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Magnetic Phase Diagram of UCoAl

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We report precision c -axis magnetic measurements on a high-quality single crystal of the heavy fermion metamagnet UCoAl. The metamagnetic transition at H_M changes from 1st order at low temperature to a crossover at high temperature. H_M is nearly linearly increasing with increasing temperature up to a critical temperature T_0 . The critical temperature T_0 is determined from both the field and the temperature dependences of magnetization to be ~ 11 K. The field dependence of the Sommerfeld coefficient γ is estimated from $M(T)$ by using a Maxwell relation. $\gamma(H)$ shows a step-like decrease at H_M . This behavior is consistent with the previous reports of specific heat and resistivity measurements at low temperatures.

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I. INTRODUCTION

Metamagnetism and quantum criticality in strongly correlated electron systems have attracted much attention because of the possibility of new quantum phases, and appear related to unconventional superconductivity and non-Fermi liquid behavior. In itinerant ferromagnets with tricritical points, the metamagnetic transition from the paramagnetic(PM) phase to the ferromagnetic(FM) phase possesses the wing structure of the first order transition in the temperature-pressure-magnetic field phase diagram [1–4]. The first-order metamagnetic transition at low temperature reaches a critical end point above which the transition changes to a crossover. UCoAl is one of the best materials in which to investigate such a

metamagnetic transition from a PM to a FM phase.

The heavy fermion UCoAl is known to be a unique system with a metamagnetic transition from a paramagnetic to a ferromagnetic ground state in uranium-based compounds. Its crystal structure is the ZrNiAl-type hexagonal structure (space group: $P\bar{6}2m$, No. 189) without inversion symmetry. When a magnetic field is applied along the c -axis (magnetic easy axis), the paramagnetic ground state becomes a field-induced ferromagnetic state as the magnetic field increases through the metamagnetic transition at H_M [5,6]. The magnetization curves are very anisotropic between $H||c$ -axis and $H \perp c$ -axis, indicating an Ising-like magnetic behavior. From investigations of chemical pressure and uniaxial pressure effects, UCoAl is recognized as a nearly ferromagnetically ordered system [7,8]. Although the ground state at ambient pressure is paramagnetic, a negative pressure ($P_c \sim -0.2$ GPa) will induce a ferromagnetic ground state [9].

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Fig. 1. Single-crystal ingot of UCoAl grown by using the Czochralski pulling method.

In this paper, we report the results of magnetic measurements performed on a high quality single crystal, comparing our results with those of previous investigations. We also present the magnetic phase diagram for fields along the c -axis at ambient pressure.

II. EXPERIMENTAL

A single crystal (Fig. 1) has been grown by the Czochralski pulling method using a tetra arc furnace. The starting materials of U (purity: 99.95%-3N5), Co (3N), and Al (6N) were melted on a water-cooled copper hearth under a high-purity Ar gas atmosphere. The ingot was turned over and was melted again. This process was repeated several times in order to obtain a homogeneous polycrystalline ingot. The ingot was subsequently pulled at a rate of 15 mm/h for the crystal growth. A bar-shaped sample was cut from the ingot for the magnetic measurements. The residual resistivity ratio was approximately 27 for a current along the c -axis. This indicated that the crystal was the highest quality reported up to now. The magnetic measurements were performed using a commercial SQUID magnetometer (Quantum Design MPMS) at temperature down to 1.8 K in magnetic fields up to 7 T.

