Quantum criticality in the ferromagnetic superconductor UCoGe under pressure and magnetic field

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The pressure-temperature phase diagram of the orthorhombic ferromagnetic superconductor UCoGe was determined by resistivity measurements up to 10.5 GPa. The Curie temperature T_C is suppressed with pressure and vanishes at the critical pressure $p_c \approx 1$ GPa. Superconductivity is observed in both the ferromagnetic state at low pressure, and in the paramagnetic state above p_c up to about 4 GPa. Non-Fermi-liquid behavior appears in a large pressure range. The resistivity varies linearly with temperature around p_c and evolves continuously with pressure to a T^2 Fermi-liquid behavior for $p \gtrapprox 5$ GPa. The residual resistivity as a function of pressure shows a maximum far above p_c at $p^\star = 7.2$ GPa and the amplitude of the inelastic scattering term of the resistivity decreases by more than one order in magnitude at $p^\star$, which appears to mark the entrance into a weakly correlated regime. The pressure dependence of the upper critical field for magnetic field applied along the b and c axis illustrates the drastic difference in the field dependence of the ferromagnetic superconducting pairing. While for $H \parallel b$ axis $H_{c2}(T)$ is driven by the suppression of the ferromagnetic order, it is dominated by the strong initial suppression of the ferromagnetic fluctuations for a field applied in the easy magnetization axis c.

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I. INTRODUCTION

Magnetic quantum criticality has attracted major attention in recent years. The magnetic quantum phase transition is accompanied by an increase of quantum fluctuations, which induces strong deviations from the standard Fermi-liquid behavior of a metal [1]. New quantum phases such as magnetically mediated nonconventional superconductivity (SC) [2], nematic orders [3,4], or complex modulated magnetic states [5] appear near the critical pressure p_c.

The collapse of antiferromagnetism occurs in many heavy-fermion systems through a continuous second order quantum phase transition, and a superconducting dome has been observed in the vicinity of p_c. In contrast, in the case of a ferromagnet, the quantum phase transition at p_c changes near p_c to be first order in systems with low disorder [6,7] or may lead to complex order [8–10]. Even if magnetically mediated superconductivity has been predicted for weakly ferromagnetic systems already a long time ago [11], up to now only four systems show the microscopic coexistence of ferromagnetism and SC: UGe_2 [12], URhGe [13], UIr [14], and UCoGe [15]. While for UGe_2 SC appears under high pressure in the ferromagnetic state, URhGe and UCoGe are ferromagnetic and superconducting already at ambient pressure but with very different (p,T) phase diagrams. In URhGe the Curie temperature T_C increases with pressure at least up to 12.6 GPa [16], while ferromagnetism in UCoGe is suppressed under pressure [17,18].

In this paper we focus on the high pressure phase diagram of UCoGe which orders at $T_C \approx 2.7$ K with a tiny ordered moment of $M_0 \approx 0.05\mu_B$ at $p = 0$ [15]. Ferromagnetism is due to the $5f$ electrons from uranium which hybridize with light electrons to form heavy quasiparticle bands at low temperature [19]. Bulk superconductivity coexists with ferromagnetism below $T_{sc} \approx 0.6$ K. This provides a unique opportunity to study the link between the collapse of ferromagnetism and concomitant ferromagnetic fluctuations and the appearance of SC. Previously, the (p,T) phase diagram of UCoGe has been established up to 2.5 GPa by resistivity and magnetic susceptibility measurements [17,18,20,21] and p_c is located around 1 GPa. The ferromagnetic transition may be already first order at ambient pressure [22]. Here we will determine the (p,T) phase diagram of UCoGe by resistivity measurements up to 10.5 GPa down to 50 mK using a diamond anvil cell (DAC). Our experiments illustrate the link between the normal and SC phases on the field and pressure dependence of the ferromagnetic coupling. UCoGe provides the unique opportunity to study the interplay between SC and ferromagnetism in a case where SC is known to be robust through p_c and where T_C will approach T_{sc} without a strong first order collapse of T_C as it happens, for example, in UGe_2 [23].

