

Effects of external pressure on the 5*f*-band metamagnetism in UCoAl

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UCoAl exhibits below 16 K anisotropic 5*f*-band metamagnetism induced by magnetic fields as low as ~ 1 T applied along the *c* axis of the hexagonal structure. No metamagnetic transition is observed in fields up to 40 T applied within the basal plane. The critical field of the metamagnetic transition B_c is composition dependent within the homogeneity range. The metamagnetic transition is found to be very sensitive to external pressure. The value of B_c increases with the rate $dB_c/dp=0.27$ T/kbar, whereas the magnetization gained through the transition becomes reduced. The critical pressure of 24 kbar (corresponding to $\sim 2\%$ volume contraction) for disappearance of metamagnetism in UCoAl has been estimated. Relations of UCoAl to 3*d*-band metamagnets are discussed. [S0163-1829(97)05609-9]

I. INTRODUCTION

UCoAl crystallizes in the hexagonal ZrNiAl-type crystal structure.¹ This is a layered structure consisting of two types of basal plane layers (U-Co and Co-Al, respectively) piled up along the *c* axis. Strong bonding of uranium 5*f* states within the basal plane in cooperation with the 5*f* orbital moment yields uniaxial anisotropy with the easy-magnetization direction in the *c* axis in izostructural UTX (*T*=transition metal, *X*=*p* metal) compounds.

Several controversial interpretations of the unusual low-temperature (below 16 K) magnetization behavior of UCoAl (no spontaneous magnetization, metamagnetic transition with rather a low critical field $B_c \sim 1$ T applied along the *c* axis) can be found in the literature.¹⁻⁵ First, antiferromagnetic ordering below 16 K was claimed in analogy to the antiferromagnet UNiAl ($B_c = 11.3$ T),⁶ although B_c in UCoAl is an order of magnitude lower. Then, absence of any anomaly in both the specific heat and the electrical resistivity which would indicate a magnetic phase transition around 16 K, led to the conclusion about the paramagnetic ground state. The metamagnetic transition to the state with U magnetic moment μ/U atom $\approx 0.3 \mu_B$ oriented along the *c* axis was in this context attributed to band metamagnetism.² This conclusion has been corroborated by results of *ab initio* electronic structure calculations³ and polarized-neutron-diffraction experiment on a single crystal.⁵ Later on, a ferromagnetic ground state with relatively large uranium moment ($0.8\mu_B$) was claimed after fitting powder neutron-diffraction data,⁴ but ignoring completely the previous single-crystal results.^{2,5}

In contrast to well-known band metamagnets YCo₂ and LuCo₂,⁷ in which the 3*d* electrons of Co are responsible for the extraordinary magnetic behavior, the 5*f* electrons of U determine the situation in UCoAl whereas Co does not contribute substantially to the magnetic moment (as evidenced by polarized neutron-diffraction investigation of a single

crystal⁵). In UCoAl also a strong influence of deviations from the 1:1:1 stoichiometry on the magnetic behavior was observed within rather a wide homogeneity range found for the compound. The value of B_c is enhanced (reduced) in U-rich (-poor) samples. Consequently a small spontaneous ferromagnetic moment appears in the sample with a deficit of uranium.⁸ (Note that this very weak ferromagnetism with a moment of the order of $0.01 \mu_B/U$ atom in the vicinity of the exact 1:1:1 stoichiometry has nothing in common with the canted ferromagnetism claimed for UCoAl in Ref. 4 with $\mu_U = 0.8 \mu_B$. In our case, at the U-poor boundary of the homogeneity range, the magnetic moment reaches only $0.1 \mu_B/U$ atom). A metamagnetic transition occurring from a low- to a high-moment ferromagnetic state has been also observed in 3*d*-band metamagnets, for example, in YCo₃,⁹ and a certain concentration range of the Lu (Co_{1-x}Al_x)₂ system.¹⁰

The critical parameters in the 3*d*-band metamagnets are very sensitive to external pressure.⁷ The motivation of the present work was to check pressure effects in UCoAl. We report on the results of magnetization studies of two representative single crystals from the homogeneity range, namely U_{0.9}Co_{1.05}Al_{1.05}, in which the weak ferromagnetism with μ/U atom = $0.05\mu_B$ was observed, and U_{1.1}Co_{0.95}Al_{0.95} having a purely paramagnetic ground state.

