Experimental investigation of potential topological and p-wave superconductors
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Angular variation of the magnetoresistance of the superconducting ferromagnet UCoGe

We present an extensive magnetoresistance study conducted on single-crystalline samples of the ferromagnetic superconductor UCoGe. The data show a pronounced structure at $B^* = 8.5 \, \text{T}$ for a field applied parallel to the ordered moments, $m_0$. Angle dependent measurements reveal this field-induced phenomenon has a uniaxial anisotropy. Magnetoresistance measurements under pressure show that $B^*$ increases with pressure at the significant rate of $3.2 \, \text{T/GPa}$. We discuss $B^*$ in terms of a field-induced polarization change of the U and Co moments. Upper critical field measurements corroborate the extraordinary S-shaped $B_{c2}(T)$-curve reported for a field along the b-axis of the orthorhombic unit cell of UCoGe.

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6.1 Introduction

The intermetallic compound UCoGe belongs to the select group of superconducting ferromagnets [1]. In this intriguing group of materials, superconductivity develops in the ferromagnetic state at a temperature \( T_s \) well below the Curie temperature, \( T_C \), for ferromagnetic ordering [2,3]. Moreover, below \( T_s \), superconductivity and ferromagnetic order coexist on the microscopic scale. The superconducting ferromagnets discovered so far are UGe\(_2\) (under pressure [4]), URhGe [5], UIr (under pressure [6]) and UCoGe [1]. The co-occurrence of ferromagnetism and superconductivity is at odds with the standard BCS (Bardeen-Cooper-Schrieffer) scenario for phonon-mediated spin-singlet superconductivity, since the ferromagnetic exchange field impedes spin-singlet Cooper pairing [7]. Instead, alternative models have been proposed that exploit the itinerant nature of the ferromagnetic order where critical spin fluctuations, connected to a magnetic instability, mediate an unconventional, spin-triplet type of pairing [8,9]. Indeed, these uranium intermetallics, in which the 5f-electrons are delocalized, are all close to a magnetic instability that can be induced by mechanical pressure, chemical doping or an applied magnetic field [10]. Unraveling the properties of superconducting ferromagnets might help to understand how spin fluctuations can stimulate superconductivity, which is a central theme for materials families as diverse as heavy-fermion, high-\( T_s \) cuprate and iron pnictide superconductors.

UCoGe crystallizes, just like URhGe, in the orthorhombic TiNiSi structure with space group \( Pnma \) [11]. The coexistence of superconductivity and ferromagnetism was first reported by Huy \textit{et al.} [1,12]. High-quality single crystals with a residual resistance ratio, \( RRR = R(300\text{K})/R(1\text{K}) \), of 30 have a Curie temperature \( T_C = 2.8 \text{ K} \) and show superconductivity with \( T_s = 0.5 \text{ K} \). UCoGe is a uniaxial ferromagnet. The spontaneous magnetic moment, \( m_0 \), points along the \( c \)-axis and attains the small value of 0.07 \( \mu_B \) per U-atom in the limit \( T \to 0 \). Proof for the microscopic coexistence of superconductivity and ferromagnetic order is provided by \( \mu \text{SR} \) (muon spin relaxation and rotation) [13] and \( ^{59}\text{Co-NQR} \) (nuclear quadrupole resonance) [14] experiments. Evidence for spin-triplet Cooper pairing has been extracted from the magnitude of the upper critical field \( B_{c2}^\perp \) (measured for the field directed perpendicular to \( m_0 \)), which greatly exceeds the Pauli limit for spin-singlet superconductivity [12,15]. The important role of spin fluctuations in promoting superconductivity is established by the large anisotropy of the upper critical field, \( B_{c2}^\perp \gg B_{c2}^\parallel \) [12,15]. For \( B \parallel m_0 \) the magnetic transition becomes a cross-over, spin fluctuations are rapidly quenched and, accordingly, superconductivity is suppressed, while for
Angular variation of the magnetoresistance of the superconducting ferromagnet UCoGe

$B \perp m_0$ spin fluctuations become more pronounced and superconductivity is enhanced. At the microscopic level, the close link between anisotropic critical magnetic fluctuations and superconductivity was recently put on a firm footing by $^{59}$Co-NMR (nuclear magnetic resonance) [16,17] and inelastic neutron scattering [18].