In UCoGe SC survives deep inside the paramagnetic (PM) domain roughly up to $p \approx 4p_c$. Up to this pressure magnetic fluctuations are dominant and the transport shows a non-Fermi-liquid regime in the normal state above the SC dome. A Fermi-liquid ground state in zero field can be detected only for $p \gtrapprox 5$ GPa, basically when SC has fully collapsed. The magnetic field and pressure variations of ferromagnetism and its concomitant fluctuations have a strong impact on the unusual temperature dependence of the superconducting upper critical field H_{c2} with an upward curvature observed initially for both the hard b and the easy c axis at low pressure [21,24].

II. EXPERIMENTAL DETAILS

High quality single crystals of UCoGe (orthorhombic TiNiSi structure) were grown in a tetra-arc furnace by the Chzochralski method. Measurements under hydrostatic pressure were performed in a DAC ($p < 11$ GPa) and a piston cylinder cell ($p < 2.5$ GPa) with argon and Daphne oil 7373 as pressure transmitting media, respectively, ensuring very good pressure conditions [25]. The resistivity ratio of the sample

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used in the DAC after polishing was RRR = 28. A dilution fridge with a system allowing to change pressure *in situ* at low temperature was used to cool the sample down to 50 mK [26,27]. The pressure was determined *in situ* by fluorescence of ruby. An ac electrical current ($j < 100\ \mu$A) was applied in the *ab* plane. A low temperature transformer has been used to improve the signal to noise ratio of the measured voltage. A magnetic field up to 7 T was applied along the c axis. In a second experiment, the upper critical field H_{c2} for $H \parallel b$ has been determined by resistivity with a sample of similar quality using a piston cylinder cell up to 2.5 GPa and magnetic field up to 16 T.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the resistivity ρ of UCoGe versus temperature T for various pressures p in the DAC and (b) the (p,T) phase diagram extracted from these measurements which is in good agreement with previous reports [17,18,20,21]. At the lowest pressures the resistivity shows a kink at T_C, as indicated in Fig. 1(a) for $p = 0.4$ GPa. T_C decreases with pressure from $T_C \approx 2.5$ K at $p = 0$ GPa and coincides with the superconducting transition temperature at $p \approx 0.8$ GPa and $T_{sc} = 0.8$ K. The extrapolation of the pressure dependence of T_C down to zero temperature determines $p_c \approx 0.9$ GPa. The exact p dependence of T_C in the SC dome is still unclear [18,20] but from symmetry arguments the transition line should be first order [28]. For the second sample measured in the piston cylinder cell a similar phase diagram has been determined [see Fig. 1(b)] with slightly higher T_C and T_{sc}. Compared to previous reports [18,20,21] the value of T_C and also the exact location of p_c appears strongly sample dependent. However, each study shows that T_{sc} is maximum at or slightly below the pressure where it reaches T_C. The inset in Fig. 1(b) shows the transition width ΔT_{sc} as a function of pressure. At the maximum of T_{sc} the superconducting transition is sharpest ($\Delta T_{sc} = 40$ mK) and broadens with increasing pressure in the PM phase. T_{sc} is maximal at 0.65 GPa, below p_c. Zero resistivity can be observed up to 3.5 GPa. Taking the midpoint or the onset of the transition criteria SC vanishes around 4 or 4.5 GPa, respectively. Thus, SC survives in the PM regime far above p_c. The broadening of the superconducting transition under pressure is independent of sample quality and high pressure conditions and it is clearly related with the strength of $\partial T_{sc}/\partial p$.

