# Magnetic properties of the 5*f* itinerant electron metamagnet UCoAl under high pressure

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The magnetization and susceptibility of single-crystalline UCoAl were measured under high pressures to $P=1.2$ GPa. A sharp metamagnetic transition was observed only in magnetic fields along the $c$ axis. The critical field is $B_c=0.65$ T at $P=0$ GPa, which increases with pressure and temperature. The susceptibility for ambient pressure shows a broad maximum at $T_{\mathrm{max}}=20$ K. The value of $T_{\mathrm{max}}$ increases with pressure. A theory of the itinerant metamagnetic transition has been generalized for the case of anisotropic spin fluctuations. The observed pressure dependence of the inverse susceptibility at $T_{\mathrm{max}}$, the temperature $T_0$ for the disappearance of the metamagnetic transition, and the temperature dependence of $B_c$ can be explained with this theory. [S0163-1829(99)04409-4]

## I. INTRODUCTION

Itinerant electron metamagnetism is a first-order field-induced transition from the paramagnetic ground state to the ferromagnetic state in the itinerant electron system. This phenomenon has been observed up to now in various Co-based compounds such as $\mathrm{Co(S_{1-x}Se_x)_2}$ and $R\mathrm{Co_2}$ with $R=$Y, Lu.[1,2] The metamagnetic transition is considered to originate from a special shape of the density-of-states curve around the Fermi level.

The UCoAl compound belongs to a wide group of uranium intermetallics which crystallize in the hexagonal ZrNiAl-type structure. Andreev *et al.*[3] found that this compound shows a metamagnetic transition to a ferromagnetic state with a U magnetic moment of $0.3\mu_B$ in a weak magnetic field less than 1 T applied along the $c$ axis. The ground state of UCoAl is considered to be paramagnetic from the observation of the absence of anomalies in the temperature variations of the specific heat, the electrical resistivity,[4] and the lattice parameters $a$ and $c$.[5] Wulff *et al.* confirmed this state from polarized-neutron diffraction experiments.[6] Band structure calculations of UCoAl suggest that the observed transition is a 5*f* itinerant metamagnetic transition.[7] The UCoAl compound has very similar characteristics to typical 3*d* itinerant metamagnets: (1) The susceptibility is exchange enhanced and exhibits a broad maximum around a characteristic temperature $T_{\mathrm{max}}$, (2) the high-field susceptibility is large even in magnetic fields above the metamagnetic transition, (3) the critical field of the metamagnetic transition $B_c$ is proportional to $T^2$ at low temperatures, (4) the transition is very sensitive to external pressure[8] and alloying.[9,10]

However, some characteristics of UCoAl are quite different from those of 3*d* itinerant metamagnets. In $R\mathrm{Co_2}$ and $\mathrm{Co(S_{1-x}Se_x)_2}$, the 3*d* electrons of Co are responsible for the magnetic properties. In UCoAl, on the other hand, the 5*f* electrons of U contribute mainly to the magnetism, while 3*d* electrons of Co do not.[6] Although the metamagnetic phenomena in 3*d* itinerant metamagnets are isotropic,[2] the metamagnetic transition in UCoAl is observed only in magnetic fields along the $c$ axis. In the basal plane, UCoAl shows weak paramagnetism and no metamagnetic transition. Large uniaxial magnetic anisotropy is found in all the U*TX* compounds ($T=$late *d*-transition element, $X=p$ element) independent of the ground state. In the 3*d* itinerant metamagnets of the Laves phase, a strong correlation between $B_c$ and $T_{\mathrm{max}}$ is found.[11] In UCoAl-type metamagnets, however, such a correlation has not been established yet.

Spin fluctuations are considered to play an important role in the itinerant metamagnetic transition at finite temperatures. Several theoretical approaches taking into account the effects of spin fluctuations have been proposed to explain the observed itinerant metamagnetic transition.[12–15] The magnetic behavior observed in $\mathrm{Y(Co_{1-x}Al_x)_2}$ and $\mathrm{Co(S_{1-x}Se_x)_2}$ can be interpreted successfully using a theory of itinerant electron metamagnetism at finite temperatures.[1,2] However, it is not clear whether this theory is applicable to the metamagnetic transition in the 5*f* itinerant metamagnet UCoAl as well.

The magnetic properties of U*TX* compounds depends on the overlap of the 5*f* wave functions of neighboring U atoms and the hybridization of the 5*f* states of U with the *s*, *p*, and *d* valence states of ligands (*T* and *X* atoms). In UCoAl, the 5*f*-3*d* plus 5*f*-3*p* hybridization is believed to be too strong for the formation of U magnetic moment. Andreev *et al.*[16] examined the influence of the Ga substitution for Al on the magnetic properties of UCoAl. A stable ferromagnetic ground state is found to appear at a Ga concentration of more than 20%. The appearance of the ferromagnetism is considered to be due to the weakening of the 5*f*-*p* hybridization because the 5*f*-4*p* (Ga) hybridization is weaker than the 5*f*-3*p* (Al) hybridization. Recently, we measured the magnetization curves of $UCoAl_{1-x}Ga_x$ at $T=4.2$ K under high pressure up to 1.2 GPa. We observed that the application of high pressure suppresses the ferromagnetism and itinerant electron metamagnetism occurs in the concentration region $0.2\leqslant x \leqslant 0.3$. Details of the experimental results will be published elsewhere.[17]

The first study of the pressure effect on the magnetization process of UCoAl was performed at $T=4.2$ K with two single-crystalline samples in the homogeneity range $U_{0.9}Co_{1.05}Al_{1.05}$ and $U_{1.1}Co_{0.95}Al_{0.95}$, which only gives information about the metamagnetic properties of the ground state.[8] In order to obtain the information at finite temperatures, we have studied in detail the pressure effects on the susceptibility and the magnetization process of UCoAl in a wide temperature range using a high-quality single crystal. Since the metamagnetic phenomena of UCoAl are very anisotropic, we have proposed a theory of the metamagnetic transition in strongly anisotropic itinerant electron systems. The experimental results are discussed in terms of this theory.

