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Coexistence and interplay of superconductivity and ferromagnetism in URhGe

F Lévy[1], I Sheikin[2], B Grenier[3], C Marcenat[4] and A Huxley[5]

[1] Département de Physique de la Matière Condensée, Université de Genève, quai Ernest-Ansermet 24, CH1211, Genève 4, Switzerland
[2] GHMFL, CNRS BP166, 38042 Grenoble, France
[3] Université Joseph Fourier & CEA, INAC/SPSMS/MDN, F-38054 Grenoble Cedex 9, France
[4] CEA, INAC, SPSMS, F-38054 Grenoble Cedex 9, France
[5] Scottish Universities Physics Alliance, School of Physics, King's Buildings, University of Edinburgh, Edinburgh EH9 3JZ, UK

E-mail: florence.levy@physics.unige.ch

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Abstract
As ferromagnetism and superconductivity are usually considered to be antagonistic, the discovery of their coexistence in UGe_2, URhGe, UIr and UCoGe has attracted a lot of interest. The mechanism to explain such a state has, however, not yet been fully elucidated. In these compounds superconductivity may be unconventional: Cooper pairs could be formed by electrons with parallel spins and magnetic fluctuations might be involved in the pairing mechanism. URhGe becomes ferromagnetic below a Curie temperature of 9.5 K, with a spontaneous moment aligned to the c-axis. For temperatures below 260 mK and fields lower than 2 T, superconductivity was first observed in 2001. Recently, we discovered a second pocket of superconductivity. This new pocket of superconductivity appears at higher fields applied close to the b-axis, enveloping a sudden magnetic moment rotation transition at $H_R = 12$ T. Detailed studies of the field induced metamagnetic transition and superconductivity are presented. The possibility that magnetic fluctuations emerging from a quantum critical point provide the pairing mechanism for superconductivity is discussed.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The problem of the coexistence of superconductivity and ferromagnetism, *a priori* two antagonistic properties, was raised in 1957 by Ginzburg [1]. In conclusion, Ginzburg estimated that such a coexistence might be possible if the induction created by the magnetization did not exceed the critical field for superconductivity. From the critical field values and the spontaneous magnetizations measured for ferromagnets at that time it was very unexpected to observe this coexistence. In 1958, Matthias and collaborators demonstrated that even a small concentration of magnetic impurities was enough to destroy lanthanum's superconductivity [2]. Then, in $ErRh_4B_4$ and $HoMo_6S_8$ ferromagnetism and superconductivity were observed simultaneously, but, again the two properties turned out to be antagonistic [3, 4]. Both compounds become superconductors, and then at lower temperature develop ferromagnetic order. For decreasing temperature the ferromagnetic order evolves via a modulated ferromagnetic structure to classical uniform ferromagnetism. When the classical ferromagnetic order is established the superconductivity disappears. Where superconductivity co-exists with the modulated magnetic state, the magnetic modulation period is smaller than the coherence length of the Cooper pairs, so the average magnetic field felt by the Cooper pairs is zero [5].

It is only since 2001 that a true coexistence of ferromagnetism and superconductivity has been experimentally observed. So far, such a coexistence has been reported in four compounds: UGe_2 [6], URhGe [7], UIr [8] and UCoGe [9]. In this paper, we will review why superconductivity and ferromagnetism are supposed to be antagonistic. Then we summarize some of the detailed studies made on URhGe that reveal a spectacular re-entrance of superconductivity with magnetic field enveloping a magnetic transition [10]. The transition field depends on the field orientation with respect to the crystal axes. The re-entrant superconductivity robustly tracks the magnetic transition as the field orientation is changed extending up to fields of at least 32 T, firmly establishing the link between superconductivity and the magnetic transition [11]. Finally, we discuss the pairing mechanism for the superconductivity.

2. Ferromagnetism and superconductivity: antagonism?

For conventional superconductors the superconducting state corresponds to a condensate of Cooper pairs made up from opposite spin electrons. An applied magnetic field tends to destroy superconductivity. Two causes of this destruction can be distinguished: orbital and paramagnetic limiting.

