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Ferromagnetic and spin-glass properties of single-crystalline U_2NiSi_3[*]

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A single crystal of U_2NiSi_3 was investigated by means of magnetization, electrical resistivity and heat capacity measurements. Whereas the DC magnetic data clearly manifest strongly anisotropic ferromagnetism, the AC magnetic susceptibility data are consistent with spin-glass behaviour, reported previously for polycrystalline samples. Moreover, no distinct anomalies around T_C occur in the specific heat and electrical resistivity characteristics. Altogether the results obtained for single-crystalline U_2NiSi_3 indicate rather an unusual state of coexistence of ferromagnetism and spin-glass freezing.

Key words: *uranium; ternary silicide; spin glass; ferromagnetism*

1. Introduction

Most of the ternary intermetallic phases U_2TSi_3 (T = 3d-, 4d- or 5d-electron transition metal) crystallize in a hexagonal structure of the AlB_2 type or its disordered derivatives [1, 2]. Recently, they have attracted much attention because of their unusual magnetic properties related to atomic disorder or topological frustration in the uranium sublattice [3, 4]. Polycrystalline samples of U_2NiSi_3 were previously studied and characterized as a cluster-glass system with the spin-freezing temperature $T_f = 22$ K [3, 5]. On the contrary, a neutron diffraction experiment performed on a single crystal has revealed a long-range ferromagnetic ordering below $T_C = 30$ K with sizeable uranium magnetic moments of $0.6\mu_B$ oriented perpendicular to the hexagonal c axis [6]. This indispensable discrepancy motivated us to undertake a reinvestigation of the bulk properties of U_2NiSi_3 on single-crystalline specimens.

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2. Experimental

Single crystal of U_2NiSi_3 was grown by the Czochralski pulling method in a tetra-arc furnace under argon atmosphere. Using a KUMA diffraction four circle diffractometer equipped with a CCD camera, its crystal structure was proved to be hexagonal AlB_2-type. DC magnetic measurements were performed in the temperature range 1.72–400 K and in magnetic fields up to 5 T using a Quantum Design SQUID magnetometer. The AC magnetic susceptibilities were measured between 10 and 40 K within the frequency range 10–10 000 Hz using an Oxford Instruments AC-susceptometer. The electrical resistivities were measured from 5 to 300 K by a conventional four-point DC technique. Heat capacities were studied within the temperature interval 2–100 K employing a quantum design PPMS platform.

3. Results and discussion

Figure 1 shows low-temperature dependences of the magnetization measured in zero-field-cooled (ZFC) and field-cooled (FC) regimes in a magnetic field applied parallel ($\sigma_{\|}$) and perpendicular ($\sigma_{\perp}$) to the *c* axis. The isothermal magnetization measured at 2 K with the field applied along the two characteristic directions is presented in Fig. 2. The overall shapes of these curves as well as the magnitude of $\sigma_{\perp}$ obtained in the FC mode unambiguously indicate strongly anisotropic ferromagnetism with a pronounced domain effect. The magnetic moments are confined to the basal hexagonal plane. The Curie temperature, defined as the inflection point on the $\sigma_{\perp}(T)$ variation, amounts to 26 K.

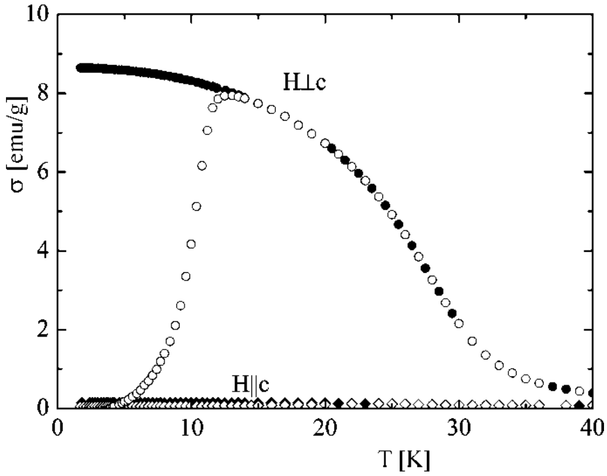

Fig. 1. Temperature dependences of the magnetization in U_2NiSi_3 measured in the ZFC (open symbols) and FC (full symbols) regimes with magnetic field ($H = 1$ kOe) applied parallel and perpendicular to the *c*-axis