## II. EXPERIMENTAL DETAILS

A single crystal of UCoAl was grown with the Czochralski method from the melt of stoichiometric amounts of the constituent elements (U of 99.95% purity, Co of 99.99% purity, and Al of 99.9999% purity) in a tetra-arc furnace. The x-ray powder diffraction of the top and bottom parts of the crystal indicates that the crystal is of a single phase with the ZrNiAl-type structure. The lattice parameters $a=667.5$ pm and $c=396.6$ pm are in good agreement with previous data.[5] Two samples cut from the top and bottom parts were found to have the same critical field of the metamagnetic transition $B_c$. This indicates that the single crystal is very homogeneous since the value of $B_c$ is very sensitive to its composition.[5] The residual resistance ratio of the crystal measured is $R(290\ \text{K})/R(4.2\ \text{K})=20$, which is larger than those of UCoAl single crystals grown previously by a modified Bridgman method $[R(290\ \text{K})/R(4.2\ \text{K})=3]$.[3–5,8] This result suggests that the single crystal used in this study has better quality. The shape of the sample for the magnetization measurements under pressure is nearly cubic with a size $\sim 1.8\ \text{mm}^3$ and a weight 63 mg.

The UCoAl sample was compressed in a Teflon capsule filled with a liquid pressure medium, mixture of two types of Fluorinert (FC 70:FC 77=1:1), in a nonmagnetic high-pressure clamp cell made of a Ti-Cu alloy. The pressure produced in the sample at low temperatures was calibrated by measuring the Meissner effect of Pb, for which the pressure dependence of the superconducting transition temperature is known to high accuracy. The maximum pressure was 1.2 GPa. The total magnetization of the sample and the surrounding high pressure cell was measured with an extraction-type magnetometer in magnetic fields up to 9 T produced by a superconducting magnet. Although the magnetization of the high pressure cell was extremely small, its value was subtracted from the total magnetization to obtain the precise value of the sample magnetization. Magnetic fields were applied only along the *c* axis of the sample. [In magnetic fields along the *c* plane, the sample behaves Pauli-paramagnetically with a susceptibility of 0.004 $(\mu_B/U)/T$ and no metamagnetic transition is observed in pulsed magnetic fields up to 42 T.]

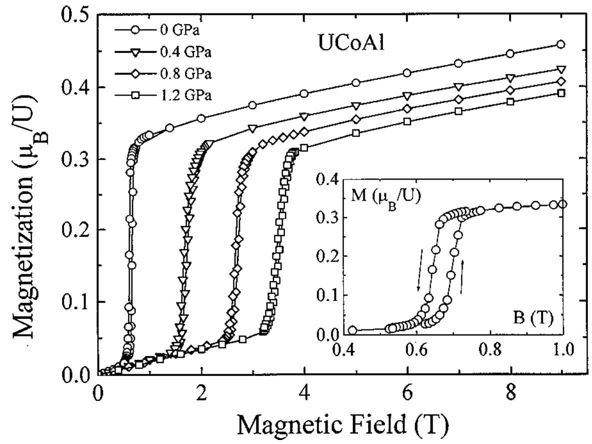


FIG. 1. Magnetization curves of single-crystalline UCoAl For $T=1.6$ K in magnetic fields along the *c* axis at various pressures. The inset shows the magnetization curve on a finer magnetic field scale.

## III. EXPERIMENTAL RESULTS

In order to examine the magnetic properties of the ground state of stoichiometric UCoAl, we measured the magnetization curves along the *c* axis at T=1.6 K for various pressures. The results are shown in Fig. 1. The ground state is paramagnetic in zero field. The metamagnetic transition is found to be much sharper than the previous results.[3–5,8] This indicates that the crystal has better quality. As shown in the inset, the magnetization curve has hysteresis around the transition. This shows that the metamagnetic transition is first order. The critical field of the transition is defined as the field at which the field derivative of magnetization $dM/dB$ becomes maximum. The critical field increases with pressure, in agreement with previous data.[8]

The average critical field $B_c$ for $T=1.6$ K is plotted as a function of pressure in Fig. 2. The value of $B_c$ increases nearly linearly for $P<1$ GPa and is written as

$$B_c(P)=B_c(0)+kP, \tag{1}$$

where $B_c(0)$ is the critical field at ambient pressure. The coefficient $k$ is given by 2.62 T/GPa at $T=1.6$ K. This value is very close to that of 2.7 T/GPa for $U_{0.9}C_{1.05}Al_{1.05}$ determined at $T=4.2$ K.[8]

The temperature dependences of the magnetization curve of UCoAl at ambient pressure and at $P=1.2$ GPa are shown in Fig. 3. The observed metamagnetic transition is very sharp at low temperatures, the slope $dM/dB$ is close to the demagnetization factor of the sample. With increasing temperature, the transition broadens slowly for $T<10$ K and rapidly for $T>10$ K.

The difference between the critical fields determined at ambient pressure in increasing and decreasing fields $\Delta B_c$ is

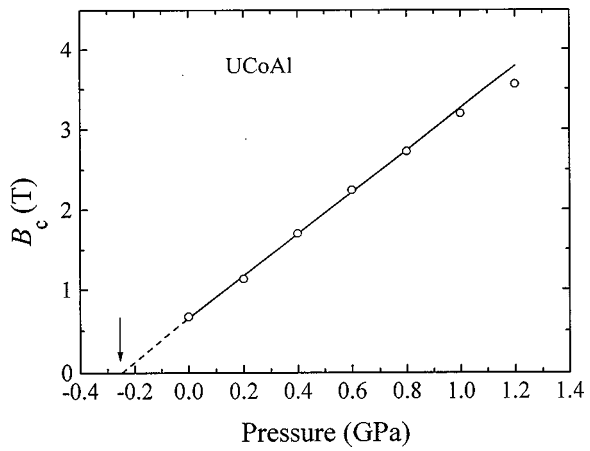


FIG. 2. Pressure dependence of the critical field of the metamagnetic transition $B_c$ at $T=1.6$ K. The solid line represents a linear fitting in the pressure range 0–0.8 GPa. The arrow indicates the critical pressure for the onset of ferromagnetism.

plotted as a function of temperature in Fig. 4(a). The value of $\Delta B_c$ decreases with increasing temperature and vanishes at a critical temperature $T_0$. This temperature is considered as the temperature at which the first-order metamagnetic transition disappears. Figure 4(b) shows the value of $dM/dB$ at the critical field as a function of temperature at ambient pressure. Sharp decrease of $dM/dB$ reflects the broadening of the metamagnetic transition around the temperature where $\Delta B_c$ becomes zero.

