Upper critical field under hydrostatic pressure in UCoGe

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Abstract.
We report on the pressure dependence of the upper superconductivity critical field $H_{c2}$ of the ferromagnetic superconductor UCoGe for field along the $c$ axis, which is the easy magnetization axis. The surprising result is the upward curvature and strong enhancement of the upper critical field on cooling. This enhancement is even more pronounced above the critical pressure $p_c$ in the paramagnetic phase than in the ferromagnetic phase below $p_c$. This effect cannot be explained by models based on the suppression of pair building magnetic fluctuations under magnetic field. That points out either decrease of the pair breaking mechanism under field or fluctuations favorable for the Cooper pairing linked to the wing structure of ferromagnetism near the critical pressure, where ferromagnetism is suppressed.

1. Introduction
Ferromagnetic superconductors have attracted much attention due to the striking interplay between itinerant ferromagnetism and spin triplet superconductivity. Up to now four ferromagnetic superconductors have been reported : UGe$_2$\textsuperscript{1} and UIr\textsuperscript{2} under pressure and at ambient pressure URhGe \textsuperscript{3} and UCoGe \textsuperscript{4} which will be discussed in the present paper. UCoGe orders ferromagnetically below the Curie temperature $T_C = 2.7$ K. This ferromagnetic transition may be already weakly first order at ambient pressure \textsuperscript{5}. Under pressure $T_C$ is suppressed and vanishes at a critical pressure $p_c \approx 1$ GPa \textsuperscript{6,7,8}. At ambient pressure bulk superconductivity which coexists with the ferromagnetism sets in below $T_{sc} \approx 0.6$ K. However, $T_{sc}$ depends strongly on the sample quality. The microscopic coexistence of ferromagnetism and superconductivity has been shown by \textmu SR \textsuperscript{9} and NMR experiments \textsuperscript{5} at ambient pressure. NMR studies have also shown that superconductivity is coupled to longitudinal magnetic fluctuations along the easy magnetization axis $c$ \textsuperscript{12}. Superconductivity survives in the paramagnetic regime above $p_c$ \textsuperscript{6,7,8}. $T_{sc}$ is highest at $p_c$.

The upper critical field $H_{c2}$ in UCoGe is strongly anisotropic \textsuperscript{10,11}. While it is higher than 15 T for field in the $ab$ plane, $H_{c2}$ is around 0.5 T for field along the $c$ axis. An ”S shape” of $H_{c2}$ as a function of temperature was observed for field along the $b$ axis \textsuperscript{11}. For field along the $a$ and the $c$ axis, $H_{c2}(T)$ is nearly linear in temperature, with a small upward curvature \textsuperscript{10,11}. Previously the pressure dependence of $H_{c2}$ for $T \to 0$ has been published in Ref. \textsuperscript{13} and it has been found that $H_{c2}$ for $H \parallel c$ is increasing with pressure while it is decreasing for both directions perpendicular to $c$. In the present paper we report on the temperature and pressure variation of $H_{c2}$ for field along the easy magnetization axis $c$ on both sides of $p_c$, in the ferromagnetic and paramagnetic regime, respectively. $H_{c2}(T)$ versus temperature shows an upward curvature, which is surprisingly stronger above $p_c$ in the assumed paramagnetic state.
2. Experimental techniques

An UCoGe single crystal was grown by Czochralski method in a tetra-arc furnace. The residual resistivity and the $\rho(T = 300 K)/\rho(T \to 0 K)$ ratio are 16 $\mu\Omega \cdot$cm and 36 respectively. Resistivity measurements have been performed under high pressure in a piston cylinder cell with Daphne 7373 oil as pressure transmitting medium. The maximal applied pressure has been 1.9 GPa. A dilution fridge was used to cool the sample down to 50 mK. The ac electrical current ($j < 50 \mu A$) has been applied along the $a$ axis and magnetic field along the $c$ axis. A low temperature transformer has been used to improve the signal to noise ratio of the measured voltage. The pressure has been determined by the superconducting transition of lead measured by ac susceptibility. The measurement performed at 0.06 GPa will be considered as zero pressure reference in the following.

3. Results

The temperature dependence of the resistivity $\rho(T)$ of UCoGe for current $j$ along the $a$ axis is represented in fig. 1 for different pressures up to 1.9 GPa. At the lowest pressure $p = 0.06$ GPa, $\rho(T)$ shows a pronounced kink at $T_C = 2.1$ K and a superconducting transition at $T_{sc} = 0.55$ K is observed. $T_{sc}$ is defined at zero resistivity, $\rho = 0$. With increasing pressure $T_C$ decreases and can be observed up to 0.6 GPa. $\rho(T)$ at 1.1 GPa does not show any signature of a magnetic transition in the normal state above $T_{sc}$. The superconducting transition temperature $T_{sc}$ first increases with pressure and shows a broad maximum close to the critical pressure $p_c \approx 0.8$ GPa, where ferromagnetism vanishes. In concordance with the maximum of $T_{sc}$ under pressure, the width of the superconducting transition is sharpest close to the critical pressure and gets much broader at lower and also at higher pressures. Above $p_c$ several steps appear inside the superconducting transition, as shown in the inset for $p = 1.1$ GPa. This has been already reported in previous high pressure work [6] and shows the sensitivity of UCoGe to intrinsic and/or pressure inhomogeneities.