III. RESULTS AND DISCUSSION

Figure 2 shows magnetization curves at selected temperatures. At 2 K, a sharp jump in the magnetization is observed at a metamagnetic transition field H_M near 0.7 T. With increasing temperature, the metamagnetic transition field shifts to higher field and becomes broader. Finally, the transition smears at high temperatures. We determined the transition field from the peak position of the derivative of the magnetization with respect to the field and plot those fields in the temperature vs magnetic field phase diagram shown in Fig. 3. As mentioned in previous reports, this transition is first order and has magnetic hysteresis around H_M at low temperatures. Figure 2(b) shows the hysteresis loop of the magnetization curve below 11 K. The hysteresis width decreases

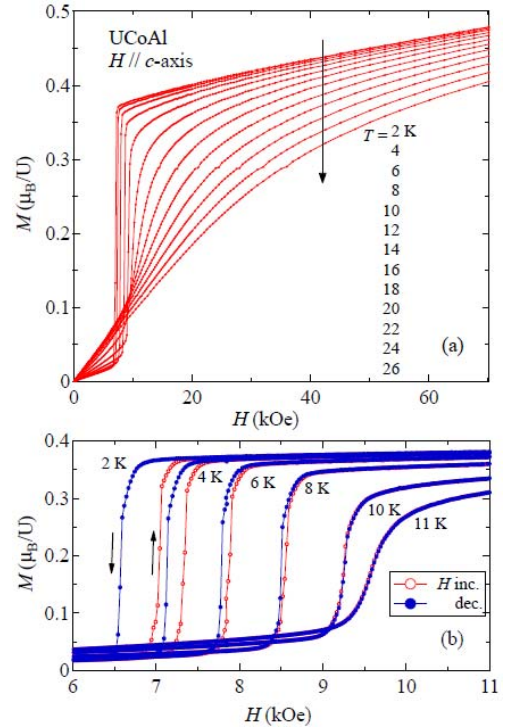


Fig. 2. (Color online) (a) Magnetization curves at selected temperatures. (b) Hysteresis loop of magnetization around a metamagnetic transition at low temperatures.

with increasing temperature as shown in Fig. 3. At temperature above 12 K, the hysteresis can not be observed within the experimental accuracy. This fact indicates that the first order transition terminates around $T_0 \sim 11$ K. This result is in good agreement with the recent results of magnetoresistance and magnetostriction [4].

From thermodynamics, we expect a slope of dH_M/dT to be zero at $T = 0$ K. However, at temperature above 4 K, H_M is roughly linearly increasing with increasing temperature up to T_0 . This positive slope of the first-order boundary indicates that the entropy S decreases with increasing field across the H_M . To estimate ΔS , we use the Clausius-Clapeyron relation

$$\frac{dH_M}{dT} = -\frac{\Delta S}{\Delta M}. \quad (1)$$

ΔS at 4 K is estimated to be 55 mJ/K·mol where $dH_M/dT \sim 0.033$ T/K and $\Delta M \sim 0.3\mu_B/\text{f.u.}$. This value is in good agreement with the previous results for the magnetocaloric effect [10].

We also precisely measured the temperature dependence of magnetization $M(T)$ for fields along the c -axis at selected fields. Figure 4 displays these results for increasing temperature. At fields below 0.6 T, $M(T)$ shows a broad peak structure around 20 K. This behavior is typical for heavy-electron systems, such as CeRu_2Si_2 , UPt_3 , and URu_2Si_2 . This peak structure shifts to lower temperature with increasing field, and it seems to merge with the field-induced ferromagnetic state at low tem-

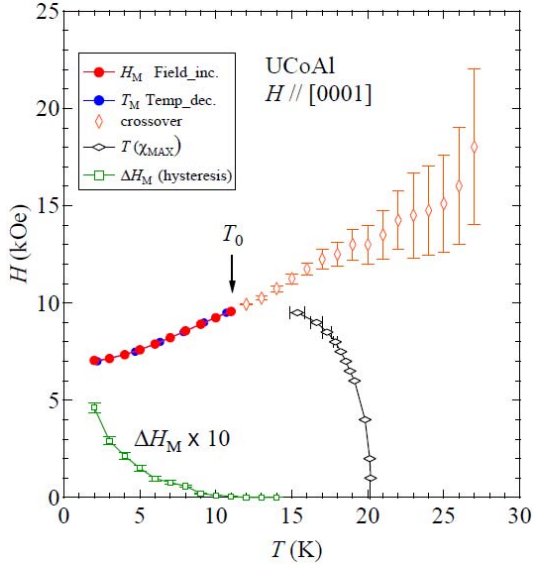


Fig. 3. (Color online) Magnetic field vs Temperature phase diagram and the width of the hysteresis at H_M of UCoAl for fields along the c -axis.