Yet another salient property of UCoGe is the unusual S-shaped curvature of the upper critical field for a field direction along the $b$-axis, $B^b_{c2}$, which yields the large value of $\sim 18$ T when $T \to 0$ [15]. This field-reinforced superconductivity seems to be closely connected to a field-induced quantum critical point as a result of the progressive depression of the Curie temperature [3,15,19]. The peculiar response of the magnetic and superconducting phases to a magnetic field calls for a detailed investigation of the anisotropy in the magnetic, thermal and transport properties. Here we present an extensive angle dependent magnetotransport study on high-quality single crystals of UCoGe for fields directed in the $bc$- and $ac$-planes of the orthorhombic unit cell. We identify a pronounced maximum in the magnetoresistance when the component of the field along the $c$-axis reaches a value $B^c = 8.5$ T. Measurements of $B^*$ as a function of pressure show a roughly linear increase of $B^*$ at a rate $dB^*/dp = 3.2$ T/GPa. The uniaxial nature of $B^*$ and its large variation with pressure provide strong indications for a close connection to an unusual polarizability of the U and Co moments. Transport measurements around the superconducting transition in fixed magnetic fields $B \parallel b$ reveal that also our samples exhibit the S-shaped $B_{c2}$-curve when properly oriented in the magnetic field.

6.2 Sample preparation

A polycrystalline batch with nominal composition U$_{1.01}$CoGe was first fabricated 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. Then, with the help of a modified Czochralski technique, a single crystalline rod was pulled from the melt in a tri-arc furnace under a high-purity argon atmosphere. The crystal was shown to be comprised of a single-phase, by means of electron micro-probe analysis. Single-crystallinity was checked by x-ray Laue backscattering. Bar-shaped samples with typical dimensions $5 \times 1 \times 1$ mm$^3$ were cut from the crystals by means of spark erosion. Magnetotransport measurements were carried out on two samples with the current, $I$, along the $b$-axis and $c$-axis, with $RRR$-values of 30 (sample #1) and 8 (sample #2), respectively. The former had already been used in the past for a previous study of the upper-critical field of UCoGe [12].
6.3 Magnetoresistance

In Fig. 6.1 we show the resistivity of UCoGe (sample #1) as a function of the magnetic field applied along the c-axis. At the lowest temperature $T = 0.27$ K ($< T_s$) the initial steep rise signals the suppression of superconductivity at $B_{c2} = 0.2$ T. Next, $\rho(B)$ steadily increases and passes through a pronounced maximum at $B^* = 8.5$ T. Increasing temperature shows that the maximum at $B^*$ is a robust property and it can be identified in the data up to at least 10 K. Although $B^*$ can be observed to exist up to $T = 10$ K, its field value varies only weakly with temperature, as can be seen in the right inset of Fig. 6.1. We remark that the overall resistivity rapidly increases with temperature and in the paramagnetic phase the initial low-field magnetoresistance is negative. In the left inset of figure 6.1, we show data taken at $T = 0.065$ K in strong magnetic fields up to 33 T. The maximum at $B^*$ is very pronounced indeed. For fields exceeding 12 T (i.e. above $B^*$) the magnetoresistance starts a steady increase that leads to the large value of 40 $\mu\Omega$cm at the maximum field used of 33 T.

