

Unusual Upper Critical Field of the Ferromagnetic Superconductor UCoGe

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We report upper critical field $B_{c2}(T)$ measurements on a single-crystalline sample of the ferromagnetic superconductor UCoGe. $B_{c2}(0)$ obtained for fields applied along the orthorhombic axes exceeds the Pauli limit for $B \parallel a, b$ and shows a strong anisotropy $B_{c2}^a \approx B_{c2}^b \gg B_{c2}^c$. This provides evidence for an equal-spin pairing state and a superconducting gap function of axial symmetry with point nodes along the c axis, which is also the direction of the uniaxial ferromagnetic moment $m_0 = 0.07\mu_B$. An unusual curvature or kink is observed in the temperature variation of B_{c2} , which possibly indicates UCoGe is a two-band ferromagnetic superconductor.

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Recently, a new member of the family of itinerant ferromagnetic superconductors has been reported, UCoGe [1]. Itinerant ferromagnetic superconductors attract much attention because they form an exception to the general rule which says that superconductivity (SC) and ferromagnetic (FM) order are mutually exclusive: in the standard BCS theory, magnetic interactions impede phonon-mediated singlet superconductivity [2]. The coexistence of superconductivity and metallic ferromagnetism therefore calls for an exotic explanation, which is offered by spin fluctuation models: on the border of ferromagnetic order, critical magnetic fluctuations mediate superconductivity by pairing the electrons in triplet states [3,4]. The ferromagnetic superconductors known to date are UGe₂ (under pressure) [5], possibly UIr (under pressure) [6], URhGe [7], and the newcomer UCoGe [1]. The latter two materials are of special interest because superconductivity occurs at ambient pressure, which provides a unique opportunity to apply a wide range of experimental techniques for the investigation of magnetically mediated superconductivity. Moreover, since URhGe and UCoGe both crystallize in the orthorhombic TiNiSi structure (space group P_{nma}) [8], but have different magnetic interaction strengths, systematics in the variation of the SC properties may be studied.

The magnetic and SC parameters reported for UCoGe so far have been extracted from measurements on polycrystalline samples [1]. Magnetization measurements show that UCoGe is a weak FM with a Curie temperature $T_C = 3$ K and a small ordered moment $m_0 = 0.03\mu_B$. The itinerant nature of the FM state is corroborated by the small value of the magnetic entropy (0.3% of $R\ln 2$) associated with the magnetic transition. The SC properties depend sensitively on the quality of the samples. SC is observed with a resistive transition temperature $T_s = 0.8$ K for the best sample. Thermal expansion and specific-heat measurements provide solid evidence for bulk magnetism and superconductivity.

In this Letter, we report the first results of magnetic and transport measurements on single-crystalline UCoGe. Magnetization measurements show that UCoGe is a uni-

axial FM, with an ordered moment $m_0 = 0.07\mu_B$ pointing along the orthorhombic c axis. The suppression of the SC transition in a magnetic field B was studied by resistivity measurements. The upper critical field $B_{c2}(T)$ measured for B applied along the orthorhombic axes is not Pauli limited for $B \parallel a, b$ and shows a large anisotropy. The observed anisotropy provides evidence for a SC gap function of axial symmetry with point nodes along the direction of the ordered moment. Such a SC gap function is in agreement with recent order parameter calculations for orthorhombic ferromagnetic spin-triplet superconductors [9–11]. A pronounced positive curvature or kink is observed in $B_{c2}^{a,b}(T)$ which possibly indicates two-band superconductivity [3,11,12].

A polycrystalline batch with nominal composition U_{1.01}CoGe was prepared by arc melting the constituents (natural U 3N, Co 3N, and Ge 5N) in a water-cooled copper crucible under a high-purity argon atmosphere. Next, a single-crystalline rod was pulled from the melt using a modified Czochralski technique in a tri-arc furnace under a high-purity argon atmosphere. Electron micro-probe analysis confirmed the single-phase nature of the grown crystal. Single-crystallinity was checked by x-ray Laue backscattering. Samples for various measurements were cut by spark erosion. Transport measurements on the as-grown samples show a residual resistance ratio, $RRR = R(300\text{ K})/R(1\text{ K}) \approx 5$, and rather poor FM and SC properties. However, after an annealing procedure, like applied for URhGe [13], the RRR increases to 30, and proper FM and SC transitions appear.