The T dependence of the resistivity has been parametrized by fitting a power law $\rho(T) = \rho_0 + A_n T^n$ in the normal state. Here ρ_0 is the residual resistivity and A_n gives the strength of the temperature dependent scattering term. In the case of a Fermi liquid $n = 2$ and the coefficient A_2 is related to the average effective mass $m^\star$ of the quasiparticles. In multiband heavy-fermion materials $A_2 \propto (m^\star)^2$ is generally obeyed. To evaluate the pressure and temperature dependence of the exponent n we performed fits with the power law on a sliding window of 0.3 K in the normal state. This allows one to plot $n(p,T)$ in Fig. 2. The resistivity follows a T^2 behavior at ambient pressure and low temperature in the ferromagnetic state. Remarkably, $\rho(T)$ is linear around p_c above the superconducting transition T_{sc}. The T^2 behavior of the Fermi-liquid regime is recovered only above 5 GPa in the PM state far above p_c below T_{FL}.

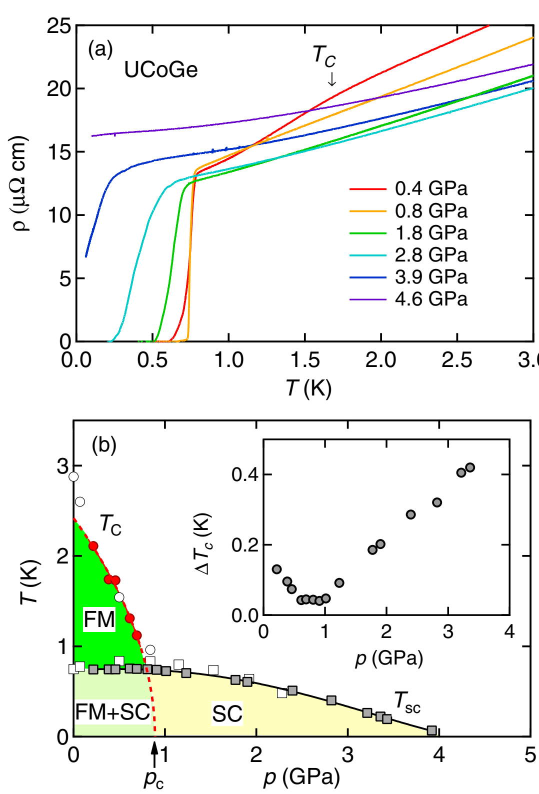

FIG. 1. (a) Temperature dependence of the resistivity in UCoGe for different pressures in the diamond anvil cell. For $p = 0.4$ GPa the Curie temperature T_C is denoted by the arrow. (b) (p,T) phase diagram of UCoGe. Closed (open) symbols correspond to the experiment in the DAC (piston cylinder cell). T_C vanishes at $p_c \approx 0.9$ GPa in both experiments. The superconducting transition T_{sc} is determined by the midpoint of the transition. The inset shows the pressure dependence of the width of the superconducting transition, ΔT_{sc}.

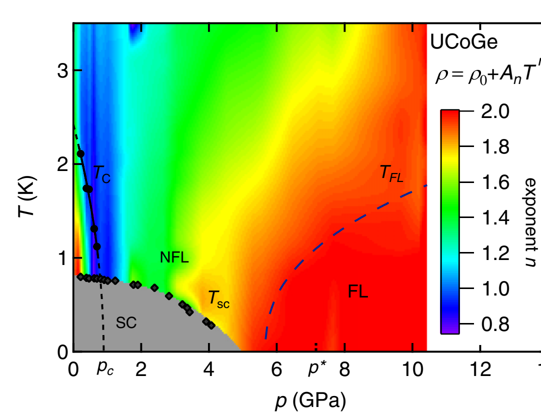

FIG. 2. Color plot of the resistivity exponent n as a function of temperature and pressure from fitting $\rho = \rho_0 + A_n T^n$ over a sliding window of 300 mK. T_C and the onset of the superconducting transition as a function of pressure are represented by solid circles and diamonds, respectively. Linear resistivity is observed around p_c. At high pressure Fermi-liquid behavior is recovered and the upper limit of the Fermi-liquid regime T_{FL} is indicated by the dashed line.

