Electronic properties of UCoAl$_{0.75}$Sn$_{0.25}$ single crystal

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Received 19 November 2003; accepted 28 November 2003

Abstract

A single crystal of the UCoAl$_{1-x}$Sn$_x$ quasiternary compound has been grown and studied by measuring the magnetization, specific heat and electrical resistivity at various temperatures and magnetic fields. Similar to the parent compounds UCoAl and UCoSn, the magnetization of their solid solution exhibits uniaxial anisotropy with a strong magnetic response along the c-axis and a weak and nearly temperature independent paramagnetic signal in the basal plane. The evolution of the c-axis magnetization isotherms, as well as the anomalies in the temperature dependence of the specific heat and resistivity point to ferromagnetism in UCoAl$_{0.75}$Sn$_{0.25}$ below $T_C \approx 5.5$ K. The low value of the spontaneous magnetic moment, poor saturation of the magnetization, the pronounced negative magnetoresistance with increasing magnetic field and the temperature and magnetic field dependence of specific heat provide indications of strong fluctuations of the U magnetic moment also in the ferromagnetic state.

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Keywords: Uranium intermetallics; UTX compounds; Ferromagnetism; Magnetic anisotropy; Magnetoresistivity; Specific heat

1. Introduction

The magnetism in UTX compounds (T: transition metal, X: p-electron element) with the hexagonal ZnNiAl-type structure is dominated by the uranium 5f electron magnetic moments. The 5f electron states, however, strongly hybridize with the T and X ligand valence electron states and so the 5f electron magnetism considerably depends on the ligand species although the ligands carry no stable magnetic moments. The UCoAl and UCoSn compounds serve as good examples of this scenario. UCoAl is till now the only known itinerant 5f electron metamagnet[1]. It remains paramagnetic down to lowest temperatures but exhibits a maximum in the temperature dependence of the c-axis magnetic susceptibility around 20 K. When a magnetic field larger than the critical field for metamagnetism $B_c \approx 0.7$ T is applied along the c-axis of UCoAl at low temperatures ($T < 10$ K) a ferromagnetic ordering of the U magnetic moments $\mu_U \approx 0.3 \mu_B$ (just above $B_c$) is induced, which is followed by a considerable increase of $\mu_U$ with further increasing field [2]. The c-axis magnetization does not saturate even in magnetic fields in the range of 40 T [3]. UCoSn, on the other hand, is a ferromagnet with a high Curie temperature ($T_C = 83$ K), a spontaneous magnetic moment along the c-axis $\mu_s = 1.24 \mu_B/\text{f.u.}$, which is almost identical with the saturated moment [1]. UCoAl and UCoSn, as well as the other UTX compounds of this family, exhibit a huge uniaxial magnetic anisotropy with the dominating magnetic response along the c-axis. Within the basal plane, only a weak Pauli susceptibility is observed irrespective the type of ground state [1].

The striking difference between magnetic properties of the two nearly isoelectronic compounds naturally motivated studies of the magnetic behavior in of the UCoAl$_{1-x}$Sn$_x$ solid solutions. Krylov and co-workers [4,5] studied the hyperfine field transferred from U to the Sn nuclei by measuring the Mossbauer effect on the $^{119}$Sn isotope. The composition of the investigated samples covered the entire concentration range between UCoAl to UCoSn by concentration steps of $\Delta x = 0.2$. Ferromagnetism has been observed already for UCoAl$_{0.8}$Sn$_{0.2}$, which is followed by a...
monotonous development of $T_C$ and $\mu_s$ with further increasing Sn content. A more detailed study in the Al-rich region revealed a more complicated concentration dependence of magnetism [6]. Already the slightly Sn-doped compound UCoAl$_{0.75}$Sn$_{0.25}$ proved to be ferromagnetic. However, the concentration dependence of $T_C$ and $\mu_s$ has not been found to be monotonous (see Fig. 1). Both parameters exhibit a local minimum for $x$ in the interval 0.20–0.25. In the concentration range $0.25 < x < 0.6$, $T_C$ as well as $\mu_s$ steeply increase with $x$ and the magnetic properties than rapidly approach the behavior of UCoSn. In particular, the materials become magnetically hard and show a rectangular hysteresis loop. The coercive force ($H_C$) for UCoSn equals 1 T in contrast to the essentially soft magnetism found in the compounds with low Sn content ($H_C < 10$ mT).