We measured the temperature dependence of the magnetization curve at various pressures and determined the critical temperature $T_0$ as a function of pressure to make the magnetic phase diagram in the $P$-$T$ plane. Figure 5 shows the phase diagram. The UCoAl compound is paramagnetic in the entire region and the metamagnetic transition appears in the region below the $T_0$ line. The value of $T_0$ decreases slightly with increasing pressure.

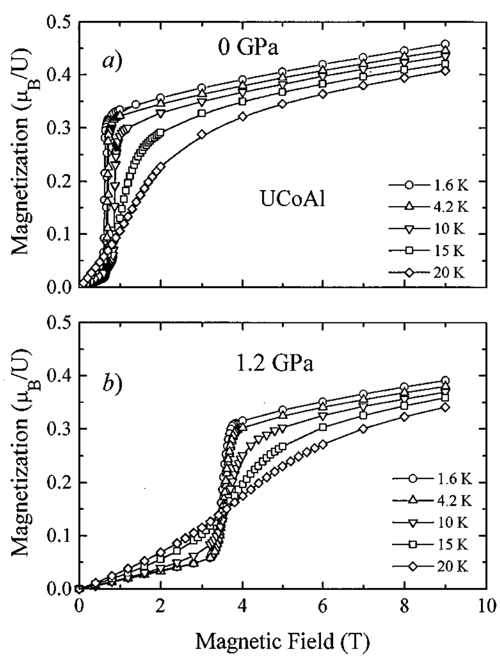


FIG. 3. Magnetization curves of UCoAl for ambient pressure (a) and for $P=1.2$ GPa (b) at various temperatures.

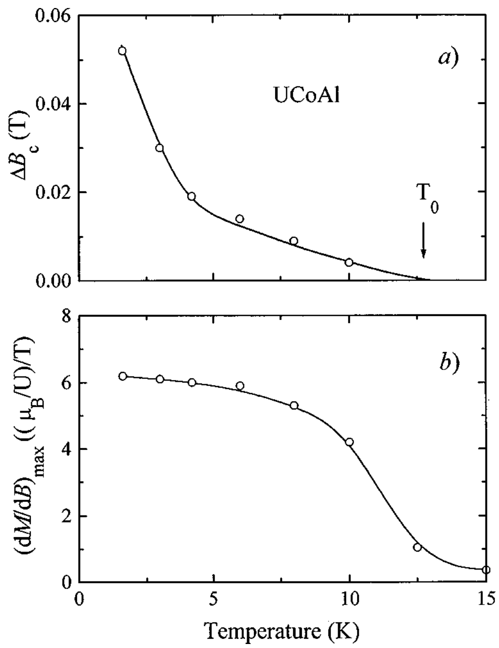


FIG. 4. Width of hysteresis of the metamagnetic transition $\Delta B_c$ (a) and the slope of the magnetization curve $dM/dB$ at $B=B_c$ (b) as functions of temperature at ambient pressure. Lines are guides for the eyes.

The average critical field $B_c$ of UCoAl is plotted as a function of the square of temperature for different pressures in Fig. 6. The critical field increases nearly linearly with $T^2$ for all the pressures. Such behavior of $B_c$ has already been found in Co-based itinerant metamagnets, Laves phase compounds,[2,18,19] and $Co(S_{1-x}Se_x)_2$.[1] In polycrystalline samples of $UCo_{1-x}Ni_xAl$ solid solutions,[9,10] this behavior is also observed. However, the value of $B_c$ determined for $UCo_{1-x}Ni_xAl$ is not accurate because the metamagnetic transition becomes very broad in these polycrystals.

The temperature dependence of the susceptibility $\chi$ measured at $B=0.02$ T is shown for various pressures in Fig. 7. With increasing temperature, the susceptibility for all the pressures increases, reaches a maximum value at $T=T_{max}$ and then decreases. The temperature dependence of $\chi$ is very similar to that of Co-based itinerant metamagnets.[2] The sus-

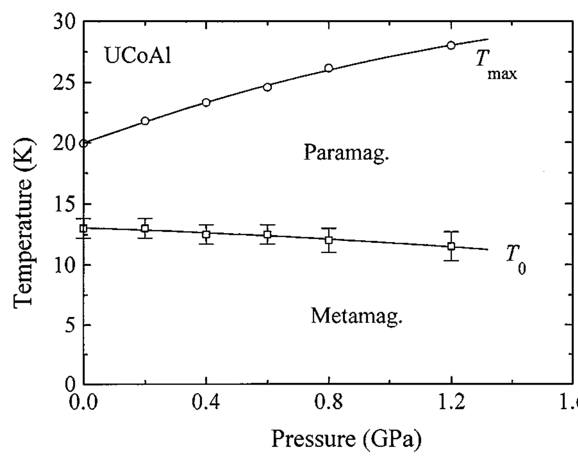


FIG. 5. Magnetic phase diagram of UCoAl in the $P$-$T$ plane. The metamagnetic transition appears only below the $T_0$ line.

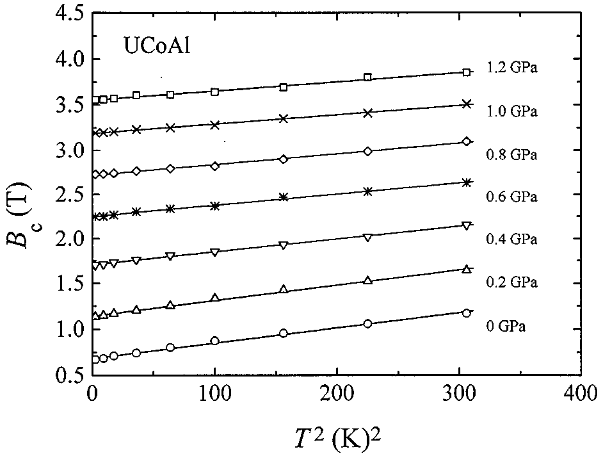


FIG. 6. Average critical field $B_c$ of UCoAl as a function of squared temperature for various pressures.

ceptibility maximum is remarkably suppressed in UCoAl by the application of pressure. This behavior is also found in the Laves phase compounds $Lu(Co_{1-x}Ga_x)_2$.[2] The value of $T_{max}$ is plotted as a function of pressure on the magnetic phase diagram shown in Fig. 5. The temperature $T_{max}$ increases gradually with pressure.