Magnetization measurements were carried out down to temperatures of 2 K and magnetic fields up to 5 T in a SQUID magnetometer on bar-shaped samples cut along the different crystallographic directions. Corrections due to demagnetizing fields are neglected because the demagnetization factor is small ($N \approx 0.1$). Four-point low-frequency ac-resistivity data were obtained using a phase-sensitive bridge for $T = 0.02$ –10 K on a bar-shaped sample with the current along the a axis and $RRR = 30$. In Fig. 1, we show the resistivity $\rho(T)$ measured for a current along the a axis. The magnetic transition is observed as a

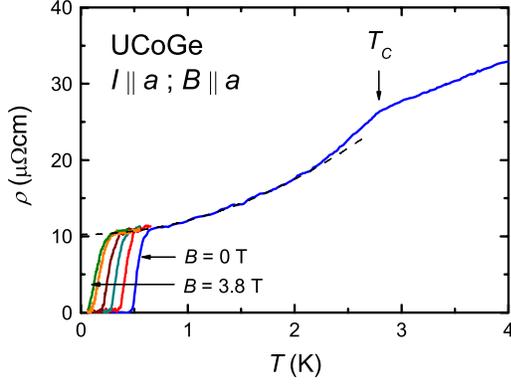


FIG. 1 (color online). Temperature variation of the resistivity of UCoGe for a current $I \parallel a$. Ferromagnetic ordering at $T_C = 2.8$ K shows up as a sharp kink. The dashed line represents $\rho(T) = \rho_0 + AT^2$ with $\rho_0 = 10.2 \mu\Omega \text{ cm}$ and $A = 1.8 \mu\Omega \text{ cm K}^{-2}$. Superconductivity appears below 0.6 K. In a magnetic field ($B \parallel a$), superconductivity is suppressed (B -values from right to left are 0, 0.8, 1.6, 2.4, 3.4, and 3.8 T).

sharp kink at $T_C = 2.8$ K. Below 2 K, $\rho \propto T^2$ due to scattering at magnons. Superconductivity appears below 0.6 K and the transition width $\Delta T_s \approx 0.1$ K. Thermal expansion data taken on the same sample demonstrate superconductivity is a bulk property [14].

The suppression of superconductivity was investigated by resistivity measurements in fixed magnetic fields applied along the orthorhombic a (longitudinal) and b and c axis (transverse). In a field, ΔT_s gradually increases to 0.15 K at the highest fields (5 T). The temperature at which SC is suppressed is taken by the midpoints of the transitions. In an applied field, the FM state rapidly forms a mono domain ($B < 0.01$ T), and we did not observe any hysteric behavior in B_{c2} . The main results are shown in Fig. 2. At least three remarkable features appear in the data: (i) the large value of $B_{c2}(0) \approx 5$ T for $B \parallel a, b$, (ii) the large anisotropy, $B_{c2}^a \approx B_{c2}^b \gg B_{c2}^c$, of a factor ~ 10 , and (iii) for all B -directions $B_{c2}(T)$ has a pronounced upturn when lowering the temperature. Clearly, this behavior is at odds with standard BCS spin-singlet pairing.

Let us first address the large $B_{c2}^{a,b}(0)$. The Pauli paramagnetic limit for a weak coupling spin-singlet superconductor is $B_{c2}^{\text{Pauli}}(0) = 1.83T_s$ [15]. By including the effect of spin-orbit coupling, a comparable $B_{c2}(0)$ value [16] results. Combined Pauli and orbital limiting could therefore in principle account for the small value of $B_{c2}^c(0)$, but not for the large $B_{c2}^{a,b}(0)$ values. Also, a strong-coupling scenario is unrealistic as this would involve a huge coupling constant $\lambda \approx 20$ [17]. The absence of Pauli limiting therefore points to a triplet SC state with equal-spin pairing (ESP). However, a prerequisite for triplet pairing is a sufficiently clean sample, with a mean free path ℓ larger than the coherence length ξ [18]. An estimate for ℓ and ξ can be extracted within a simple model [19] from the (large) initial slope $dB_{c2}^{a,b}/dT = -7.9\text{--}8.4$ T/K. With the