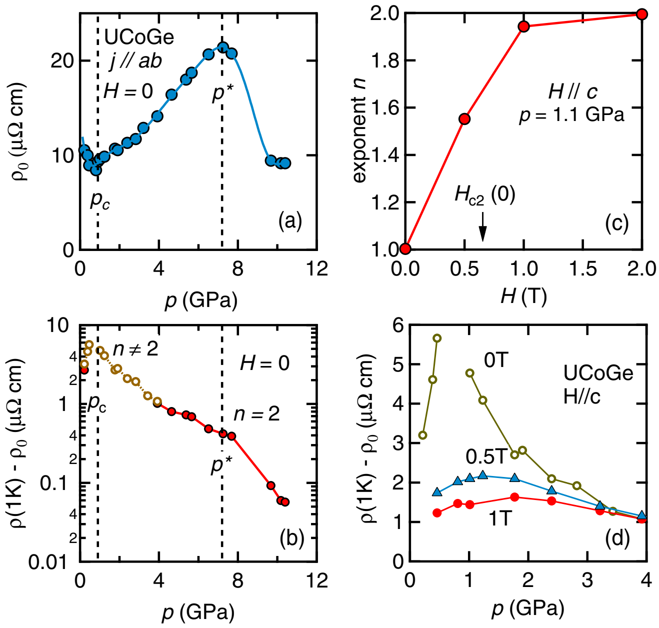

FIG. 3. (a) Residual resistivity ρ_0 as a function of pressure. ρ_0 shows a minimum at $p_c \approx 0.9$ GPa and a maximum at $p^\star = 7.2$ GPa. (b) $\rho(1\ \mathrm{K}) - \rho_0$ on a logarithmic scale as a function of pressure. (c) Field dependence of n slightly above p_c obtained from fits in the normal state for $T < 1.5$ K. (d) $\rho(1\ \mathrm{K}) - \rho_0$ for different magnetic fields as a function of pressure.

The unusual quasilinear T dependence of $\rho(T)$ observed close to the ferromagnetic instability is not expected in the spin-fluctuation theory. Theoretically, a $T^{5/3}$ temperature dependence is predicted in the quantum critical regime for a three-dimensional ferromagnet [29–33]. This is a consequence of an underlying quasiparticle scattering rate that varies linearly with the excitation energy E of a quasiparticle near the Fermi level. A $T^{5/3}$ behavior was reported in several itinerant ferromagnetic systems such as, e.g., $NiAl_3$ [34], URhAl [35], and U_3P_4 [36]. However, strong deviations from this $T^{5/3}$ have been reported for MnSi [37,38] and $ZrZn_2$ [39,40] where a $T^{3/2}$ dependence has been observed up to $3p_c$.

A T-linear resistance has been observed in different strongly correlated electron systems, such as high-T_c cuprate or iron-pnictide superconductors close to the optimal doping [41–43], in organic superconductors [44], in ruthenates [45], and also in several heavy-fermion systems, when these are close to quantum criticality such as $CeCoIn_5$ [46], $CeRhIn_5$ [47,48], or $YbRh_2Si_2$ [49]. For the antiferromagnetic heavy fermions, different theoretical scenarios have emerged to explain the unusual T dependence, such as a reduced dimensionality of the magnetic fluctuations [31,33,50], critical valence fluctuations [51,52], or fluctuations associated with the change of the electronic structure from the ordered to the PM state [53–55]. The specific case of ferromagnetic fluctuations remains to be treated.