In order to investigate the magnetocrystalline anisotropy of $B^*$, we have measured the angle dependence of the magnetoresistance. The data taken in a dilution refrigerator at $T = 0.15$ K are shown for a field rotation in the $bc$-plane and in the $ac$-plane in the upper and lower panels of Fig. 6.2, respectively. The major experimental observation is the steady upward shift of the maximum in $\rho(B)$ when the field is rotated away from the c-axis. The value $B^*(\theta)$ is proportional to $B^*(0)/\cos\theta$, where $\theta$ is the angle at which the field is tilted away from the c-axis. This functional behavior is illustrated in the inset in Fig. 6.2 and holds for the $bc$- as well as for the $ac$-plane. For $\theta > 58^\circ$ the maximum in $\rho(B)$ falls outside the magnetic field range probed in the dilution refrigerator. We remark that the value of the maximum magnetoresistance $\rho^*$ at $B^*$ is essentially independent of the field-angle. This tells us the angle dependent magnetoresistance data could be collapsed onto a single reduced curve $\rho/\rho^* \text{ versus } B/B^*$. We conclude the maximum in $\rho(B)$ occurs when the component of the magnetic field along the c-axis reaches $B^* = 8.5$ T. This behavior confirms its uniaxial nature, and it has this uniaxial character in common with the ferromagnetic order in UCoGe. The suppression of superconductivity in the field-angle interval probed in Fig. 6.2 takes place at a low value of the upper-critical field $B_{c2}$ for $B \parallel c$-axis [15].
Angular variation of the magnetoresistance of the superconducting ferromagnet UCoGe

Figure 6.1 Resistivity of UCoGe (sample #1) as a function of the magnetic field $B \parallel c$ at temperatures of 0.27, 2, 3, 4, 6, 8 and 10 K, as indicated. The current was applied along the $b$-axis. Left inset: High-field magnetoresistance ($B \parallel c; I \parallel b$) up to $B = 33$ T at $T = 0.065$ K. Right inset: $B^*$ as a function of temperature determined from the maximum in the magnetoresistance.

Figure 6.2 Angular variation of the magnetoresistance of UCoGe (sample #1) at $T = 0.15$ K. Upper panel: field rotation in the $bc$-plane; $\theta = -58, -53, -48, -43, -38, -33, -28, -18, -8, 2, 12, 22$ and 27 degrees, where $0^\circ$ corresponds to $B \parallel c$. Lower panel: field rotation in the $ac$-plane; $\theta = -53, -48, -43, -38, -28, -18, -8, 2, 12, 22$ and 27 degrees. The current is always applied along the $b$-axis. Inset: $B^*$ as a function of $\theta$. The solid line represents $B^*(\theta) = B^*(0)/\cos\theta$.
The pressure variation of \( B^* \) was investigated for sample #2 for \( B \parallel I \parallel c \) for pressures up to 1.29 GPa in the \(^3\text{He} \) refrigerator. The lower residual resistance ratio (\( RRR = 8 \)) for this sample results in a very different field variation \( \rho(B) \), as shown in Fig. 6.3 for \( T = 0.25 \) K. Similar magneto-resistance data were recently reported in Ref. [20]. After the initial steep rise, due to the suppression of superconductivity, \( \rho(B) \) steadily decreases and shows a kink rather than a maximum near 8.5 T. The field at which the kink appears identifies \( B^* \). Under pressure \( B^* \) increases at an initial linear rate of 3.2 T/GPa (see inset to Fig. 6.3 for data at \( T = 0.25 \) K and 1.0 K). As the inset shows the temperature variation is weak. At our highest pressure, \( B^* \) falls outside the available magnetic field range in the \(^3\text{He} \) refrigerator.

\[ \text{Figure 6.3 Magnetoresistance of UCoGe (sample \#2) for } B \parallel I \parallel c \text{ at pressures of 0.26, 0.52, 0.77, 1.03 and 1.29 GPa as indicated. The temperature is } T = 0.25 \text{ K. Inset: } B^* \text{ as a function of pressure at } T = 0.25 \text{ K (circles) and } T = 1.0 \text{ K (squares). The value of } B^* \text{ at ambient pressure (triangle) is taken from sample \#1. The solid lines connect the data points. Data taken by A. Nikitin.} \]