The orbital effect corresponds to the action of the magnetic field on the electron charge. As the field is increased the electrons describe circular motions with decreasing radius. Roughly, when this radius is smaller than the coherence length of the order parameter the superconductivity is destroyed. The critical field can be expressed as (see [12])

$$H_{C2}^{\text{orbital}}(T) = \frac{\Phi_0}{2\pi\xi^2(T)}$$

where $\Phi_0 = \hbar/(2e)$ is the magnetic flux quantum and ξ is the coherence length. The coherence length is proportional to the inverse of the electron effective mass $(m^*)^{-1}$, and thus the orbital critical field is proportional to the square of the effective mass:

$$H_{C2}^{\text{orbital}}(T) \propto (m^*)^2.$$

The paramagnetic effect corresponds to the action of the magnetic field on the electron spin. The field tends to align the spins of the electrons that make up the Cooper pairs. At some point, as the field is increased, it is energetically favorable to lose the condensation energy by breaking the Cooper pairs to align the spins with the field. At zero temperature, the critical field can be expressed as [13]

$$H_{C2}^{\text{paramagnetic}}(T=0) = \frac{\sqrt{2}\Delta(T=0)}{g\mu_B}$$

where $\Delta(T = 0)$ is the superconducting gap at zero temperature, μ_B is the Bohr magneton and g is the Landé factor for the electrons. Within the BCS theory (and for $g = 2$) the paramagnetic field is expressed in tesla as

$$H_{C2}^{\text{paramagnetic}}(T=0) = 1.85\ T_S$$

where T_S is the critical temperature for superconductivity expressed in kelvin.

As a ferromagnet generates an internal magnetic field, ferromagnetism and superconductivity are usually considered to be antagonists.

3. URhGe: interplay of ferromagnetism and superconductivity

Many theories suggest that magnetic fluctuations provide the glue forming superconductivity coexisting with ferromagnetic order (instead of phonons for the BCS theory). Theories differ on the nature of the magnetic excitations and on the respective spin orientation of the electrons forming Cooper pairs. Abrikosov [15] and Suhl [14] suggest a model with magnetic excitations generated by localized spins giving Cooper pairs comprising antiparallel spins, while others, like Fay and Appel [16], assume that the ferromagnetism is itinerant and that Cooper pairs are formed from electrons with parallel spins.

The results obtained for URhGe and presented here tend to support the view that magnetic fluctuations play an important role in the pairing mechanism. URhGe becomes ferromagnetic below a Curie temperature, T_C, of 9.5 K, with a spontaneous moment aligned to the c-axis of its orthorhombic crystal structure. For temperatures below 260 mK and fields lower than 2 T, superconductivity occurs in clean samples [7]. Our recent work has revealed a second pocket of superconductivity engulfing a magnetic moment rotation transition at $H_R = 12$ T. The re-entrant superconductivity was observed up to 32 T and seems to be linked beyond doubt to the magnetic transition. The inference is that superconductivity is a consequence of the proximity to a magnetic quantum critical point associated with this transition.

Figure 1 illustrates, for magnetic fields applied along the b-axis, the interplay between the metamagnetic transition (magnetic transition induced by field) and the re-entrant superconductivity. The moment direction sketched in figure 1(a) was determined at 2 K by elastic neutron scattering on the D23 instrument (ILL, Grenoble). As an increasing magnetic field is applied along the b-axis the magnetic moment tends to rotate towards the applied field. At $H_R = 12$ T there is metamagnetic transition: the moment suddenly aligns with the b-axis. Resistivity measurements for different temperatures, shown in figures 1(b) and (c), clearly point out the link between the metamagnetic transition and re-entrant superconductivity. At 500 mK a peak is observed at H_R, the critical field of the moment rotation transition, while at 40 mK two pockets of superconductivity are observed, one below 2 T and one between 8 and 13 T. The re-entrant superconductivity cannot be explained by the Jaccarino–Peter mechanism: compensation of the applied field by an internal field produced by the magnetic moments [17]. Indeed, in the 8–13 T range the magnetic moments are inclined up to 55° to the applied field direction, and therefore cannot give a null total field. The phase diagram of figure 1(c) highlights that the re-entrant superconductivity is more robust than the low field superconducting pocket with the maximum critical temperature for superconductivity reached exactly at the critical field of the magnetic transition, H_R. We interpret the peak in resistivity observed above the