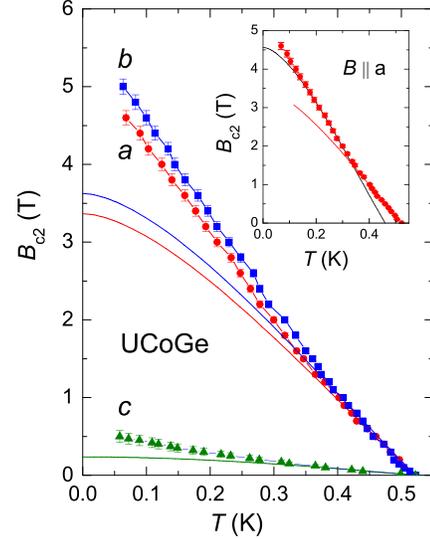


FIG. 2 (color online). Temperature dependence of the upper critical field of UCoGe for B along the a , b , and c axis. The solid lines show the calculated dependence for a superconducting gap function with axial (along c) and polar symmetries (along a and b) (see text). Inset: $B_{c2}(T)$ for $B \parallel a$, plotted together with the polar state functions for two superconducting phases with zero-field transitions at 0.46 and 0.52 K.

specific-heat coefficient $\gamma = 0.057$ J/mol K² [1] and $\rho_0 = 10.2 \mu\Omega \text{ cm}$, we calculate $\ell \approx 900$ Å and $\xi \approx 120$ Å and conclude our single crystal satisfies the clean-limit condition.

The order parameter in an orthorhombic itinerant ferromagnetic superconductor with spin-triplet pairing and strong spin-orbit coupling has been worked out by Fomin [9] and Mineev [10]. Under the assumption that the exchange splitting of the Fermi surface is sufficiently large, such that the pairing between electrons from spin-up and down bands is negligible, equal-spin pairing will give rise to two-band superconductivity with gap functions $\Delta_{\parallel}(R, k) = -\eta_1(R)f_-(k)$ and $\Delta_{\perp}(R, k) = \eta_2(R)f_+(k)$, where $\eta_{1,2}$ are the order parameter amplitudes. From the symmetry-group analysis for an orthorhombic uniaxial FM, it follows that only two SC gap structures $f_{\pm}(k)$ are possible. Assuming that the ordered moment m_0 is directed along the z axis, then the SC gap has zeros (nodes) parallel to the magnetic axis ($k_x = k_y = 0$, A phase) or a line of zeros on the equator of the Fermi surface ($k_z = 0$, B phase). In other words, the A phase has a gap function of axial symmetry with nodes along m_0 , and the B phase has a gap of polar symmetry with a line of nodes perpendicular to m_0 . Before relating the anisotropy in B_{c2} to the structure of the SC gap, we first examine the magnetization $M(B)$.

$M(B)$ for a field along the a , b , and c axis is presented in Fig. 3. The data reveal UCoGe is an uniaxial FM with the ordered moment $m_0 \parallel c$. The inset shows $M_c(T)$ measured in a small field $B = 0.01$ T. By smoothly extrapolating the data for $T \rightarrow 0$, we obtain $m_0 = 0.07\mu_B$. This value is in

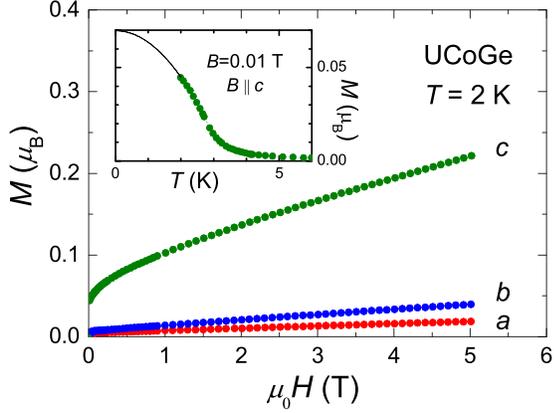


FIG. 3 (color online). Magnetization of UCoGe for fields along the a , b , and c axis at $T = 2$ K. Ferromagnetic order is uniaxial with m_0 pointing along the c axis. The inset shows $M(T)$ for $B = 0.01$ T along c . In the limit, $T \rightarrow 0$ $m_0 = 0.07\mu_B$.

agreement with the powder averaged value $m_0^{\text{powder}} = 0.03\mu_B \approx m_0^{c\text{-axis}}/2$ [1]. For $B \parallel a, b$, the induced magnetization is small. The large initial increase in $M_c(B)$ is related to the relatively high temperature of $0.7T_C$ at which the data are taken.

