in Fig. 1. For small fields the \(M_{\text{ZFC}}/H\) curve exhibits a sharp cusp at a temperature \(T_c=23.7\ \text{K}\) (\(H=10\ \text{G}\)) and then a strong decrease near 26 K with increasing temperature. With increasing the external magnetic field, this cusp loses its sharpness and both peak intensity and peak temperature decrease rapidly. The resultant rounded maximum under \(H=6000\ \text{G}\) are observed at about 7 K. The appearance of a well defined peak in ZFC magnetization at characteristic temperature \(T_c\), which depends strongly on the applying magnetic field is a typical feature of spin-glass-like systems \(^9\). From this result the \(T_c\) vs \(H\) phase diagram, as displayed in the inset of Fig.1, is established.

![Fig.1 M_{ZFC}/H vs T curves for U_2NiSi_3 in various DC magnetic fields. The inset shows the T_c vs H phase diagram.](image)

It is well known that the susceptibility of usual long-range-order Heisenberg antiferromagnet shows a sharp peak at Néel temperature $T_N$. At low fields, however, $T_N$ decreases very slowly with increasing $H$. The rapid drop of the temperature $T_c$ in the low field region illustrated in the inset of Fig.1 appears to be an intrinsic feature of spin glasses. On the other hand, above $T_F$, the magnetization of
U$_2$NiSi$_3$ decreases rapidly with increasing temperature like a ferromagnet with an inflection point of $T_c=28$ K. The negative value of $M_{ZFC}$ observed in a field of 10G arises because the sample is cooled under a net negative field, present above the superconducting solenoid as described in detail by Rao et al$^3$. Figure 2 shows the temperature dependence of the reduced ZFC and FC magnetizations for U$_2$NiSi$_3$ in a field of 1000G. It is clear from this figure that the ZFC and the FC curves show the evident different features below a temperature $T_c=16$ K. The former is strongly dependent on both the magnetic history and the elapsed time, and exhibits a maximum at about $T_f$ (H=1000 G)=14 K. Whereas the latter is reversible, i.e. when we cycled the temperature back and forth in a constant field, the $M_{RC}/H$ curve traced the same path, and has a tendency to approach a constant value at low temperatures. This indicates that the metastable and irreversible states are formed by the applied magnetic field applied below $T_c$. It is note that the $M_{ZFC}/H$ curve begins to deviate from the $M_{RC}/H$ curve below $T_c=16$ K. This behavior is typical for a re-entrant spin glass$^8$.

$\text{Fig.2 Difference between the ZFC and FC magnetizations for U}_2\text{NiSi}_3$.  

The complex susceptibility of U$_2$NiSi$_3$ was studied in an AC field of 1 Oe at several frequencies ranging from 0.1 to 1000Hz. As shown in Fig. 3, both the real and the imaginary components of the susceptibility show pronounced maxima, the positions of which are frequency dependent. Note the sharp cusp in $\chi'$ at 23.7K ($\nu=0.1$Hz) denoting the freezing temperature $T_f$ and how $T_f$ shifts to higher temperatures with increase of $\nu$. Using the data shown in Fig. 3 the $T_f$ vs $\nu$ phase diagram is also established and displayed in the inset of Fig. 3. We can calculate the initial frequency shift of $T_f$: 

\[ \delta T_f = \frac{\Delta T_f}{T_f \Delta \log \nu} = 0.021 \pm 0.005 \]  

for U$_2$NiSi$_3$. This value is typical for metallic spin glasses, e.g., CuMn:0.005 and URh$_2$Ge$_2$:0.025. On the other hand, the maxima occur in $\chi'$ and $\chi''$ at slightly different temperatures. At 1000Hz, for example, the $\chi'$ curve exhibits a maximum at 24 K whereas the peak in the $\chi''$ curve occurs at 23 K. Also $\chi''$ is not zero up to $T$ being risen to $T_c=28$ K. These are the characteristic features of re-entrant spin glasses$^8$ indicating a ferromagnetic transition to occur between the high-temperature paramagnetic and low-temperature spin-glass states. This result is consistent with the DC magnetization measurements (Fig. 1). These features show that U$_2$NiSi$_3$ behaves like a re-entrant spin glass$^9$.

Fig.3 Temperature dependence of the AC susceptibility of U$_2$NiSi$_3$ measured at various frequencies. A $T_f$ vs $\nu$ phase diagram is shown in the inset.

Remanence effect and magnetic relaxation on macroscopic time scale are also the striking phenomenon for spin glasses$^8$. These features are apparently observed in the U$_2$NiSi$_3$ sample as displayed in Figs. 4 and 5. Fig. 4 shows the hysteresis loop measured at 5 K. The remanence is about 0.02 $\mu_T$ per U atom. The remanence effect can be explained to be due to the existence of anisotropy. The magnetic relaxation of U$_2$NiSi$_3$ was studied by measuring the isothermal remanent magnetization $M_{REM}$ as a function of time $t$ below the freezing temperature $T_f$. The sample was cooled in zero field from $T \gg T_f$ to 5 K, and then a field of 5000 G was applied for about 5 min. After switching off the field, $M_{REM}$ was recorded as a function of $t$. As shown in Fig. 5, the remanent magnetization decays very slowly, a nonzero remanent magnetization could still be detected after 3 h. Fischer and Hertz$^{10}$ explained the long time magnetic relaxation effect observed in spin glasses based on the qualitative ideas about the glassiness of the spin glass state, with many possible configurations separated by barriers of varying heights.

Fig.4 Magnetic field dependence of the magnetization of U$_2$NiSi$_3$ at 5K.

the $T$-Si sublattice is strongly dependent on the nature of the $T$ element. For instance, Ru and Si atoms are perfectly ordered in U$_2$RuSi$_3$ (and thus no spin glass behavior was observed in this compound); Rh-Si network in U$_2$RhSi$_3$ exhibits a partial random arrangement; in U$_2$PdSi$_3$, the distribution between Pd and Si atoms is almost statistical (and thus a spin glass state is formed). For U$_2$NiSi$_3$, it is believed that at least part of the Ni and Si atoms are randomly distributed in the 2D Ni-Si network. The statistical arrangement of Ni and Si atoms seems to introduce the randomly frustrated U-U exchange interaction necessary for the occurrence of the spin glass state. In this respect, we can expect that if the stoichiometric relation between Ni and Si atoms in U$_2$NiSi$_3$ is changed within certain limits, the similar derived AlB$_2$-type structure and spin-glass-like behavior should also be observed.

4. Conclusion

In conclusion, our results of DC magnetization, AC susceptibility, magnetic relaxation and specific heat measurements on a well annealed polycrystalline U$_2$NiSi$_3$ sample can be considered as clear evidence for the formation of a spin glass state. However, from the characteristic behavior of the AC susceptibility we suggest that the U$_2$NiSi$_3$ intermetallic is not a simple spin glass but an re-entrant spin glass which exhibit ferromagnetic-like properties between the high-temperature paramagnetic and low-temperature spin glass regions. The statistical distribution of Ni and Si atoms on one site of the U$_2$NiSi$_3$ crystal lattice varies the electronic environment around the U atoms and seems to introduce the randomly frustrated U-U exchange interactions necessary for the occurrence of the spin glass state.

Reference