The non-monotonous development of magnetism in the UCoAl$_{1-x}$Sn$_x$ system reflects the complicated interplay between the lattice expansion (the lattice parameters increase linearly with increasing the Sn content [6]) and variations of the valence electron states, especially the doping by the 5p electrons provided by Sn instead of the Al 3p electrons. Both factors should influence the formation of the U 5f electron moment, as well as the exchange interactions responsible for magnetic ordering.

In the present paper, we have focused our attention on the UCoAl$_{0.75}$Sn$_{0.25}$ compound, which exhibits the mentioned minimum values of $T_C$ and $\mu_s$. In order to extract the intrinsic anisotropic properties, we have grown a single crystal and subjected it to measurements of the magnetization, specific heat and electrical resistivity with respect to temperature and magnetic field.

2. Experimental details

The UCoAl$_{0.75}$Sn$_{0.25}$ single crystal has been pulled under protective high-purity Ar atmosphere from a 10-g stoichiometric melt by a modified Czochralski tetra-arc method with W electrodes. The purity of the used elementary metals was 3N for U and Co, and 5N for Sn and Al. A pulling speed of 10 mm$^3$h$^{-1}$ was applied, using a W wire as a seed. The check of crystal quality as well as the orientation of the crystal for cutting has been done using the X-ray Laue method. The phase purity of the crystal and the lattice parameters were determined by standard X-ray diffraction of a powder sample prepared from a part of the crystal. The determined lattice parameters $a = 679.2$ pm and $c = 396.4$ pm are in a good agreement with those reported in [6].

The samples used for the study have been spark-erosion cut from the crystal. The magnetization and specific heat as a function of magnetic field and temperature were measured in the temperature range 2–300 K using a Physical Properties Measurement System PPMS-14 (Quantum Design) with a superconducting coil providing a magnetic field up to 14 T. The field and temperature dependence of electrical resistivity were measured by a standard four-point ac method; the measurements at temperatures from 2 to 300 K were made in magnetic fields up to 18 T generated by a superconducting magnet (Oxford Instruments).

3. Results and discussion

Fig. 2 shows the magnetization curves measured on the UCoAl$_{0.75}$Sn$_{0.25}$ single crystal in a magnetic field applied along the $c$-axis for several temperatures and in a field along the $a$-axis at 2 K. By inspecting the data obtained at 2 K for the two principal crystallographic directions, one can see the presence of strong uniaxial anisotropy with the easy magnetization axis along the $c$-axis, typical for the whole family of the UTX intermetallics with the hexagonal ZnNiAl-type structure. In the easy direction, a spontaneous
magnetization is found as a clear sign of ferromagnetism, however, along the $a$-axis a paramagnetic response with a low magnetic susceptibility is measured, which is comparable to the corresponding $a$-axis signals observed for UCoAl and UCoSn. The spontaneous magnetization of UCoAl$_{0.75}$Sn$_{0.25}$ is very small, only 0.17 $\mu_B$/f.u. as derived from Arrott plots (shown in Fig. 3), which is even smaller than the magnetization in UCoAl above the metamagnetic transition. The $c$-axis magnetization moreover shows poor saturation; the high differential susceptibility leads to a magnetization of 0.66 $\mu_B$/f.u. in 14 T. Note that this result rather resembles the high-field behavior of UCoAl, and on the other hand, it contrasts with the very good saturation of magnetization in UCoSn. From the Arrott plot also the value of the Curie temperature, $T_C \approx 5.5$ K, has been derived.

UCoAl$_{0.75}$Sn$_{0.25}$ is magnetically extremely soft, which is unexpected especially for such a highly anisotropic ferromagnet. The coercive field is only 4 mT at 2 K (compared with 100 mT at $x = 0.3$ in UCoAl$_{1-x}$Sn$_x$ [6]).

As seen in Fig. 4, the very large anisotropy persists in the paramagnetic range. The temperature dependence of magnetic susceptibility measured along the $c$-axis down to 70 K obeys the Curie–Weiss law with an effective moment $\mu_{\text{eff}} = 2.66 \mu_B$ per U atom and a paramagnetic Curie temperature $\theta_p = 10$ K. The $\mu_{\text{eff}}$ value is considerably lower than the single U ion values for the $5f^2$ and $5f^2$ configurations (3.58$\mu_B$ and 3.62$\mu_B$, respectively) and falls in the range of $\mu_{\text{eff}}$ values typical for most of U intermetallics [1]. The positive value of $\theta_p$ that roughly corresponds to the $T_C$ value confirms that ferromagnetic interactions dominate in UCoAl$_{0.75}$Sn$_{0.25}$. The magnetic susceptibility along the $a$-axis exhibits weak temperature dependence even at temperatures below $T_C$. This nearly temperature-independent susceptibility in the basal plane is a common feature of all members of the family of the ZrNiAl-type of UTX intermetallics.