Sakakibara *et al.*[11] found a strong correlation between the values of $B_c$ for $T=0$ K and $T_{max}$ in various kinds of itinerant metamagnets including heavy fermion systems. Rather good proportionality holds between $B_c$ and $T_{max}$ for $M(Co,Al)_2$ ($M=$Y, Lu) with $B_c/T_{max}=(0.25\pm0.05)$ T/K. Figure 8 shows the value of $B_c$ for UCoAl observed at various pressures as a function of $T_{max}$. There exists a linear relation between $B_c$ and $T_{max}$. The estimated slope $dB_c/dT_{max}=0.36$ T/K is rather close to that for Co-based metamagnets.[11]

## IV. DISCUSSION

### A. Analyses of the magnetization curves

First, we try to analyze the magnetization process quantitatively since the metamagnetic transition observed in the high-quality single-crystalline sample of UCoAl is very

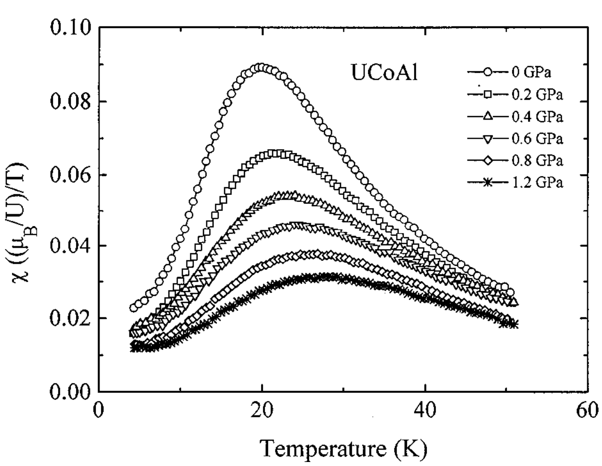


FIG. 7. Temperature dependence of the susceptibility $\chi$ of UCoAl along the $c$ axis measured in a magnetic field of 0.02 T at different pressures.

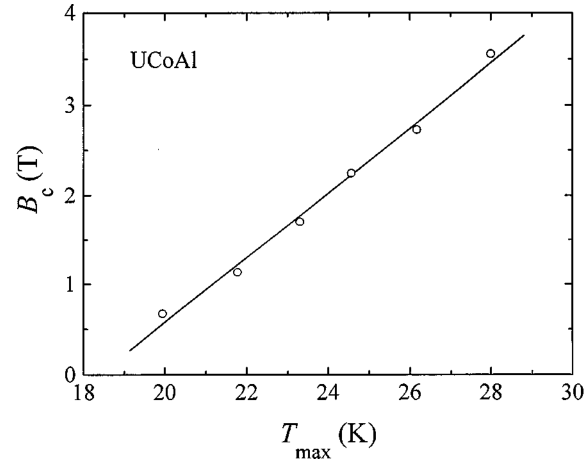


FIG. 8. Critical field of the metamagnetic transition $B_c$ at 1.6 K as a function of the temperature of the susceptibility maximum $T_{max}$ at various pressures.

sharp and shifts systematically with pressure and temperature. The magnetization curve for an itinerant metamagnet can be described in the ground state by the following magnetic equation of state:

$$B=a_0M+b_0M^3+c_0M^5, \tag{2}$$

where $B$ is magnetic field, $M$ is the uniform magnetization, and $a_0$ is the inverse susceptibility at $T=0$ K, $a_0=\chi(0)^{-1}$. The coefficients $a_0$, $b_0$, and $c_0$ are functions of the electronic density of states and its derivatives at the Fermi level. Here, we express $M$ and $\chi$ in terms of $\mu_B/U$ ($=\mu_B$/f.u.) and $(\mu_B/U)/T$, respectively. The metamagnetic transition appears under the condition

$$a_0>0, \quad b_0<0, \quad c_0>0, \quad \text{and} \quad 3/16<a_0c_0/b_0^2<9/20, \tag{3}$$

where $M(B)$ in Eq. (2) becomes a triple-valued function. In the equilibrium condition, the metamagnetic transition occurs at the critical field $B_c$, where the free energies of two states, that is the paramagnetic and ferromagnetic states, are equal to each other. It should be noted that in UCoAl the metamagnetic transition occurs only in magnetic fields along the $c$ axis. Therefore, we consider that $B$ and $M$ in Eq. (2) are the $z$ component of magnetic field and that of the magnetization, respectively. In principle, we can estimate the values of $a_0$, $b_0$, and $c_0$ by fitting the curve calculated from Eq. (2) to the measured magnetization curve. However, since the high-field susceptibility of UCoAl is large and the magnetization has no tendency to saturate after the metamagnetic transition, we must take into account an additional paramagnetic contribution. This contribution may originate from the polarization of conducting electrons or quantum spin fluctuations. The estimated value of the additional paramagnetic susceptibility is $\chi_0=0.011$ $(\mu_B/U)/T$, independent of temperature and pressure. This value is in reasonable agreement with the high-field susceptibility of paramagnetic $UCo_{0.9}T_{0.1}Al$ ($T=$Ni or Pd), which shows no metamagnetic transition.[20] Since the coefficient $a_0$ in Eq. (2) is the inverse susceptibility at $T=0$ K, we can directly estimate the value of $a_0$ from the initial slope of the magnetization curve and the value of $\chi_0$:

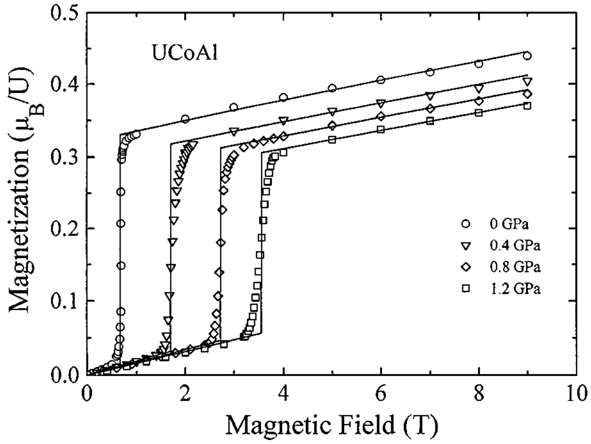


FIG. 9. Magnetization curves of UCoAl for different pressures at 1.6 K (open symbols) together with the best fitted curves using Eq. (2) (solid lines).