II. EXPERIMENTAL

The UCoAl-based alloys were prepared by melting the components (3N8 U; 4N Co and Al) with nominal compositions selected around 1:1:1 in an arc furnace with helium atmosphere. In order to increase the grain size, the ingots were remelted in a resistance furnace with a high-temperature gradient and cooled slowly through the melting point. The x-ray analysis confirmed the single-phase state of the alloys. Samples for the measurements were cut out of

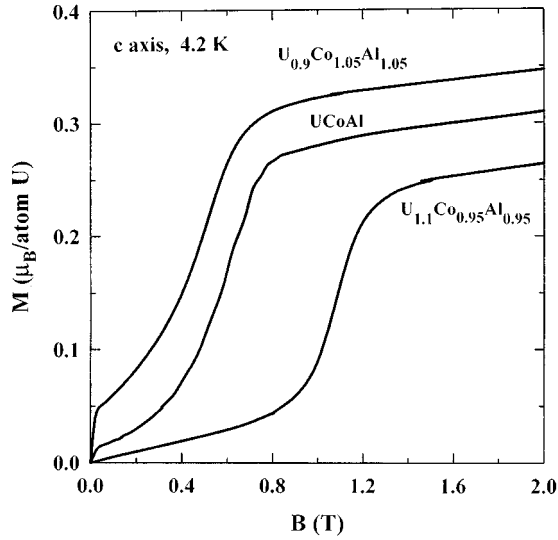


FIG. 1. Low-field magnetization curves of the stoichiometric and off-stoichiometric UCoAl single crystals in field applied along the c axis at 4.2 K under ambient pressure.

large (~ 3 mm) grains and polished on planes perpendicular to the $\langle 100 \rangle$, $\langle 210 \rangle$, and $\langle 001 \rangle$ axes.

The magnetization at ambient pressure was measured at 4.2 K by an induction method in pulsed field up to 40 T applied along the a and c axis, respectively. High-pressure measurements (up to 10.1 kbar) have been performed by an extraction-type magnetometer with steady magnetic fields up to 9 T, applied along the c axis. The high-pressure clamp cell was made of Cu-Ti alloy.

III. RESULTS AND DISCUSSION

Low-field magnetization (along the c axis) curves of single crystals of UCoAl and two representative off-stoichiometric compounds at 4.2 K in ambient pressure are shown in Fig. 1. The magnetization process of $U_{1.1}Co_{0.95}Al_{0.95}$ starts by a paramagnetic straight-line dependence with the susceptibility of $3.5 \times 10^{-8} \text{ m}^3/\text{mol}$. The metamagnetic transition occurs within the interval of 0.8–1.3 T yielding a moment $\mu = 0.25 \mu_B/\text{U}$ atom in the upper field. (Note that in this paper all values of magnetic moment are related to one U atom, not per formula unit due to noninteger indexes.) The magnetization curve of $U_{0.9}Co_{1.05}Al_{1.05}$ is considerably different and exhibits a spontaneous moment of $0.05 \mu_B/\text{U}$ atom and a much lower transition field. After saturating the ferromagnetic component, the moment starts to grow much faster than in $U_{1.1}Co_{0.95}Al_{0.95}$ mainly due to proximity of the metamagnetic transition which is completed already in 0.8 T where μ/U atom reaches $0.31 \mu_B$.

At 2 K, both crystals exhibit a noticeable (20 mT) magnetic hysteresis at the metamagnetic transition indicating its first-order character. Since the pressure measurements were performed at 4.2 K, where hysteresis becomes rather small (~ 5 mT, less than linewidth in Fig. 1), we neglect it in further consideration. As seen from Fig. 1, the stoichiometric single crystal exhibits intermediate behavior in all aspects which reflects monotonous development of magnetic properties with composition within the homogeneity range. Since

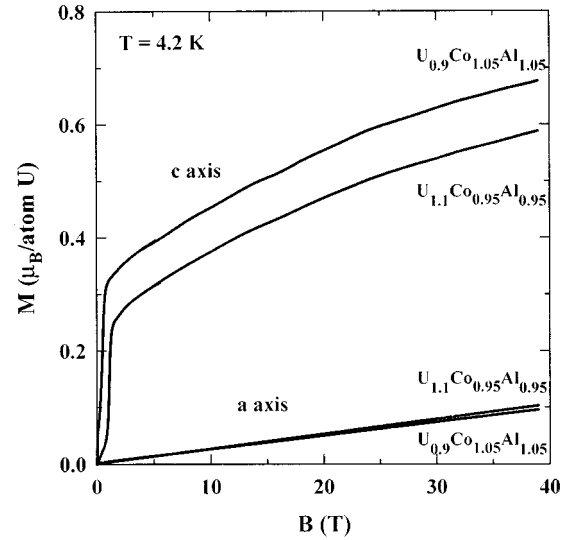


FIG. 2. High-field magnetization curves of the UCoAl-based single crystals in field applied along the c and a axes at 4.2 K under ambient pressure.