Having established that UCoGe is a uniaxial ferromagnet with $m_0 \parallel c$, we now turn to the anisotropy in B_{c2} . The effect of an anisotropic p -wave interaction on B_{c2} has been investigated by Scharnberg and Klemm [20]. For a p -wave interaction that favors one direction over the other two, a polar state has the highest T_s , while conversely, if the p -wave interaction is weakest in one direction, an axial state is favored. Since the SC gap is fixed to the crystal lattice by spin-orbit coupling, B_{c2} in general will show a strong anisotropy. We first consider the case of a polar state with the maximum gap direction along the uniaxial direction $m_0 \parallel c$ (i.e., the strength of the pairing interaction along c is stronger than in the ab plane, $V_c > V_{ab}$). The upper critical fields have been calculated for B in the plane of the nodes ($B \perp m_0$) (CBS = Completely Broken Symmetry state) and $B \parallel m_0$ along the maximum of the gap (polar state) [20]. For $V_{ab} = 0$, their ratio for $T \rightarrow 0$ is given by $B_{c2}^{\perp}(0)/B_{c2}^{\parallel}(0) = 0.466(m_{ab}/m_c)^{1/2}$, where m_{ab}/m_c reflects the anisotropy in the effective mass. For our orthorhombic material, m_{ab}/m_c is of the order of 1, and the model predicts $B_{c2}^{\perp}(0)/B_{c2}^{\parallel}(0)$ is $\sim 1/2$. This is at variance with the results in Fig. 2 where the anisotropy ratio ~ 10 , and we conclude that a polar gap cannot explain the anisotropy in B_{c2} . For the axial state (nodes along $m_0 \parallel c$ and the maximum gap in the ab plane), the anisotropy in B_{c2} is reversed [20] $B_{c2}^{\perp}(0) > B_{c2}^{\parallel}(0)$. This is the situation in UCoGe. $B_{c2}^{\parallel}(T)$ has been calculated in Ref. [21] (Anderson-Brinkman-Morel state), but calculations for $B_{c2}^{\perp}(T)$ are not at hand. However, since the gap is maximum, one may assume that $B_{c2}^{\perp}(T)$ can be represented by the polar function. In Fig. 2, we compare the anisotropy

in B_{c2} calculated in this way with the experimental results. We conclude that the measured anisotropy supports an axial state, but the model calculations do not track the experimental results at lower T . This is due to the pronounced positive curvature in $B_{c2}(T)$, which we discuss next.

For $B \parallel a$ and b , the slope $-dB_{c2}/dT$ has initial values of 7.9 and 8.4 T/K but increases to 11.4 and 12.1 T/K at lower T . For $B \parallel a$, the increase is rather abrupt (kink-like) and takes place in a narrow T range centered around 0.33 K (see inset in Fig. 2), while for $B \parallel b$, the increase is more gradual. An overall smooth increase of B_{c2} is observed for $B \parallel c$. We stress that if we define T_s in another way, like by T_s^{onset} or by the 10% or 90% points (measured by the drop in resistance), the absolute values for B_{c2} change slightly, but the upward curvature remains. An upward curvature in $B_{c2}(T)$ was also reported for the polycrystalline samples [1].