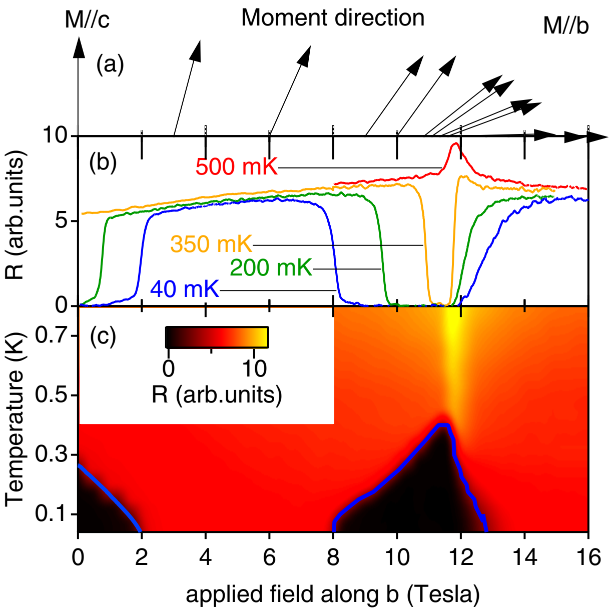

Figure 1. Superconductivity and metamagnetic transition for applied magnetic field along the b-axis. (a) Moment direction established by neutron diffraction. For 12 T applied along $\vec{b}$ the magnetic moments suddenly rotate from the c-axis to the b-axis. (b) Resistivity measurement for different temperatures. At 500 mK, the peak indicates the position of the metamagnetic transition. At 40 mK two pockets of superconductivity are observed, one below 2 T and one between 8 and 13 T. (c) Temperature–applied magnetic field phase diagram obtained from resistivity measurements. Black regions correspond to superconductivity and the white (yellow) line is the signature of the metamagnetic transition.

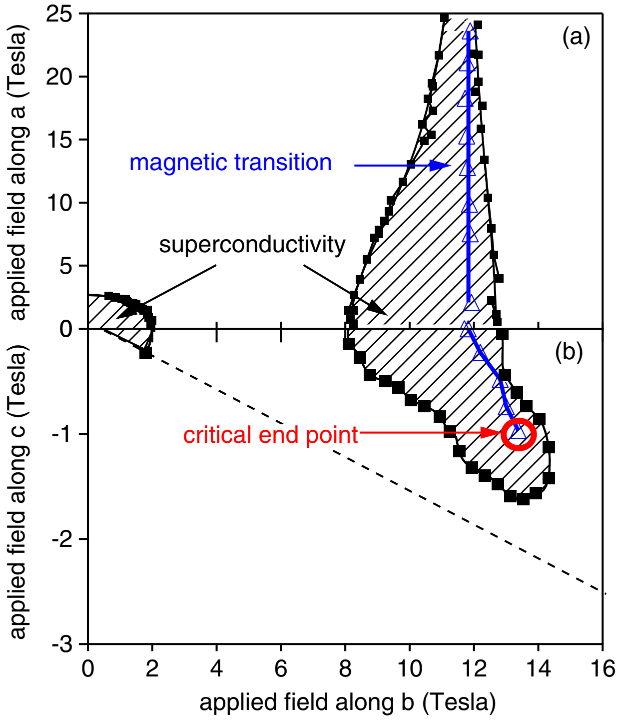

Figure 2. Superconductivity and metamagnetic transition for applied magnetic fields in the (bc) and (ab) planes. Results obtained by resistivity measurements for *in situ* rotation of the applied magnetic field. Black hatched area: extent of superconductivity measured at $T \sim 40$ mK. Blue line with triangles: first order metamagnetic transition, measured at $T \sim 500$ mK. Red circle: critical end point. Dashed line: limit of the angular range over which experimental data were collected.

superconducting transition temperature (shown for 500 mK) as the sum of two contributions. One contribution is a continuous enhancement of the electron effective mass as the magnetic transition is approached because of magnetic fluctuations. The other contribution is a delta-function-like peak arising from extra scattering of the electrons by magnetic domain walls present at the transition; an increased residual resistivity owing to domains is well known in iron [18]. This interpretation assumes that the transition is (weakly) first order, so at the transition there is a coexistence of domains with moments with and without a component along the c-axis. For field along the b-axis, hysteresis, expected at a first order transition, was not clearly resolved in early resistivity and torque measurements. However, hysteresis is clearly visible in more recent torque measurements when an additional field component is applied along the magnetically hard a-axis, cf. figure 3.