The residual resistivity ρ_0 obtained from the fit is represented in Fig. 3(a) as a function of pressure. $\rho_0(p)$ shows a shallow minimum at $p_c \approx 0.9$ GPa. It increases up to $\rho_0 \approx 22\ \mu\Omega$ cm at $p^\star \approx 7.2$ GPa and decreases strongly with pressure. Finally it saturates around 9.5 GPa at $\rho_0 = 10\ \mu\Omega$ cm. $p^\star$ is independent of magnetic field at least up to 7 T. The low temperature inelastic electronic scattering contribution to the resistivity $\rho(1\ \mathrm{K}) - \rho_0$ at zero magnetic field as a function of pressure is represented in Fig. 3(b). $\rho(1\ \mathrm{K}) - \rho_0$ shows a clear maximum at p_c. The decrease of $\rho(1\ \mathrm{K}) - \rho_0$ with pressure gets stronger above $p^\star = 7.2$ GPa. The compressibility of UCoGe was computed by density functional theory [56] and experimentally determined by an x-ray scattering experiment under hydrostatic pressure up to 30 GPa [57]. At $p = 10$ GPa the volume of the unit cell is reduced by 3%. No structural transition was observed in the x-ray scattering experiment and only tiny anomalies in the lattice parameters as a function of pressure at $p^\star$ [57]. In the ferromagnetic state at $p = 0$ the valence of UCoGe is close to the U^{3+} configuration [58]. It was estimated at 3.2 by local-density approximation calculations for ambient pressure [59] and is expected to be closer to the U^{4+} under pressure. Thus the anomaly at $p^\star$ may be related to a weak valence crossover as observed in various Ce or Yb based heavy-fermion systems under pressure [51,52,60].

For a small magnetic field of 1 T along the c axis, slightly higher than the upper critical field H_{c2}, the exponent $n = 2$ is recovered [see Fig. 3(c)] and Fermi-liquid behavior appears in the entire pressure range. The low temperature electronic scattering, determined by $\rho(1\ \mathrm{K}) - \rho_0$, is plotted for different magnetic fields as a function of pressure in Fig. 3(d). Its acute enhancement at p_c is suppressed under magnetic field and a rather smooth pressure dependence is achieved. This indicates that the low field behavior of UCoGe is determined by magnetic fluctuations which are rapidly suppressed by a magnetic field applied along the c axis as has been shown at zero pressure by NMR [61], thermal transport [62], and specific heat experiments [63,64].

A striking property of UCoGe is its unusual upper critical field H_{c2} with the very strong anisotropy between H_{c2} for a field along the a (H_{c2}^a) or b (H_{c2}^b) hard axis and H_{c2} along the easy magnetization c axis (H_{c2}^c). While H_{c2}^a and H_{c2}^b exceed values above 20 T at ambient pressure, H_{c2}^c is as small as 1 T or even less, depending on the sample quality [21,65,66]. In addition, the temperature dependence $H_{c2}(T)$ for all directions is very unusual with a strong upward curvature.

$H_{c2}^b(T)$ for a field applied along the b axis is represented in Fig. 4(a) for different pressures. At ambient pressure $H_{c2}^b(T)$ shows a strong upward curvature with decreasing temperature. This measurement does not show the "S" shape of H_{c2} observed in [66]. It may be due to a small misalignment along the a axis of the sample inside the pressure cell. The unusual behavior of H_{c2}^b along the hard axis is related to the field decrease of T_C in the transverse magnetic field and the concomitant reinforcement of the magnetic fluctuations [66,67]. This mechanism is quite similar to that of the field reentrance of SC in URhGe [68,69]. The upward curvature of H_{c2}^b is reduced under pressure and vanishes around $p \approx 1$ GPa. Up to the highest pressure $p = 2.28$ GPa, $H_{c2}^b(T)$ is still nearly linear in T. $H_{c2}^b(0)$ decreases almost linearly with pressure from about $H_{c2}^b(0) \approx 25$ T at zero pressure to 7 T at 2.28 GPa. The upward curvature disappears when ferromagnetism collapses [70].

For $H \parallel c$, the field and pressure evolution of the inelastic low temperature electronic scattering contribution $\rho(1\ \mathrm{K}) - \rho_0$ shows that magnetic fluctuations are strongly suppressed with field. The relatively low value of H_{c2}^c and