Several appealing explanations for a kink ($B \parallel a$) or upward curvature ($B \parallel b$) in $B_{c2}(T)$ have been given in the literature, of which a crossover between two phases in a two-band ferromagnetic superconductor [11] is perhaps the most appealing. Mineev and Champel [11] have evaluated the linearized Ginzburg-Landau equations including gradient terms for a cubic two-band FM superconductor with gaps Δ_{\uparrow} and Δ_{\downarrow} . Depending on the strength of the pairing interactions measured by $g_1 = V_{\uparrow\uparrow}\langle |f_-(k)|^2 N_{0\uparrow}(k) \rangle$ and $g_2 = V_{\downarrow\downarrow}\langle |f_+(k)|^2 N_{0\downarrow}(k) \rangle$ (here $N_{0,\uparrow}$ is the angular dependent density of states) and a number of anisotropy coefficients, an upturn in $B_{c2}(T)$ is predicted. In the more anisotropic situation of an orthorhombic system, calculations are more tedious, but also then a crossover between ESP states in field is possible. Notice that in the two-band model, it is not *a priori* known what the ground state is ($|\uparrow\uparrow\rangle$ or $|\downarrow\downarrow\rangle$), as this depends on g_1 and g_2 and the anisotropy coefficients. In order to illustrate the possibility of the two-band superconductivity scenario, we have plotted in the inset in Fig. 2 the polar state functions [21] for two SC phases with zero-field transition temperatures of 0.46 and 0.52 K. A description with two polar gaps tracks the experimental data down to $T \approx 0.16$ K, but fails there below. Note however that in the two-band model of Ref. [11] in zero field, only one superconducting transition occurs and that the upturn in B_{c2} is due to a field-induced crossover. This is in accordance with the zero-field thermal expansion data, which show a single SC transition [14]. Another interesting scenario for an upwards curvature in B_{c2} is a rotation of the quantization axis of the ESP state from along m_0 at low fields, towards the field direction in high fields. The upward curvature in B_{c2} is then possibly explained by the field not being perfectly aligned along c . Scharnberg and Klemm [20] have also investigated the possibility of a kink in B_{c2} . For the axial symmetric case, a kink appears for $B \parallel c$ for a specific ratio of the strength of the pairing interaction, namely $V_c/V_{ab} < 0.866$, which could give rise to a transition from the Scharnberg-Klemm

state to the polar state. However, an upturn is not predicted for B directed in the ab plane. The only other unconventional superconductor which shows a clear kink in B_{c2} is the heavy-fermion material UPt_3 [22]. Here, the kink is attributed to the intersection of two $B_{c2}(T)$ curves of the two SC phases [23] which appear even in the absence of a magnetic field. The split transition is attributed to the coupling of a two-component order parameter, belonging to the E_{1g} or E_{2u} representation in the hexagonal crystal, to a symmetry breaking field.

The upper critical field of the FM superconductor URhGe (isostructural to UCoGe) has been investigated by Hardy and Huxley [13]. B_{c2} , measured on single crystals with RRR 's of 21 and 34, i.e., comparable to the value of the UCoGe single-crystal, also show a large anisotropy. The data do not show any sign of an upward curvature and are well described by the model functions for a polar state with a maximum gap parallel to a , and for the CBS state [20] with nodes in the direction of b and c . This polar gap structure takes into account the easy- (bc) -plane nature of the magnetization: $m_0 = 0.42\mu_B$ points along c , but for $B \parallel b$ rotates towards the b axis [24]. Surprisingly, superconductivity reappears when the component of the applied field along the b axis reaches ~ 12 T [24] and the ordered moment rotates towards the b axis. Compelling evidence is at hand that in the high-field as well as in the low-field SC phase, superconductivity is stimulated by critical magnetic fluctuations associated with the field-induced spin-reorientation process. Near the quantum critical point, an acute enhancement of B_{c2} is observed [25] and superconductivity survives in fields as large as 28 T applied along a . It will be highly interesting to investigate whether the apparent lack of saturation of B_{c2} for $T \rightarrow 0$ in UCoGe has a similar origin, namely, the proximity to a field-induced quantum critical point.

In conclusion, we have reported the first measurements of the magnetic and superconducting parameters of a single crystal of UCoGe. We find that ferromagnetic order is uniaxial with the ordered moment $m_0 = 0.07\mu_B$ pointing along the c axis. The upper critical field for $B \perp m_0$ exceeds the upper critical field for $B \parallel m_0$ by a factor 10. The magnitude and anisotropy of B_{c2} support p -wave superconductivity and point to an axial SC gap function with nodes along the moment direction (A phase [10]). For $B \perp m_0$, we observe a pronounced upward curvature ($B \parallel b$) or kink ($B \parallel a$). An appealing explanation for this unusual phenomenon is that UCoGe is a two-band ferromagnetic superconductor [3,11,12]. The structure in B_{c2} could then mark a crossover between two SC phases with equal-spin pairing states. On the other hand, UCoGe is close to a ferromagnetic instability as the Curie temperature is low and m_0 is small. This brings to the fore

another exotic explanation for the positive curvature of $B_{c2}(T)$, namely, field-tuning of critical magnetic fluctuations, which promote and enhance magnetically mediated superconductivity.

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