$$a_0=(\chi_{\rm exp}-\chi_0)^{-1}, \tag{4}$$

where $\chi_{\rm exp}$ is the susceptibility determined from the initial slope at 1.6 K. We selected the best fitted magnetization curve by changing the values of the two parameters $b_0$ and $c_0$. In this process, we made the calculated critical field equal to the average one determined experimentally $B_c$. Figure 9 shows the calculated magnetization curves for various pressures. They reproduce well the experimental magnetization curves of UCoAl. The values of the coefficient $a_0$, $b_0$, and $c_0$ are plotted as a function of pressure in Fig. 10.

As already described, the metamagnetic transition appears under the condition of Eq. (3) in the ground state. A ferromagnetic state appears for $a_0>0$, $b_0<0$, $c_0>0$, and $a_0c_0/b_0^2\leqslant 3/16$ although the Stoner condition ($a_0<0$) is not satisfied. Using the values of $a_0c_0/b_0^2$ for $T=1.6$ K evaluated at various pressures, we can estimate the critical pressure for the onset of ferromagnetism from the condition $a_0c_0/b_0^2=3/16$. The estimated critical pressure is $P_c=-0.25$ GPa (Fig. 10). The critical pressure can be evaluated also from the pressure dependence of the average critical field for the metamagnetic transition $B_c$ at $T=1.6$ K. The value of $B_c$ decreases with decreasing pressure and becomes zero at $P=-0.25$ GPa, as shown in Fig. 2 [and Eq. (1)]. This suggests that the critical pressure is $P_c=-0.25$ GPa. The increase in the transition field originates from the increase in the $5f$-$3d$ hybridization due to the application of high pressure. The values of the volume compressibility of several U*TX* compounds are reported to be $\kappa=(0.8-1.05)\times 10^{-2}$/GPa at room temperature.[21] Therefore, if the volume of UCoAl is increased 0.25%, this compound will become ferromagnetic. Recently, we made solid solutions of $U_{1-x}Y_xCoAl$ and measured the magnetic properties.[22] We found that the ferromagnetic state appears at $x=0.06$. The substitution of nonmagnetic Y for U leads to the volume expansion with a rate of 4.5%/$x$, indicating that the volume expansion is 0.3% at the appearance of ferromagnetism. This is consistent with the estimated volume of UCoAl at the onset of ferromagnetism in this study.

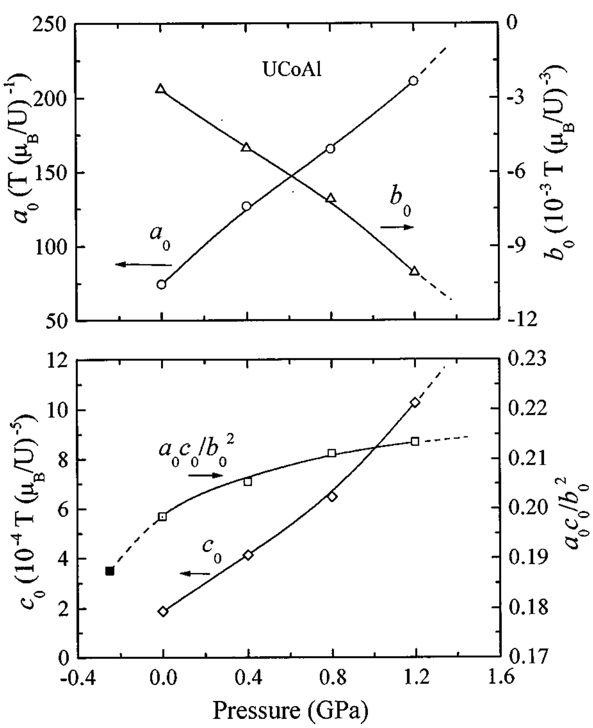


FIG. 10. Pressure variations of the Landau expansion coefficients $a_0$, $b_0$, $c_0$, and the value of $a_0c_0/b_0^2$ at 1.6 K. The closed square corresponds to the onset of ferromagnetism (see Fig. 2).

At the "upper" critical pressure for the disappearance of the metamagnetic transition, the condition $a_0c_0/b_0^2=9/20$ is satisfied. Because the pressure dependence of $a_0c_0/b_0^2$ shown in Fig. 10 is essentially nonlinear and the value of $a_0c_0/b^2$ tends to saturate at high pressures, the estimation of the critical pressure is very difficult. The real critical pressure will be considerably high. The magnetization jump $\Delta M$ at the transition decreases gradually with pressure (Fig. 1). From the extrapolation of the value of $\Delta M(P)$ to $\Delta M=0$, we can roughly evaluate the critical pressure to be $P=7$ GPa. This value is considerably larger than 2.4 GPa reported previously,[8] indicating a large uncertainty in such an estimation. Nevertheless, just 10% substitution of Ni for Co in UCoAl suppresses the metamagnetic transition in spite of the lattice expansion.[22] The suppression of the transition originates from the change in the strong $5f$-$3d$ hybridization produced by the substitution of Ni.

Next we analyze the magnetization curves for ambient pressure measured at different temperatures between 1.6 and 10 K. At a finite temperature $T$, the coefficients $a_0$, $b_0$, and $c_0$ in Eq. (3) are changed into $a(T)$, $b(T)$, and $c(T)$. We performed the same fitting procedure to estimate the values of $a(T)$, $b(t)$, and $c(T)$. Figure 11 shows the values as a function of temperature. The absolute values of all the coefficients decrease with increasing temperature. At the critical temperature for the disappearance of the metamagnetic transition $T=T_0$, the relation $a(T)c(T)/b(T)^2=9/20$ is satisfied. The value of $a(T)c(T)/b(T)^2$ for ambient pressure increases with temperature (Fig. 11) and can be fitted well with an exponential function. The extrapolation of the function gives the value of $T_0$, $T_0\cong 16$ K. This value is consistent with the critical temperature evaluated from the disappearance of hysteresis of the magnetization curve ($T_0\cong 13$ K, as shown in Fig. 4.