Fig. 5 shows the temperature dependence of the electrical resistivity $\rho$ for current along the $c$-axis. The magnetic ordering is indicated on the $\rho(T)$ curve by a small cusp-like anomaly around $T_C$. Surprisingly, this anomaly is removed already by a magnetic field of 1 T applied along the $c$-axis. The low temperature $\rho(T)$ dependence gradually changes with increasing magnetic field from an almost linear one in zero field to a quadratic one in 18 T. As is seen in Fig. 5, the low-temperature resistivity is considerably reduced by the application of a magnetic field along the $c$-axis. In Fig. 6, it...
Fig. 6. Longitudinal magnetoresistivity along the c-axis at 2 and 10 K.

is seen that the longitudinal magnetoresistivity curves measured at 2 and 10 K, respectively, are comparable yielding in both cases about ~15% at 18 T. The fact that there is a considerably negative magnetoresistance which increases with increasing magnetic field and goes hand in hand with the increase in c-axis magnetization without saturating, may be attributed to strong spin fluctuations (namely fluctuations of the U magnetic moment). The latter become gradually suppressed by increasing magnetic field along the c-axis. These aspects of suppression of spin fluctuations by a magnetic field applied along the c-axis seem to be analogous to UCoAl (in fields above 1 T).

Fig. 7 shows the temperature dependence of the specific heat represented by the $C_p/T$ versus $T^2$ plot. In zero magnetic field, a broad anomaly associated with the magnetic ordering terminates at 5.5 K in agreement with the $T_C$ value obtained from the Arrott plots. The magnetic entropy at 6 K amounts to only about 90 mJ mol$^{-1}$ K$^{-1}$, which clearly points to itinerant type of magnetism. (Note that $S = 0$ in case of weak itinerant magnetism with magnetic moments vanishing above $T_C$.) The electronic specific heat coefficient $\gamma = 113$ mJ mol$^{-1}$ K$^{-2}$ is much higher than the corresponding value determined for the both parent compounds (65 and 61 mJ mol$^{-1}$ K$^{-2}$ in UCoAl and UCoSn, respectively [1]). Similar to the resistivity, the $T_C$ related specific heat anomaly is wiped out already by a field of 1 T applied along the c-axis. However, it does not shift to higher temperatures which usually happens in ferromagnets. The $\gamma$ value gradually decreases with increasing field and amounts to 77 mJ mol$^{-1}$ K$^{-2}$ at 14 T, which corroborates the scenario proposing suppression of spin fluctuations in magnetic fields.

4. Conclusions

We have grown a UCoAl$_{0.75}$Sn$_{0.25}$ single crystal and subjected it to measurements of the magnetization, specific heat and electrical resistivity with respect to temperature and magnetic field. Similar to the parent compounds UCoAl and UCoSn, the magnetization of our solid solution exhibits uniaxial anisotropy with a strong magnetic response along the c-axis and a weak and nearly temperature independent signal in the basal plane. The temperature dependence of the c-axis magnetization, specific heat and resistivity can be interpreted in terms of ferromagnetism in UCoAl$_{0.75}$Sn$_{0.25}$ below $T_C \approx 5.5$ K. The low value of the spontaneous magnetic moment at 2 K, poor saturation of the magnetization, negative magnetoresistance with increasing magnetic field and the temperature and magnetic field dependence of the specific heat (especially the $\gamma$ coefficient) reflect strong fluctuations of the U magnetic moment in the ferromagnetic state, which become gradually frozen out in magnetic field applied along the c-axis.

Acknowledgements

The work is a part of the research program MSM113200002, which is financed by the Ministry of Education of the Czech Republic. We acknowledge also the support of the Czech Grant Agency under the grant number 202/02/0739. It was also partly supported by a grant from NSF (grant number: DMR-0094241). Work at the National High Magnetic Field Laboratory was performed under the auspices of the NSF, the US Department of Energy and the State of Florida.
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