we present nominal composition of the samples we are aware that the true composition may be slightly deviated in each case. The certain uncertainty in the true composition amplified by strong composition sensitivity of the magnetization is most probably responsible for the scatter of values of the spontaneous moment over the range 0– $0.018 \mu_B$ found in different samples (two single crystals and several polycrystals) prepared with the nominal composition UCoAl. For further study, we have chosen the representative crystals $U_{1.1}Co_{0.95}Al_{0.95}$ with a purely paramagnetic ground state and $U_{0.9}Co_{1.05}Al_{1.05}$ with a relatively large spontaneous moment.

Figure 2 shows the high-field magnetization curves recorded on both crystals in ambient pressure at 4.2 K in magnetic field applied along the c and a axes. Considering that the positive offset in the c -axis magnetization of $U_{0.9}Co_{1.05}Al_{1.05}$ gained below 1 T, we can conclude that the high-field behavior of both samples is almost identical. Similar to the other izostructural UTX compounds, the huge uniaxial magnetocrystalline anisotropy (easy magnetization direction along the c axis) causes Ising-like magnetism. Above the metamagnetic transition the c -axis magnetization further grows showing only a slight saturation tendency. The magnetization in the basal plane is very weak ($\sim 0.1 \mu_B$ in 39 T) and linear with magnetic field. The anisotropy field B_a is definitely much larger than the highest field used in our experiment. The lower estimate of 120 T may be made from the hard-direction magnetization extrapolation. In this field we arrive at $0.3 \mu_B$, which is the typical magnetization gained through the metamagnetic transition.

Figure 3 displays the c -axis magnetization curves of the $U_{0.9}Co_{1.05}Al_{1.05}$ and $U_{1.1}Co_{0.95}Al_{0.95}$ single crystals under external pressure. In both cases, the pressure causes considerable increase of B_c and reduction of the moment gained ΔM across the metamagnetic transition. Hysteresis at the transition remains rather small with increasing pressure in both crystals. The spontaneous moment in $U_{0.9}Co_{1.05}Al_{1.05}$ vanishes already in 2.6 kbar.

In Fig. 4, the dM/dB vs B plots relevant to magnetization

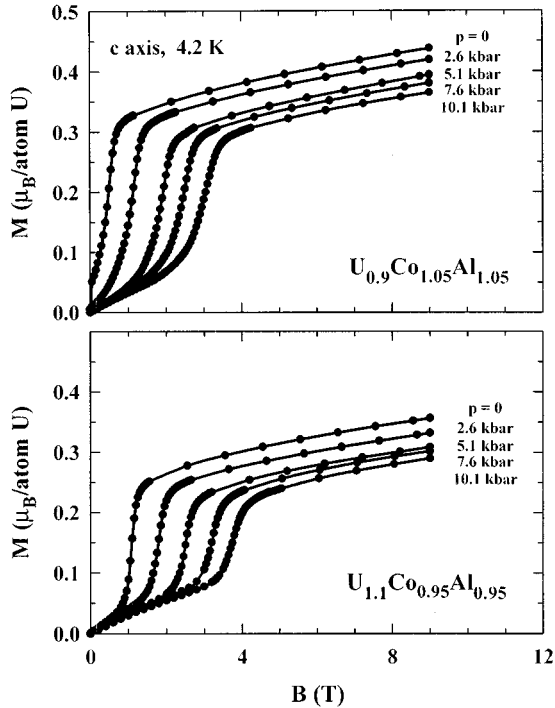


FIG. 3. Magnetization curves of the $U_{0.9}Co_{1.05}Al_{1.05}$ and $U_{1.1}Co_{0.95}Al_{0.95}$ single crystals in field applied along the c axis under varying pressure at 4.2 K.

curves in Fig. 3 are shown. B_c is the field in which the maximum in dM/dB occurs. Inspecting the dM/dB vs B curves, one can see clearly that the metamagnetic transition gradually broadens with increasing pressure, but still remains