6.4 Upper critical field \( B_{c2} \)

The upper critical field \( B_{c2}^{\perp} \) for a field direction (\( B \parallel a \) or \( B \parallel b \)) perpendicular to the ordered moment (\( m_0 \parallel c \)) is extremely sensitive to the precise orientation of the magnetic field [15]. In order to substantiate this unusual \( B_{c2}^b \) -behavior of our single crystals we measured sample \#1 as a function of field orientation in the dilution refrigerator. Special care was taken to enable field rotation in the \( bc \)-plane. After fine tuning to \( B \parallel b \) we measured the resistivity in fixed magnetic fields. The results are shown in Fig. 6.4. For \( B = 0 \) the superconducting transition sets-in at 0.6 K and has a width \( \Delta T_c = 0.1 \) K. On applying a magnetic field, the superconducting transition progressively shifts to lower temperatures, and is still visible up to the highest field (16 T). Striking features are \( (i) \) the change of the rate of the reduction of \( T_c \) in
the field range 5-9 T, and (ii) the narrowing of $\Delta T_s$ across the same field range. The upper critical field, determined by taking the midpoints of the transitions, is shown in Fig. 6.6. $B_{c2}(T)$ has an unusual curvature for $B > 4$ T and extrapolates to the large value of 17 T in the limit $T \to 0$, in good agreement with the results reported previously [15]. In the inset we show the strong angular variation of $B_{c2}$ around $B \parallel b$ measured at $T = 0.15$ K. For a tilt-angle of typically only 2° the upper critical field has already diminished by a factor of 3 [15]. For $B \parallel a$, the upper critical field measurements displayed in Figs. 6.5 and 6.7 show there to be a remaining misorientation of the order $\sim 2^\circ$, resulting in a rather small value of $B_{c2}'$ ($B_{c2}' \sim 10$ T at $T = 0$), whereas in Ref. [15], $B_{c2}' \sim 30$ T.

**Figure 6.4** Superconducting transition of UCoGe (sample #1) measured by resistivity for $B \parallel b \parallel I$ in fixed magnetic fields from 0 to 16 T with steps of 1 T (from right to left).

**Figure 6.5** Superconducting transition of UCoGe (sample #1) measured by resistivity for $B \parallel a, I \parallel b$ in fixed magnetic fields from 0 to 8 T with steps of 1 T (from right to left).

**Figure 6.6** Temperature variation of the upper critical field $B_{c2}(T)$ of UCoGe (sample #1) measured for $B \parallel b$. Inset: Angular variation of $B_{c2}(T)$ in the $bc$-plane at $T = 0.15$ K.

**Figure 6.7** Temperature variation of the upper critical field $B_{c2}(T)$ of UCoGe (sample #1) measured for $B \parallel a$. 
6.5 Discussion

The major result from these angle dependent magnetoresistance measurements is the pronounced maximum at a field $B^*$, which occurs when the component of the magnetic field along the $c$-axis reaches a value of 8.5 T. This characteristic field $B^*$ is a robust property of our samples, but the shape of the magnetoresistance $\Delta \rho \equiv \rho(B) - \rho(0)$ is different for sample #1 (with a maximum at $B^*$, Fig. 6.1) and sample #2 (with a kink at $B^*$, Fig. 6.3). We remark that there are two obvious differences between the experiments. Firstly, the sample quality is very different as quantified by the residual resistivity value $\rho_0$ of 10 and 75 $\mu\Omega\text{cm}$, respectively. Possibly, for sample #2 magnetic disorder makes a large contribution to $\rho_0$, which is then reduced by the magnetic field, resulting in a negative $\Delta \rho$. The second difference is the measurement geometry, i.e. transversal ($B \parallel c$, $I \parallel b$ for sample #1) versus longitudinal ($B \parallel c \parallel I$ for sample #2) magnetoresistance, since the Lorentz force on the current in general leads to more scattering and a positive $\Delta \rho$. Moreover, in the transverse configuration scattering is expected to be more effective since the current is perpendicular to $m_0$, compared to the longitudinal configuration where the current and $m_0$ are aligned. Measurements in the transverse geometry with $B \parallel c$ have not appeared in the literature so far, while longitudinal ($c$-axis) magnetoresistance data have been reported on two samples of different quality: (i) a magnetoresistance trace taken on a sample with $\text{RRR} = 30$ at $T = 0.04 \text{ K}$ shows a weak initially positive $\Delta \rho$ with a small structure near $B^* \approx 9 \text{ T}$ and three additional kink-like features in the field range 17-30 T [21], and (ii) the magnetoresistance of a sample with $\text{RRR} = 5$ has an overall negative $\Delta \rho$ with a kink at $B_k$ or $B^* \approx 9 \text{ T}$ [20], as in our Fig. 6.3. In the latter study the angular variation of $B^*$, measured at $T = 0.04 \text{ K}$ by tilting the field from the $c$-axis towards an arbitrary direction in the ab-plane, was also found to follow the $B^*(\theta = 0)/\cos \theta$-law. The large variation of $\Delta \rho$ with the $\text{RRR}$-value and geometry is uncommon and its understanding is highly relevant in view of the strongly anisotropic magnetic properties of UCoGe.