Figure 2 reveals the link between the magnetic transition and the re-entrant superconductivity for fields applied in the easy (bc)-magnetic plane, panel (b), and in the (ab)-plane, panel (a). The results were obtained at GHMFL by resistivity measurements with *in situ* rotation of the applied field respectively in the (bc) and (ab) planes. The limits of the superconductivity pockets were determined from measurements made at ~40 mK, while the position of the magnetic transition was determined at a temperature of ~500 mK from the position of the sharp peak in resistivity. In the easy (bc)-magnetic plane, figure 2(b), the critical field of the spin rotation transition increases with the angle of the field from the b-axis. For a 5° misorientation the transition is no longer sharp: the transition changes from first order to a crossover. The point at which the first order magnetic transition line ends is a critical end point: the transition is continuous at this point. At zero temperature this point is a quantum critical end point. Quantum fluctuations are expected to become divergent as this point is approached and could provide a pairing mechanism for superconductivity. Experimentally, in high quality samples, superconductivity engulfs this point and extends along the region over which the first order nature of the magnetic transition is weak. In the (ab)-plane, figure 2(a), the critical field for the magnetic transition corresponds to a fixed field component along the b-axis; i.e., the field along b at which the transition occurs is unaffected by any additional field applied along the hard magnetic a-axis up to 25 T. The magnetic transition remains a first order transition; experimental evidence is (i) the peak in resistivity remains sharp and (ii) a clear hysteresis observed in torque measurement, cf figure 3. In the (ab)-plane the re-entrant superconductivity envelops the magnetic transition and is particularly robust: we observed it up to 32 T, the maximum field available then at GHMFL; cf figure 4. For fields in the (bc)-plane superconductivity extends along the weak first order transition lines away from the quantum critical end points. The extension of the superconducting pocket to very high field as the field is rotated towards the a-axis demonstrates

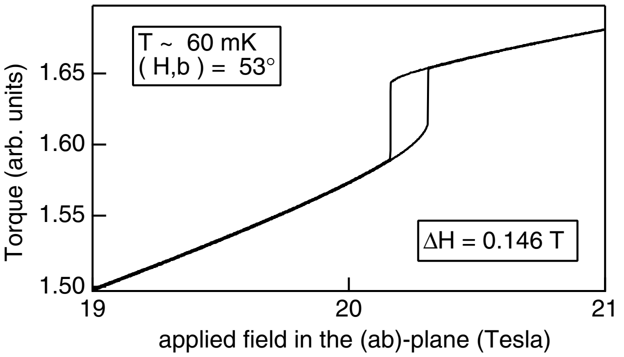

Figure 3. Metamagnetic transition in the (ab)-plane. Torque measurements made at 50 mK for an applied field in the (ab)-plane such that $(\vec{H}, \vec{b}) = 53°$. A clear hysteresis is observed: the transition is first order.

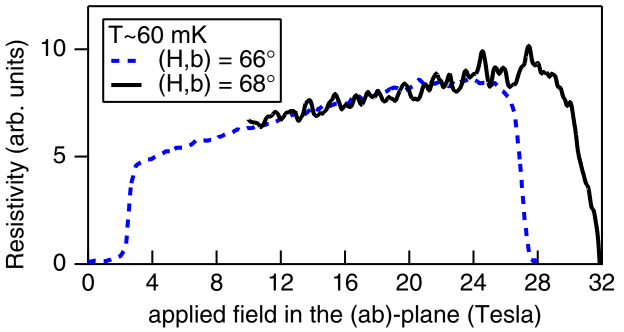

Figure 4. Superconductivity up to 32 T. Resistivity measurement made at 45 mK for an applied field in the (ab)-plane such that $(\vec{H}, \vec{b}) = 66°$ and 68°.

dramatically that the mechanism for the superconductivity is correlated with the magnetic phase transition.

4. Mechanism of the superconductivity?

The work done on URhGe suggests that the superconductivity observed at both low fields and high fields has the same origin and probably a similar order parameter. Given that the superconductivity occurs over such a large continuous range of field along each direction compared with the Pauli paramagnetic limitation of $\sim 1.85 T_S$, equal spins must be paired. If this is indeed driven by the quantum critical fluctuations, the nature of the magnetic transition suggests that these excitations may involve rotation of the spin direction in the (bc)-plane. Such rotation excitation would naturally propagate most easily along the direction perpendicular to the rotation plane i.e. the a-axis. This suggests an order parameter with an orbital structure of the form $\Psi(\vec{k}) \propto k_a$, which has the required odd parity compatible with equal spin pairing. It is curious to note that such an order parameter was already proposed for URhGe by Hardy and Huxley before the discovery of the re-entrant superconductivity [19] based on fitting the anisotropic critical fields for the low field superconducting pocket. Mineev demonstrated that this order parameter is the only one compatible (for a triplet pairing) with a magnetic moment direction inclined at an arbitrary angle