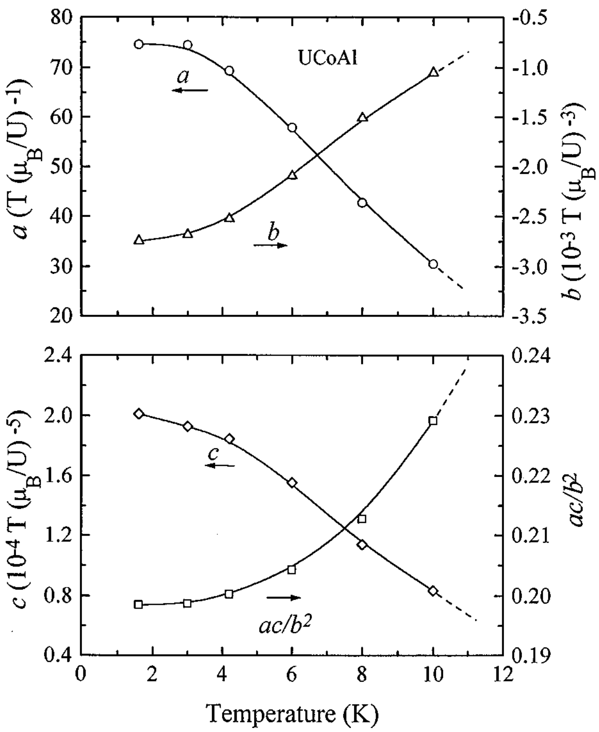


FIG. 11. Temperature dependence of the Landau expansion coefficients $a(T)$, $b(T)$, $c(T)$, and $a(T)b(T)/c(T)^2$ at ambient pressure.

### B. Theory of the itinerant metamagnetic transition and its comparison with the experimental results

A theory of the itinerant metamagnetic transition based on the spin-fluctuation model has been developed for isotropic magnetic systems by Yamada.[15] However, the metamagnetic transition observed in UCoAl occurs only in magnetic fields along the $c$ axis. Therefore, we generalize this theory to the case of uniaxial anisotropic systems. At a finite temperature $T$, the coefficients $a_0$, $b_0$, and $c_0$ in Eq. (12) are renormalized by thermal spin fluctuations and the magnetic equation of state is given by

$$B=a(T)M+b(T)M^3+c(T)M^5, \tag{5}$$

where the coefficients $a(T)$, $b(T)$, and $c(T)$ are functions of $a_0$, $b_0$, $c_0$, and the thermal average of fluctuating magnetic moment $\langle(\delta m_i)^2\rangle$.[15] The suffix $i$ means the component of the moment. The thermal average is a function of temperature and depends on the component. We introduce $Q(T)$ and $\alpha$ defined by

$$\langle(\delta m_\parallel)^2\rangle=Q(T), \tag{6}$$

$$\langle(\delta m_\perp)^2\rangle=\alpha Q(T), \tag{7}$$

where $\alpha$ is the parameter to represent the uniaxial anisotropy of the spin fluctuations ($0\leqslant\alpha\leqslant1$). Considering the anisotropic fluctuations given by Eqs. (6) and (7), the expressions for $a(T)$, $b(T)$, and $c(T)$ given in Ref. 15 can be rewritten as

$$a(T)=a_0+b_0(3+2\alpha)Q(T)+c_0(15+12\alpha+8\alpha^2)Q(T)^2, \tag{8}$$

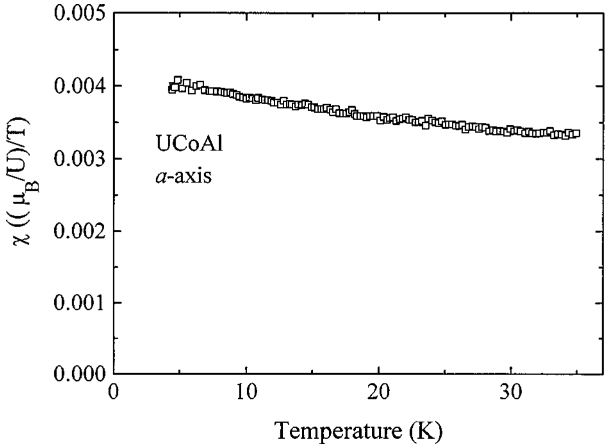


FIG. 12. Temperature dependence of the susceptibility of UCoAl along the $a$ axis.

$$b(T)=b_0+2c_0(5+2\alpha)Q(T), \tag{9}$$

$$c(T)=c_0. \tag{10}$$

The temperature dependence of the inverse paramagnetic susceptibility is given by

$$\chi(T)^{-1}=a(T)=a_0+b_0(3+2\alpha)Q(T)+c_0(15+12\alpha+8\alpha^2)Q(T)^2. \tag{11}$$

Under the condition $a_0>0$, $b_0<0$, $c_0>0$, and $a_0c_0/b_0^2>(3+2\alpha)^2/[4(15+12\alpha+8\alpha^2)]$, the susceptibility $\chi(T)$ increases and then decreases through a broad maximum at a characteristic temperature $T_{\max}$ with increasing temperature. If the magnetic system exhibits the metamagnetic transition, this condition is satisfied. The UCoAl compound shows the transition in the pressure region $0\leqslant P\leqslant1.2$ GPa, where the susceptibility has a broad maximum (Fig. 7), consistent with the theoretical result. The maximum of $\chi(T)$ is given by the relation $\partial\chi(T)^{-1}/\partial Q(T)=0$ and we get

$$Q(T_{\max})=\frac{|b_0|}{2c_0}\frac{3+2\alpha}{15+12\alpha+8\alpha^2}, \tag{12}$$

$$\chi(T_{\max})^{-1}=a(T_{\max})=a_0-\frac{b_0^2}{4c_0}\frac{(3+2\alpha)^2}{15+12\alpha+8\alpha^2}. \tag{13}$$

$$B(T_{\max})=|b_0|\frac{4\alpha(1-\alpha)}{15+12\alpha+8\alpha^2}. \tag{14}$$

In order to estimate the anisotropy parameter $\alpha$ for UCoAl, we measured the temperature dependence of the susceptibility along the $a$ axis at ambient pressure, as shown in Fig. 12. The susceptibility in the basal plane is smaller than that along the $c$ axis and decreases gradually with increasing temperature. Moreover, the magnetization increases linearly with increasing magnetic field and no metamagnetic transition occurs in magnetic fields up to 42 T. These results indicate that the spin fluctuations in UCoAl are very anisotropic and the anisotropy parameter $\alpha$ is nearly zero. Therefore, we assume $\alpha=0$ for UCoAl.