An appealing scenario that has been put forward to explain the change in magnetoresistance at $B^*$ is a ferro-to-ferrimagnetic transition [20]. This proposal is largely based on a recent polarized neutron diffraction experiment on UCoGe carried out for $B \parallel c$ [22]. In low magnetic field (3 T) the small ordered moment $m_0$ is predominantly located at the U atom, but in a large field of 12 T a substantial moment, antiparallel to the U moment, is induced on the Co site. This unusual polarizability of the Co 3$d$ orbitals may give rise to a field-induced ferri-magnetic-like spin arrangement. Support for this scenario was obtained by
field-dependent ac-susceptibility data [20] which exhibit a maximum near \( B^* \). However, hitherto we were unable to confirm this result in our AC-susceptibility measurement set-up. Recently, the DC-magnetization \( M(B) \) was measured at \( T = 1.5 \) K in pulsed magnetic fields up to \( 53 \) T [23]. The moment polarization is large. \( M(B) \) gradually increases from \( \approx 0.05 \mu_B \) at \( B = 0 \) to \( 0.7 \mu_B \) at the maximum field. For \( B \parallel c \) the data do not show a clear sign of a (meta)magnetic transition, however, a weak structure appears near \( B^* \) in the derivative \( dM/dB \), and a second change of slope occurs near \( 23.5 \) T. Sensitive torque cantilever experiments might be helpful to resolve the possibly anomalous behavior of the magnetization around \( B^* \). Further arguments in favor of a magnetic transition are: (i) the uniaxial (Ising-type) behavior of the ferromagnetic order is reflected in \( B^* \), and (ii) the pressure variation of \( B^* \) (see Fig. 6.3) is large and has a magnitude comparable to the pressure dependence of \( T_C \) [24] assuming \( 1 \) K \( \equiv 1.5 \) T per \( \mu_B \) (the critical pressure for the suppression of ferromagnetic order is \( 1.4 \) GPa). In this scenario the pressure increase of \( B^* \) may be related to the reduced polarizability of the Co moment under pressure.

Another possible origin of the structure in \( \Delta \rho \) near \( B^* \) is a Lifshitz transition, i.e. a field-induced topological change of the Fermi surface. Notably it has been suggested that the multitude of small kink-like features observed in \( \Delta \rho \) for \( B \parallel c \parallel I \) at \( T = 0.04 \) K could hint at a Fermi surface reconstruction [21]. Quantum oscillations have been reported for UCoGe for \( B \parallel b \) but could not be detected for a field direction along or close to the \( c \)-axis. A second indication for the possibility of a field-induced Fermi surface modification comes from thermoelectric power data [25], which show two pronounced peaks at 11.1 and 14.6 T for \( B \parallel b \). While the former peak is associated with field-reinforced superconductivity, the latter peak and the ensuing sign change of the thermopower provide evidence for a topological change of the Fermi surface. In the related material URhGe the field-induced disappearance of a small Fermi-surface pocket was recently demonstrated by quantum oscillations measurements [26].