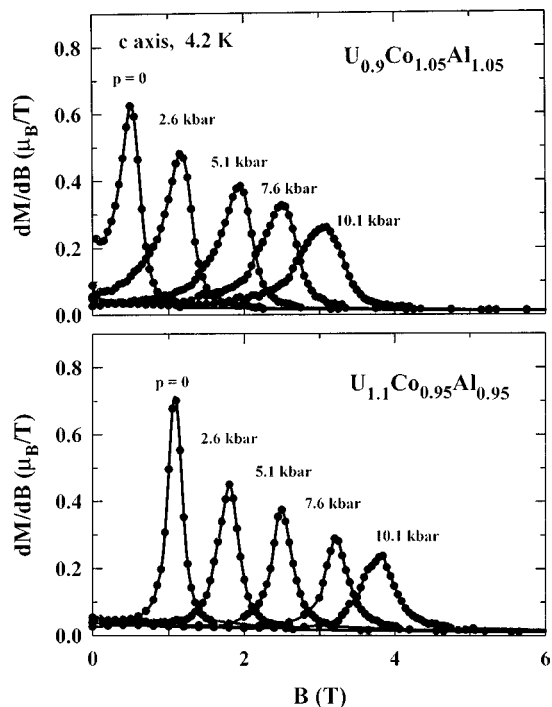


FIG. 4. Field dependences of differential susceptibility dM/dB of the $U_{0.9}Co_{1.05}Al_{1.05}$ and $U_{1.1}Co_{0.95}Al_{0.95}$ single crystals in field applied along the c axis under varying pressure at 4.2 K.

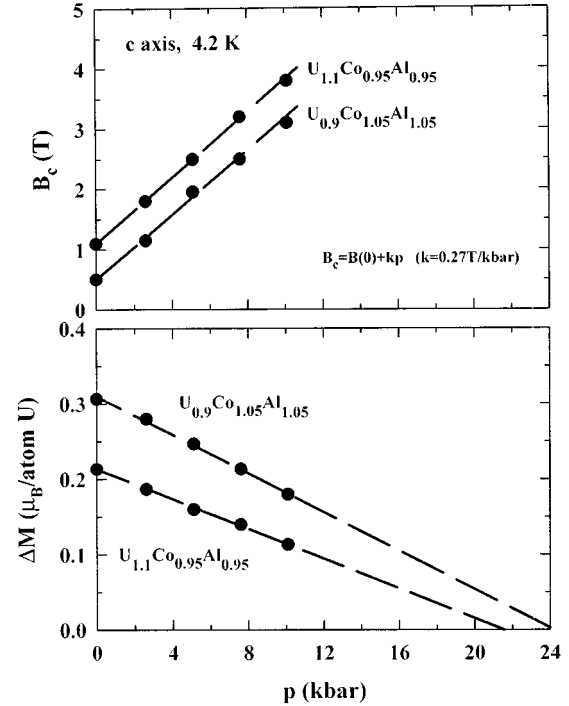


FIG. 5. Pressure dependences of the transition field B_c of the metamagnetic transition and the jump of the magnetic moment ΔM upon the metamagnetic transition in the $U_{1.1}Co_{0.95}Al_{0.95}$ and $U_{0.9}Co_{1.05}Al_{1.05}$ single crystals at 4.2 K.

pronounced enough for determination of B_c .

The pressure dependence of B_c and ΔM derived for both samples are presented in Fig. 5. Both quantities depend on pressure linearly. Moreover, the $B_c(p)$ dependence

$$B_c(p) = B(0) + k \cdot p \quad (1)$$

yields the same coefficient $k = dB_c/dp = 0.27$ T/kbar. $B(0)$ is 0.5 T in $U_{0.9}Co_{1.05}Al_{1.05}$ and 1.1 T in $U_{1.1}Co_{0.95}Al_{0.95}$. The linear extrapolation of the $\Delta M(p)$ dependences offers practically the same critical pressure (22–24 kbars) for metamagnetism in both materials.

There are several important aspects in which the 5*f*-band metamagnet UCoAl differs from the 3*d*-band metamagnets of the YCo₂- and LuCo₂-type. In particular, the 5*f* moment has a considerable orbital component in contrast to the negligible orbital magnetism in 3*d* systems. Consequently, magnetic phenomena in YCo₂ and LuCo₂ are more or less isotropic, but the huge uniaxial anisotropy of UCoAl prevents the metamagnetic behavior in the basal plane.