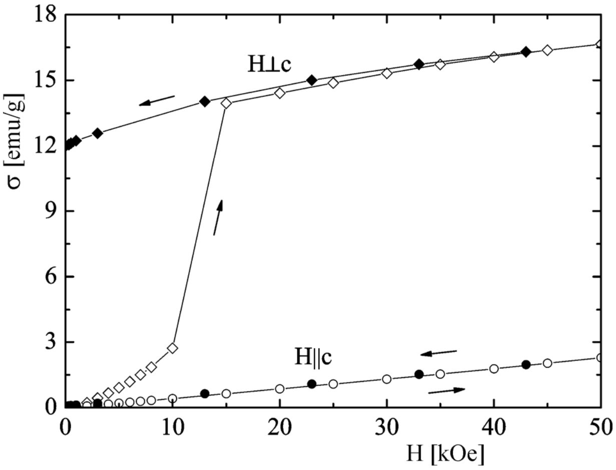

Fig. 2. Magnetization isotherms for U_2NiSi_3 taken at 1.7 K with increasing and decreasing magnetic field (H = 1 kOe) applied parallel and perpendicular to the c-axis

As shown in Fig. 3, a strong magnetocrystalline anisotropy is observed also in the paramagnetic state. The magnetic susceptibility measured along the c axis ($\chi_{\|}$) is much smaller than the component taken within the ab plane ($\chi_{\perp}$). Above ca. 100 K, the $\chi_{\|}(T)$ and $\chi_{\perp}(T)$ variations can be described by the modified Curie–Weiss law with the following parameters: $\chi_{0\|} = 6\times10^{-4}$ emu/mol, $\mu_{\mathrm{eff}\|} = 2.47\mu_B$, $\theta_{p\|} = -14$ K and $\chi_{0\perp} = 8\times10^{-4}$ emu/mol, $\mu_{\mathrm{eff}\perp} = 2.64\mu_B$ and $\theta_{p\perp} = 31$ K.

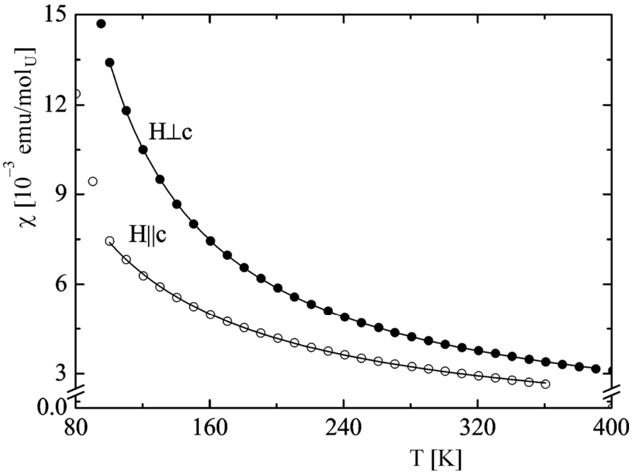

Fig. 3. Temperature dependences of the magnetic susceptibility of U_2NiSi_3 measured with magnetic field (H = 10 kOe) applied parallel and perpendicular to the c-axis. The solid lines represent fits to the experimental data of the modified Curie–Weiss law with the parameters given in the text

The ferromagnetic ordering in single-crystalline U_2NiSi_3, being evident in the magnetic characteristics, manifests only faintly in the heat capacity and electrical transport data.

The electrical resistivity, measured with the current flowing along ($\rho_{\|}$) and perpendicular ($\rho_{\perp}$) to the *c* axis, shows only tiny anomalies at T_C (see Fig. 4). Above ca. 60 K, the two resistivity components change with the temperature in a metallic manner, yet at lower temperatures some anomalous features are seen. Both curves exhibit shallow minima near 50 K, and in the ordered state the resistivities slightly increase with decreasing temperature. Worth noting is also that the overall changes in the values of $\rho_{\|}$ and $\rho_{\perp}$ over the entire temperature range are very small. The observed behaviour is likely to results from the presence of sizeable atomic disorder (i.e., Ni and Si atoms occupying same crystallographic site) in the unit cell of the compound studied.

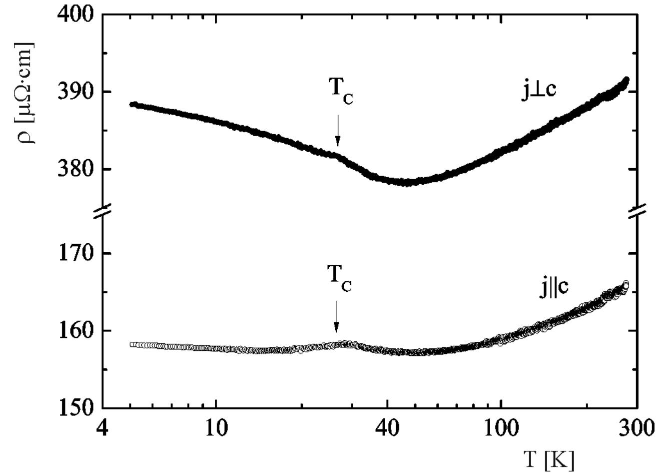

Fig. 4. Temperature dependences of the electrical resistivity of U_2NiSi_3 measured with the current flowing along and perpendicular to the *c*-axis. The arrows indicate the Curie temperature