Overall, the Ising-like nature of the ferromagnetic ground state results in a complex magnetotransport behavior. Moreover, the magnetization [12,23,27], thermal expansion [28], thermoelectric power [25] and thermal conductivity [29] all have a strong magnetocrystalline anisotropy, which makes it difficult to unravel the behavior of UCoGe. However, on the positive side, it is the strong anisotropy that results in longitudinal ferromagnetic fluctuations that are believed to play a major role in inducing spin-triplet superconductivity [17]. A greater understanding of the anisotropy is therefore likely to be important for our understanding of the superconductivity.
The unusual superconducting behavior is demonstrated by the $B_{c2}(T)$ curve reported in Fig. 6.6. We recall the upward curvature for $B > 4$ T and the large value of 17 T in the limit $T \to 0$. The sample (#1) used here comes from the same single-crystalline batch as used in our first measurements of the upper critical field [12], where $B_{c2}(0)$ was found to reach a value of 5 T for $B \parallel b$. This discrepancy can now be attributed to a small misorientation of 2° (see the inset in Fig. 6.6). Still, in the data reported in Ref. [15] the field-reinforced behavior and S-shaped $B_{c2}(T)$ curve is more pronounced than in our data. Possibly, this is due to the somewhat larger $\Delta T_s$ of our sample and/or a remaining tiny misorientation towards the ac-plane [15]. The precise orientation of the sample with respect to the magnetic field direction remains absolutely crucial for pinning down the behaviour of this material. The field-reinforced superconductivity appears to be connected to critical spin fluctuations associated with a field-induced quantum critical point, where the latter is reached by the suppression of the Curie temperature in strong magnetic fields for $B \perp m_0$ [30]. A simple McMillan-type formula [19] can then be used to link $T_s$ to the intensity of the critical spin fluctuations as probed by the effective mass, $m^*$, extracted from the coefficient $A$ of the Fermi-liquid resistivity: $\Delta \rho(T) = \rho_0 + AT^2$. A second, more recently, proposed origin of the field-reinforced superconductivity is a Lifshitz transition [26,29]. Here a vanishing Fermi velocity $v_F = \hbar k_F / m^*$, where $k_F$ is the Fermi wave vector, results in a small coherence length $\xi = \hbar v_F / \pi \Delta$, where $\Delta$ is the BCS excitation gap, which in turn leads to a high value of $B_{c2} = 2\pi \Phi_0 / \xi^2$ (here $\Phi_0$ is the flux quantum). Finally, we mention the progress made in modeling the intricate and anisotropic $B_{c2}(T)$ of UCoGe using a strong-coupling Eliashberg model, exploiting the Ising-type spin fluctuations [31]. There are also approaches being developed based on the completely broken symmetry scenario for parallel-spin $p$-wave superconductors [32].

6.6 Conclusion

In summary, we have presented an extensive angle dependent magnetoresistance study of single crystals of UCoGe for fields directed in the $bc$- and $ac$-planes of the orthorhombic unit cell. We pinpoint a pronounced structure in the magnetoresistance, which occurs when the component of the field along the $c$-axis reaches a value $B^* = 8.5$ T. This behaviour is very pronounced for transverse measurement geometry and rather weak for longitudinal geometry. Measurements of $B^*$ as a function of pressure show a roughly linear increase at a rate $dB^*/dp = 3.2$ T/GPa. The uniaxial nature of $B^*$ and its large pressure variation are consistent
with the interpretation that the change in the magnetoresistance regime at $B^*$ is related to an unusual polarizability of the U and Co moments. Transport measurements in fixed magnetic fields confirm the extraordinary S-shaped $B_{c2}(T)$-behavior reported in the literature, after carefully aligning the sample along the field $B \parallel b$. In order to further unravel these intriguing properties of UCoGe, notably with respect to the close connection between field-induced phenomena, such as a quantum critical point or Lifshitz transition, and superconductivity, an unrelenting research effort is required to probe the strongly anisotropic thermal, magnetic and transport properties of this system with the help of high-quality single crystals.
References


Angular variation of the magnetoresistance of the superconducting ferromagnet $UCoGe$