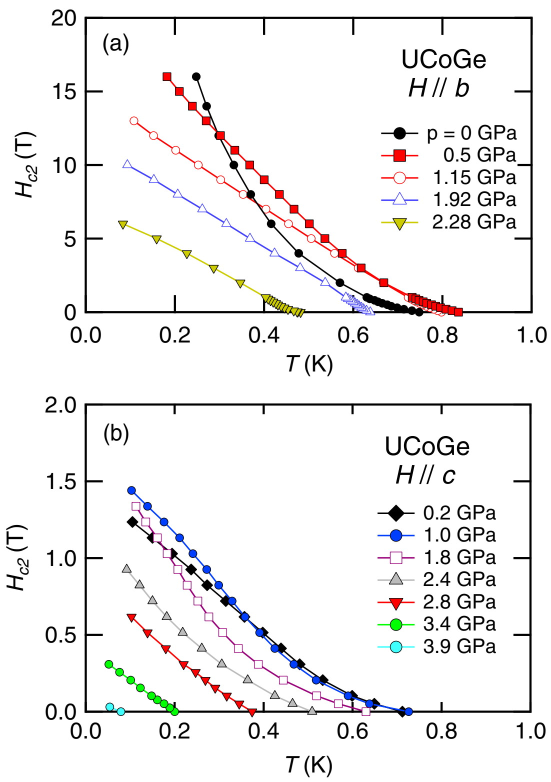

FIG. 4. (a) Temperature dependence of the upper critical field H_{c2} in UCoGe for different pressures for a field along the b axis. Measurements were performed on sample 2 in a piston cylinder cell. The midpoint of resistivity drop was chosen as criteria for the transition. (b) Temperature dependence of H_{c2} in UCoGe for different pressures for field along the c axis. Measurements were performed on sample 1 in the DAC.

the unusual upward curvature of H_{c2}^c was explained by several models including a magnetic field dependent pairing interaction [61,64,71,72]. In our experiment the unusual curvature of $H_{c2}^c(T)$ was observed in the whole pressure range of the superconducting dome. Inside the framework of equal spin triplet pairing, $H_{c2}(0)$ is governed by the orbital effect and so is proportional to $(m^\star T_{sc})^2$. Here we have to account for a field dependence $(m_H^\star)$ which reflects a field dependence of the pairing interaction. The value $T_{sc}(m_H^\star)$ at finite field is lower than $T_{sc}(m_0^\star)$ expected from zero magnetic field and this leads to the initial curvature of $H_{c2}(T)$. However, at 3.4 GPa far away from the quantum critical region, the ferromagnetic fluctuations have been strongly suppressed and their field dependence becomes weak. Therefore, at 3.4 GPa $H_{c2}^c(T)$ varies linearly with temperature and is much closer to the conventional orbital limitation with a field independent pairing than H_{c2} in the vicinity of p_c. This suggests that $H_{c2}^c(T)$ is driven by the suppression of pairing interaction under field.

IV. CONCLUSION

To summarize we determined the (p,T) phase diagram of UCoGe up to 10.5 GPa by resistivity measurements. T_C vanishes at $p_c \approx 0.9$ GPa and SC has been observed up to $4p_c$. Ferromagnetic fluctuations account for the unusual normal state transport properties observed up to 5 GPa as well as the pressure and field dependence of H_{c2}. The superconducting critical field H_{c2}^c for field along the c axis is determined by the pressure and field dependence of the ferromagnetic fluctuations which are suppressed for $H \approx 1$ T at $p = 0$. H_{c2}^b for transverse magnetic field changes drastically on crossing the ferromagnetic phase boundary below p_c. Entering in the paramagnetic regime above p_c H_{c2}^b recovers rapidly the conventional initially linear T dependence of the upper critical field. Finally, the crossover pressure $p^\star$ marks a change in the magnetic and electronic properties linked to the pressure dependence of the $5f$ occupation number and a smooth crossover from a strongly to weakly correlated regime.

Our experiment gives now a sound basis on the anisotropic SC response of H_{c2} on the static and dynamic components of the ferromagnetism. It is a unique example where the magnetic coupling is deeply modified (here for $H \parallel c$) in low magnetic fields. Close to p_c the comparable strength of T_C and T_{sc} leads to remarkable non-Fermi-liquid behavior which indicates the strong coupling of magnetism and superconductivity. This is remarkably different from other ferromagnetic superconductors driven to quantum criticality by the pressure (UGe_2) [73,74] of a magnetic field (URhGe) [75] where a Fermi-liquid regime is robust.

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