The mean-square amplitude of thermally fluctuating moment is given by

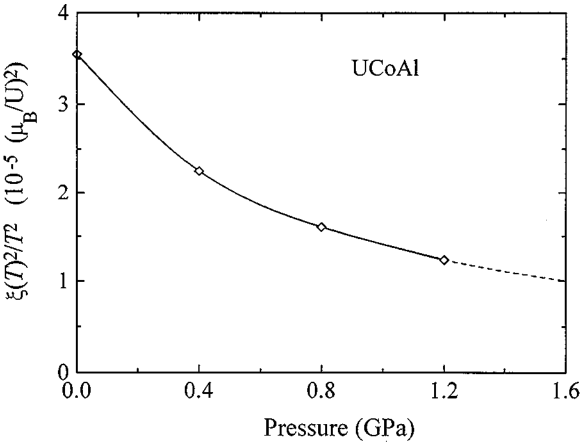


FIG. 13. Pressure dependence of the mean square amplitude of fluctuating moment $\xi(T)^2$ divided by $T^2$ estimated for UCoAl.

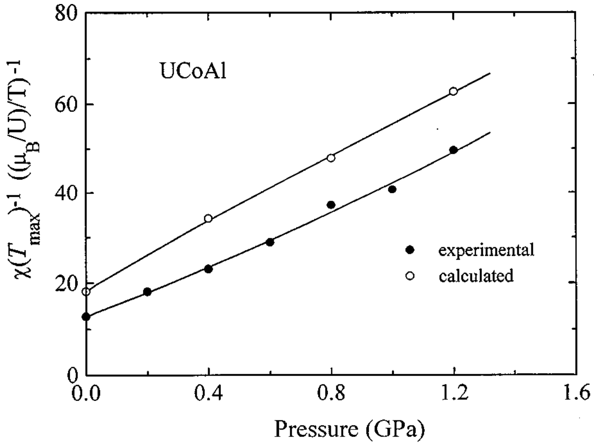


FIG. 14. Value of the inverse susceptibility of UCoAl for $T = T_{max}$ as a function of external pressure. The closed circles indicate the experimental data and the open circles the values calculated with the coefficients $a_0$, $b_0$, and $c_0$ derived from the magnetization curves for 1.6 K.

$$\xi(T)^2 = \langle(\delta m_\parallel)^2\rangle + 2\langle(\delta m_\perp)^2\rangle = (1+2\alpha)Q(T). \quad (15)$$

Using Eq. (12), we can evaluate the thermal spin fluctuation at $T=T_{max}$. In the case of strongly anisotropic systems ($\alpha=0$), it is given by

$$\xi(T_{max})^2 = -\frac{1}{10}\frac{b_0}{c_0}. \quad (16)$$

At low temperatures $\xi(T)^2$ is proportional to $T^2$.[15] We assume that this proportionality is valid up to $T=T_{max}$ in UCoAl. Figure 13 shows the pressure dependence of $\xi(T)^2/T^2$ $[=\xi(T_{max})^2/T_{max}^2]$ obtained for UCoAl with Eq. (16) and the experimental values of $T_{max}$. The pressure reduces the spin fluctuation in UCoAl. At $P=0$ GPa, the value of $\xi(T)^2/T^2$ is $3.5\times10^{-5}$ $[(\mu_B/U)^2]$. This value is about 6 times larger than that estimated at the same temperature for $Co(S_{1-x}Se_x)_2$, in which both the temperatures $T_{max}$ and $T_0$ are much higher.[1] At $T=15$ K, the average amplitude of the thermally fluctuating moment reaches about $0.1\mu_B/U$ in UCoAl.

According to Eq. (13), $\chi^{-1}(T_{max})$ can be estimated with the value of $a_0$, $b_0$, and $c_0$ determined from the magnetization curve at $T=1.6$ K. Figure 14 shows the calculated values of $\chi^{-1}(T_{max})$ for various pressures, which are compared with the experimental ones $\{=[\chi_{exp}(T_{max})-\chi_0]^{-1}\}$ for UCoAl. The experimental and theoretical values are consistent with each other. This gives an evidence of the applicability of the spin-fluctuation theory to the strongly anisotropic itinerant metamagnet UCoAl.

Equation (14) gives $b(T_{max})=0$ for $\alpha=0$. Since $Q(T)$ is an increasing function of temperature, $b_0$ is negative and $c_0$ is positive, $b(T)$ is negative at low temperatures, increases with temperature as seen from Eq. (9) and becomes zero at $T=T_{max}$. The value of $b(T)$ estimated from the magnetization curve of UCoAl at ambient pressure is negative at low temperatures and increases with temperature (Fig. 11). The value seems to become zero around the temperature at which the susceptibility becomes maximum ($T_{max}=20$ K for UCoAl), in good agreement with theory. Although the coefficient $c(T)$ is temperature independent in theory, we found that the $c(T)$ of UCoAl has a strong temperature dependence (Fig. 11). In the analysis of the magnetization curve, we assumed implicitly that the higher-order terms than the fifth-order term $c(T)M^5$ are negligibly small in the magnetic equation of state of UCoAl [see Eq. (5)]. The observed temperature dependence suggests that the higher-order terms are not negligible. [If these terms are present, the $c(T)$ estimated using Eq. (5) is affected by them and shows a temperature dependence.]