Another feature, which makes metamagnetism in UCoAl so outstanding is the extremely low value of B_c . The critical field in YCo₂ and LuCo₂ is almost two orders of magnitude higher, although suitable *p*-metal (Al, Ga) substitutions for Co may suppress this large value to the level comparable to UCoAl.^{7,10}

On the other hand, there are numerous common features of UCoAl and the 3*d*-band metamagnets suggesting certain universality in the band metamagnetism:

(1) Enhanced magnetic susceptibility with a broad maximum in the $\chi(T)$ dependence around the characteristic temperature T^* .

(2) Metamagnetic transition at low temperatures (below T^*).

(3) Comparable values of the moment jump at the transition, $\Delta M = 0.25\mu_B$ in UCoAl, $0.27\mu_B$ in YCo₂ (Ref. 11) and $0.44\mu_B$ in LuCo₂.¹²

(4) High susceptibility in fields far above the metamagnetic transition.

(5) B_c with increasing temperature satisfying the formula

$$B_c(T) = B(0) + a \cdot T^2 \quad (2)$$

with almost the same coefficient $a \cong 2 \times 10^{-3} \text{ T K}^2$, despite two orders of magnitude difference in B_c between Y(Lu)Co₂ (Ref. 7) and UCoAl.¹³

(6) Moderately enhanced values of the linear coefficient γ in the specific heat [$\gamma = 37 \text{ mJ mol}^{-1} \text{ K}^{-2}$ in YCo₂ (Ref. 7) and $70 \text{ mJ mol}^{-1} \text{ K}^{-2}$ in UCoAl (Ref. 2)].

(7) High sensitivity to alloying in both Y(Lu)Co₂ (Ref. 7) and UCoAl.¹⁴

As regards the magnetoelasticity, magnetization under pressure was studied in Lu(Co_{0.88}Ga_{0.12})₂.⁷ This is a ferromagnet at ambient pressure but the ferromagnetism is lost under pressure and metamagnetism appears with very large pressure effect on the critical field, $dB_c/dp = 1.0 \text{ T/kbar}$. In this respect, metamagnetism in UCoAl is less pressure dependent. Nevertheless, both compounds can be classified as very sensitive to the pressure.

The values of volume compressibility at room temperature for several UTX compounds with the ZrNiAl-type structure have been reported in Ref. 15. They fall in a narrow range $\kappa = (0.8 - 1.05) \times 10^{-2} \text{ kbar}^{-1}$. Velocities of longitudinal- and transverse-acoustic waves along the c axis were measured on a UCoAl single crystal at 4.2 K (Ref. 16) as $v_l = 4450 \text{ m/s}$, $v_t = 2610 \text{ m/s}$. Unfortunately, the data along one axis are not enough for proper determination of volume compressibility. However, using the formula for a polycrystal¹⁷

$$\kappa = (\rho v_l^2 - \frac{4}{3} \rho v_t^2)^{-1}, \quad (3)$$

(i.e., neglecting presumable anisotropy of the linear compressibility), we obtain $\kappa = 0.89 \times 10^{-3} \text{ kbar}^{-1}$ which is in good agreement with other UTX. Taking this κ value, the change of volume corresponding to the critical pressure may be estimated as $\Delta V/V = -2\%$.

In summary, we have studied low-temperature magnetization behavior of selected UCoAl-based single crystals in high magnetic fields (up to 40 T) and ambient pressure and pressure effects on magnetism in fields up to 9 T. The results are compared with typical $3d$ metamagnets $R\text{Co}_2$. There are pronounced differences between these two types of systems connected mainly with different character of $5f$ - and $3d$ -electron states in intermetallics. Both types of materials exhibit band metamagnetism. In UCoAl it is, however, confined only to one dimension (along the c axis) due to huge anisotropy typical for many systems with $5f$ moments. The much weaker spin-orbit interaction in $3d$ systems allows quenching of the orbital moment and the metamagnetism more or less isotropic. Also the critical-field values differ strikingly. B_c in UCoAl ($\sim 0.8 \text{ T}$) is almost two orders of magnitude lower than in $R\text{Co}_2$. Despite these striking differences a spectacular number of features which both the $5f$ and $3d$ metamagnets have in common (characteristic temperature dependence of the susceptibility, temperature dependence B_c , sensitivity to alloying and external pressure) suggest that the metamagnetism in both systems is basically of the same origin.

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