The temperature dependence of the specific heat of U_2NiSi_3 is displayed in Fig. 5. The magnetic phase transition at T_C manifests itself only as a small kink on the $C(T)$ curve. As shown in the inset to Fig. 5, the plot C/T vs. T^2 yields for $T \to 0$ rather large linear contribution to the specific heat of about 140 mJ/(mol$_U$·K^2). This enhancement of the low-temperature heat capacity can be considered as another indication of the structural disorder in the sublattice of nonmagnetic atoms.

The results of the AC magnetic susceptibility studies are summarized in Fig. 6. Both components to the AC susceptibility, real and imaginary, form pronounced peaks close to T_C. Their positions depend on the frequency of alternating field, namely they systematically shift towards higher temperatures with increasing frequency. Simultaneously, the magnitude of the real component at the maximum decreases and that of the imaginary component rapidly increases. Such a behaviour is characteristic of spin glasses. The frequency dependence of the freezing temperature, defined as a maximum on the $\chi'(T)$ curve, is shown in the inset to Fig. 6. This variation can be well described using the empirical Vogel–Fulcher law [7]

$$\omega = \omega_0 \exp\left(-E_a/k_\mathrm{B}\left(T_f - T_0\right)\right)$$

where ω_0 is the characteristic frequency, E_a denotes the activation energy and T_0 is the Vogel–Fulcher temperature.

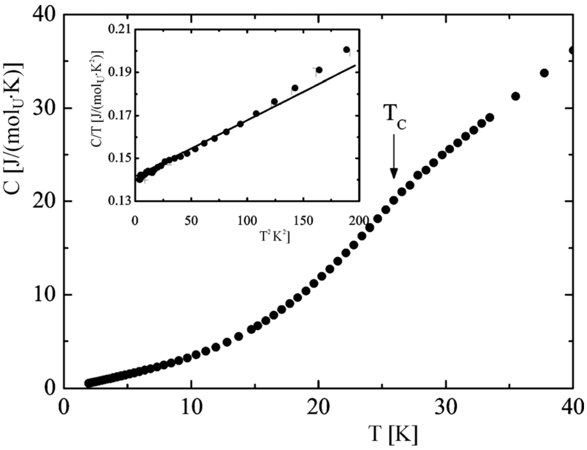

Fig. 5. Temperature variation of the specific heat of U_2NiSi_3; the arrow indicates the Curie temperature, as derived from the magnetic measurements. The inset displays the low-temperature data in the representation C/T versus T^2. The solid line emphasizes a straight-line behaviour for $T \to 0$

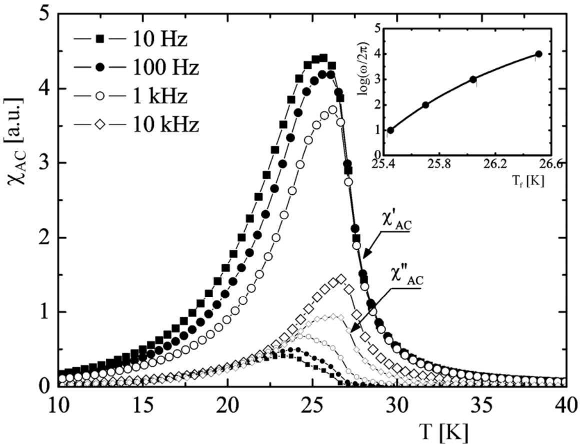

Fig. 6. Temperature dependence of real (χ'_{AC}) and imaginary (χ''_{AC}) components of the AC magnetic susceptibility of U_2NiSi_3 between 10 and 40 K (H = 10 Oe) at the frequencies 10 Hz ≤ $\omega/2\pi$ ≤ 10 kHz. The inset shows the frequency dependence of the freezing temperature T_f. The solid line represents the fit to the experimental data of the Vogel–Fulcher law with the parameters given in the text

The least squares fit of the experimental T_f versus $\omega/2\pi$ data for U_2NiSi_3 yields the following parameters: $\omega_0/2\pi = 10^9$ Hz, E_a/k_B = 31.9 K and T_0 = 23.7 K. The characteristic frequency is of the order of magnitude typical of metallic spin-glass systems.