The condition for the appearance of the metamagnetic transition at finite temperatures can be obtained by replacing $a_0$, $b_0$, and $c_0$ in Eq. (3) with $a(T)$, $b(T)$, and $c(T)$. If $a(T)c(T)/b(T)^2$ is smaller than $\frac{3}{16}$, the system becomes ferromagnetic. If $a(T)c(T)/b(T)^2$ is larger than $\frac{9}{20}$, on the other hand, no metamagnetic transition occurs. From these relations and Eqs. (9)–(11), we get the temperature for the disappearance of the metamagnetic transition $T_0$ for strongly anisotropic spin fluctuations ($\alpha=0$)

$$T_0^2 = T_{max}^2\left(1-\sqrt{\frac{10}{3}}\sqrt{\frac{a_0c_0}{b_0^2}-\frac{3}{20}}\right). \quad (17)$$

The ferromagnetic state appearing in the vicinity of $a(T)c(T)/b(T)^2=\frac{3}{16}$ becomes unstable at the critical temperature $T_1$,

$$T_1^2 = T_{max}^2\left(1-\sqrt{\frac{80}{3}}\sqrt{\frac{a_0c_0}{b_0^2}-\frac{3}{20}}\right) \quad (18)$$

for $\alpha=0$. The paramagnetic-to-ferromagnetic transition at $T=T_1$ is first order. The temperature $T_0$ can be evaluated from Eq. (17) using the values of $T_{max}$ and $a_0c_0/b_0^2$. The evaluated value is $T_0=15.4$ K at ambient pressure and increases to $T_0=20$ K at $P=1.2$ GPa. These values are slightly larger than the experimental values of $T_0\cong13$ K determined from the extrapolation of the value of $\Delta B_c(T)$ to $\Delta B_c(T)=0$ (Figs. 4 and 5), but very close to the estimation obtained from the extrapolation of the values of $ac/b^2$ to $ac/b^2=\frac{9}{20}$ (Fig. 11). Equation (18) shows that the ferromagnetic state is unstable in UCoAl even in the ground state, in agreement with the experimental fact that UCoAl is a paramagnet.

If the critical field of the metamagnetic transition is very low, the magnetization in the paramagnetic state $M_1$ and that in ferromagnetic state $M_2$ are given from Eq. (5) by

$$M_1 = B_c/a(T), \tag{19}$$

$$M_2 = M_0 + B_c/\{a(T) + 3b(T)M_0^3 + 5c(T)M_0^5\}, \tag{20}$$

$$M_0 = \left[\frac{|b(T)|}{2c(T)}\left\{1 + \sqrt{1 - 4\frac{a(T)c(T)}{b(T)^2}}\right\}\right] \tag{21}$$

at the critical field. Using the Maxwell relation

$$\int_{M_1}^{M_2} B dM = B_c(M_2 - M_1), \tag{22}$$

we obtain the relation for the critical field

$$B_c \cong \frac{M_0}{3}\left\{a(T) - \frac{b(T)^2}{8c(T)}\left[1 + \sqrt{1 - 4a(T)c(T)/b(T)^2}\right]\right\}. \tag{23}$$

Substituting Eqs. (8)–(10) into Eq. (23), we get

$$B_c \cong \frac{3}{4}\sqrt{\frac{|b_0|}{3c_0}}\left\{a_0 - \frac{3}{16}\frac{b_0^2}{c_0} + \frac{1}{4}(3 - 2\alpha)|b_0|Q(T)\right\} \tag{24}$$

in the first order approximation of $Q(T)$. Since $Q(T)$ is proportional to $T^2$ at low temperature and the coefficient of $Q(T)$ is positive, the value of $B_c(T)$ increases with temperature as $T^2$, in agreement with the experiment results, as shown in Fig. 6.

Finally, we discuss the change in the $\gamma$ value of the electronic specific heat due to the metamagnetic transition. The specific heat measurements of polycrystalline UCoAl indicate that the $\gamma$-value changes from 70 $\mathrm{mJ\,mol^{-1}\,K^{-2}}$ in zero field to 62 $\mathrm{mJ\,mol^{-1}\,K^{-2}}$ at 5 T.[4] We try to estimate the reduction of the value from the temperature dependence of the critical field and the change of the magnetization at the critical field $\Delta M$ using the relation[2]

$$\Delta\gamma = -2\beta\Delta M, \tag{25}$$

where the coefficient $\beta$ is defined by

$$B_c(T) = B_c(0) + \beta T^2. \tag{26}$$

This estimation gives $\Delta\gamma = -5.5\ \mathrm{mol^{-1}\,K^{-2}}$ at ambient pressure, consistent with the specific heat measurements. We found that the absolute value of $\Delta\gamma$ decreases nearly linear with increasing pressure. At $P = 1.2$ GPa, we have $\Delta\gamma = -2.7\ \mathrm{mJ\,mol^{-1}\,K^{-2}}$. These results also suggest that the spin fluctuation in UCoAl are suppressed by the application of high pressure. The reduction of the $\gamma$ value is only less than 10% and is much smaller than that in the typical 3*d* itinerant metamagnet $YCo_2$.[2]

## V. CONCLUSION

The UCoAl compound has the paramagnetic ground state and exhibits a metamagnetic transition to the ferromagnetic state with a magnetic moment $M = 0.3\mu_B/U$ at a critical field of $B_c = 0.65$ T applied along the $c$ axis. We determined the magnetic phase diagram in the $P$-$T$ plane. The metamagnetic transition for ambient pressure disappears at a critical temperature $T_0 = 13$ K. The value of $T_0$ decreases slightly with increasing pressure. The application of high pressure increases the critical field with a rate 2.6 T/GPa in the ground state. The temperature dependence of the susceptibility shows a broad maximum at a temperature $T_{max} = 20$ K at ambient pressure. The temperature $T_{max}$ increases with pressure. The critical field of UCoAl has a linear relation with $T_{max}$, which is very similar to that found in 3*d* itinerant metamagnetic systems.[11] By fitting the magnetization curves, we have determined the phenomenological Landau expansion coefficients for different pressures and temperatures. We have analyzed the experimental data using a theory of the itinerant metamagnetic transition generalized for the case of anisotropic thermal spin fluctuations. The observed pressure dependence of the inverse susceptibility at $T_{max}$, the temperature $T_0$ for the disappearance of the metamagnetic transition and the temperature dependence of $B_c$ can be explained by this theory with the Landau expansion coefficients determined.

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