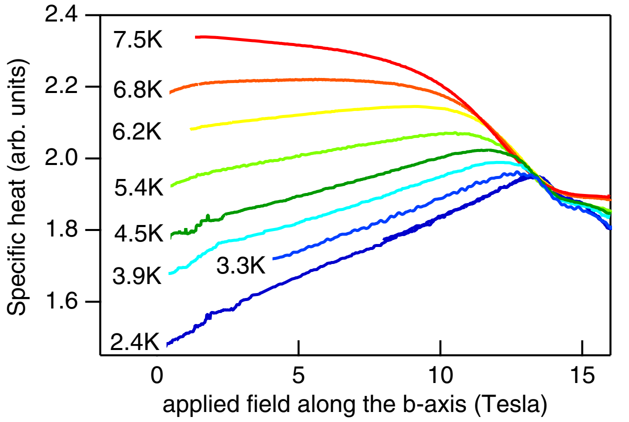

Figure 5. AC specific heat for applied field along the b-axis. These measurements were made at different constant temperatures. A single crystal of few milligrams with a flat surface perpendicular to the b-axis was used glued on to a tensioned chromel–constantan thermocouple. The ac heating was optical, delivered via an optical fiber.

in the (bc)-plane [20, 21], spanning the region of re-entrant superconductivity.

The re-entrant superconductivity feature could be explained by a competition between the critical field and the applied field. Indeed, as explained above, if we assume a triplet pairing $|\uparrow\uparrow\rangle$ only the orbital limit applies, and thus the critical field is proportional to the square of the effective mass. As the magnetic transition is approached the effective mass is enhanced, dressed by magnetic fluctuations, and so is the critical field. Near to the magnetic transition the critical field is higher than the applied field: giving the high field superconductivity pocket. At lower field the critical field is lower than the applied field: superconductivity is destroyed. Eventually below $\sim$2 T the applied field becomes sufficiently small that superconductivity again reappears. Heat capacity measurements shown in figure 5 tend to support this interpretation. At the lowest temperature measured, 2.4 K, the measurements start to reflect the behavior of the electronic γ term of the specific heat, which is proportional to the effective mass. The data then suggest that the effective mass is enhanced as the magnetic transition is approached. However, the 30% increase of heat capacity with field from 0 to 12 T observed at 2.4 K, if this is equated directly with the change of effective mass, is too small to explain the re-entrance. The relative increase in the effective mass is probably however underestimated due to the high measurement temperature. In zero field C/T at 2.4 K is known to be 25% larger than the low-temperature-limit in the normal state [7]. The heat capacity close to 12 T might plausibly also increase as the temperature is reduced below 2.4 K. In addition, although many precautions were taken, because of the huge torque acting on the sample, the applied field was slightly misaligned from the b-axis and so the maximum specific heat value is reached at 13 T instead of 12 T.

5. Conclusion

In URhGe the spectacular re-entrance of superconductivity with magnetic field appears to be correlated with the vicinity to a magnetic transition, which strongly suggests that the superconductivity is driven by the exchange of magnetic excitations. Our results suggest that superconductivity in URhGe at both low and high fields has the same origin. Our results suggest that the order parameter is a triplet polar state with the maximum gap directed along the a-axis. This might naturally result from pairing due to magnetic fluctuations propagating along the a-axis connected with the spin rotation transition of the magnetic moments in the (bc)-plane.

Among the four compounds known to present a coexistence of superconductivity and ferromagnetism, UGe_2 shares several features that invite comparison with URhGe [22]. There is a pocket of superconductivity within the ferromagnetic state engulfing a magnetic transition (induced by pressure in the case of UGe_2). The maximum critical temperature for superconductivity is reached at the pressure where the magnetic transition occurs. In addition, at a fixed pressure of 13.5 kbar, superconductivity in UGe_2 shows a re-entrant behavior as the magnetic transition is crossed by application of field, although the two pockets of superconductivity merge as the temperature is reduced [23].

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