Moreover, the characteristic temperature T_0 is somewhat lower than the Curie temperature $T_C = 26$ K, as predicted for spin glasses [7].

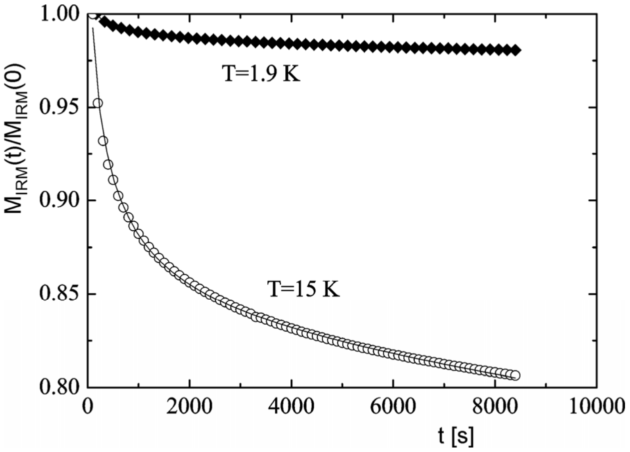

Fig. 7. Time dependence of the isothermal remanent magnetization in U_2NiSi_3 measured at 1.9 and 15 K upon applying magnetic field of 20 kOe perpendicular to the *c* axis for 5 min and switching it off. The solid lines are the fits to the experimental data of the formula $M_{\mathrm{IRM}}(t) = M_0 + \alpha \ln t + \beta \exp(-t/\tau)$ with the parameters given in the text

Figure 7 shows the time dependence of reduced isothermal remanent magnetization (IRM) in U_2NiSi_3 recorded at 1.9 and 15 K. The sample was first zero-field cooled from a temperature much higher than T_f, then a magnetic field of 20 kOe was applied perpendicular to the *c* axis for 5 min and switched off at $t = 0$. As is apparent from Fig. 7, the IRM decay at both temperatures can be accurately described by the equation:

$$M_{\mathrm{IRM}}(t) = M_0 + \alpha \ln t + \beta \exp(-t/\tau)$$

appropriate for non-magnetic atom disorder (NMAD) spin-glasses [4]. The least-squares fitting parameters: $M_0 = 11.9$ emu/g, $\alpha = -0.006$ emu/g, $\beta = -0.004$ emu/g, $\tau = 2872$ s for 1.9 K and $M_0 = 4.19$ emu/g, $\alpha = -0.02$ emu/g, $\beta = -0.01$ emu/g, $\tau = 3454$ s for 15 K are similar to the values reported for other NMAD systems [4, 8, 9]. An important feature of this class of spin glasses is the presence of the exponential term in the expression for $M_{\mathrm{IRM}}(t)$, while for other spin glasses just the logarithmic term weighted by the magnetic viscosity parameter α is sufficient to properly describe their IRM behaviour [7–9].

4. Summary

The results of AC magnetization, magnetic relaxation, specific heat and electrical resistivity measurements of single-crystalline U_2NiSi_3 can be considered as strong evidence for the formation of a spin-glass state in agreement with our initial hypothesis [3] and more recent results [5]. However, the freezing temperature $T_f = 25–27$ K (dependent on the frequency), determined in the present studies, somewhat differs from the value of 22 K given in the previous reports. On the other hand, the DC magnetic data obtained for the U_2NiSi_3 single crystal evidently indicate strongly anisotropic ferromagnetism which seems to support the conclusion derived from the neutron diffraction data [6]. Again, the Curie temperature $T_C = 26$ K is notably different from that reported before ($T_C = 30$ K [6]). Most likely these discrepancies arise due to small differences in the level of atomic disorder and/or possible deviations from the ideal stoichiometry in the previously studied polycrystalline samples and the single crystal investigated in the present work.

In conclusion, new results obtained for a single crystal of U_2NiSi_3 reveal the coexistence of long-range ferromagnetic ordering and spin-glass state with the Curie temperature equal to the freezing temperature. A similar behaviour was previously observed in isostructural compounds U_2RhSi_3 and U_2IrSi_3 which were classified as ferromagnetic cluster glass systems [5, 8]. A deeper insight into the intriguing physical behaviour of this series of uranium ternaries can be expected from neutron diffraction investigations. For this reason both elastic (to provide direct evidence for long-range ferromagnetic order) and inelastic (to study spin dynamics) neutron experiments on U_2NiSi_3 single crystals (they are large enough) are planned in the nearest future.

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