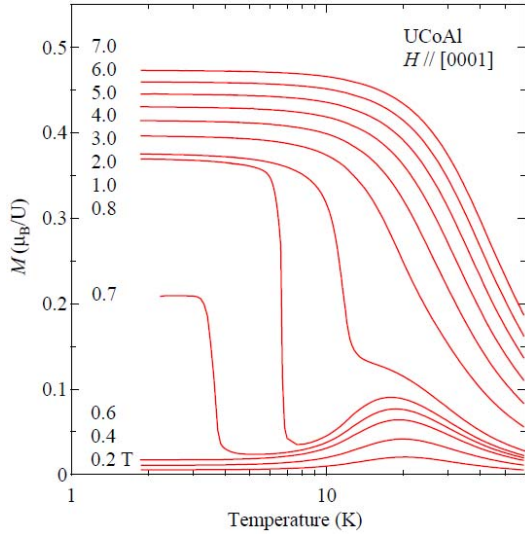


Fig. 4. (Color online) Temperature dependence of the magnetization of UCoAl for fields along the c -axis.

perature. The interesting point is that the peak position seems to be connected to the end point of the first-order transition, as shown in Fig. 3.

Using the thermodynamic Maxwell relation $d^2M/dT^2 = d\gamma/dH$, we try to estimate the field dependent part of the Sommerfeld coefficient $\Delta\gamma(H)$ based on the data of $M(T, H)$ as shown in Fig. 5, although there is a phase boundary of first order at H_M . Here we took the γ_0 value of UCoAl as $73 \text{ mJ/K}^2 \cdot \text{mol}$ at zero field [10]. Although the step-like behavior of γ in Fig. 5 is qualitatively consistent with both the field dependence of C/T and variation of the A coefficient of the resistivity [4, 10], the absolute value

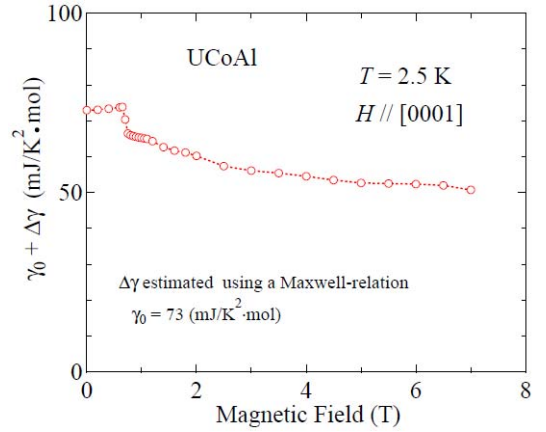


Fig. 5. (Color online) Field dependence of γ . $\Delta\gamma(H)$ was estimated from $M(T)$ by using a Maxwell-relation. The γ_0 value was taken from Ref. [10].

of $\Delta\gamma$ across H_M is only half that of the reported data [4]. This underestimate of $\Delta\gamma(H)$ can be attributed to the discontinuous volume change at H_M , which was neglected in this estimation for simplicity. Indeed, a large negative magnetostriction has been reported accompanying the metamagnetic transition for fields along the c -axis [4,11].

IV. CONCLUSIONS

We grew a high quality single crystal of UCoAl and performed precision magnetic measurements for field along the c -axis. The metamagnetic transition at H_M changes from 1st order at low temperature to a crossover at high temperature. H_M is nearly linearly increasing with increasing temperature up to a critical temperature determined to be approximately 11 K by both field and temperature dependence of magnetization. The field dependence of the Sommerfeld coefficient γ was estimated from $M(T)$ using a Maxwell relation. $\gamma(H)$ shows a step like decrease at H_M . This behavior is consistent with the previous reports on specific heat and resistivity at low temperatures.

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