The pressure dependence of $T_C$ and $T_{sc}$ is shown in fig. 2. The Curie temperature $T_C$ at $p = 0$ and the critical pressure $p_c$ are smaller for the present sample than those in previous studies [6][7][8]. The pressure temperature phase diagram seems to be shifted to lower pressure. In addition we plot the pressure dependence of $H_{c2}$ at 50 mK in fig. 2. $H_{c2}$ at 0.06 GPa is 0.46 T, it is lower than values previously obtained on other samples[10]. While $T_{sc}$ is maximum around $p_c \approx 0.8$ GPa, $H_{c2}$
Figure 2. Pressure variation of Curie temperature $T_C$, superconducting transition temperature $T_{sc}$ and upper critical field $H_{c2}$ taken at $T = 50$ mK for $H \parallel c$. FM indicates the ferromagnetic phase, the hatched area gives the critical pressure region. We did not observe $T_C < T_{sc}$, and details of the phase diagram in this range are still not resolved.

Figure 3. (a) Upper critical field $H_{c2}$ versus temperature under hydrostatic pressure for various pressures. Field is applied along the $c$ axis. b Renormalized $H_{c2}/T_{sc}$ is plotted versus dimensionless temperature $T/T_{sc}$. $T_{sc}$ was determined with resistivity measurement and the criterion $\rho = 0$. Independent of pressure $H_{c2}(T)$ shows a positive curvature, only at the lowest temperature $H_{c2}(T)$ starts to change the curvature.
Figure 4. Field variation of square root of $A$ coefficient. Resistivity was fitted with a $T$ square law $\rho = \rho_0 + AT^2$ on a small temperature range just above the superconducting transition to get these results. The values of $\sqrt{A}$ were renormalized by the value at zero field and $p = 0.06\ \text{GPa}$.

The inset shows pressure dependance of $\sqrt{A}$ at zero field.

low field, whereas in the paramagnetic phase $\sqrt{A}$ decreases more smoothly with field. Above 2 T the evolution of $\sqrt{A}$ with field gets weaker. The pressure variation of the normalized $\sqrt{A}$ at zero field shown in the inset of fig.4 seems to have a smooth maximum below 1 GPa. This is consistent with previous observations on polycrystalline samples [6].

4. Discussion

The most surprising property of the upper critical field $H_{c2}$ of UCoGe is its extremely strong anisotropy, i.e. along the $a$ axis for perfect alignment $H_{c2} > 25$ T, while along the $c$ axis a field of $H = 1$ T is sufficient to destroy superconductivity [11]. In both directions $H_{c2}$ shows an upward curvature indicating that the Pauli depairing mechanism is negligible and $H_{c2}$ is only limited by the orbital effect, in agreement with the proposed equal spin pairing state in this ferromagnetic superconductor [14]. In a simple approach, one would expect a strong anisotropy in the Fermi velocity which is inversely proportional to the effective mass. The anisotropy of the effective mass under field was estimated by resistivity [11] and specific heat measurements [15]. However, it is not strong enough to explain the huge anisotropy of $H_{c2}$.

The upward curvature of $H_{c2}$ was first observed at ambient pressure for field along the $a$ axis with a kink at 0.35 K [10]. The authors of Ref. [10] suggested it may be due to multiband superconductivity. However, multiband superconductivity is not sufficient to explain the strong temperature dependence of $H_{c2}$ in the low temperature region for $H || c$.

Recently it has been shown on the basis of NMR experiments that a key ingredient to understand the unusual $H_{c2}$ of UCoGe is the fact that longitudinal ferromagnetic fluctuations along the $c$ axis induce the superconductivity in this compound [12]. These fluctuations show a $1/\sqrt{H}$ dependence for $H || c$. Taking these fluctuations and their suppression for $H || c$ into account, the model calculation in the ferromagnetic domain based on a linearized Eliashberg theory in ref. [16] reproduces the experimental observations of (i) the strong anisotropy of $H_{c2}$, (ii) the strong suppression of $H_{c2}$ for field along the $c$ axis, and (iii) the upward curvature of $H_{c2}$ for the so-called A state, which corresponds to a gap function with point node symmetry.

However, there is no consideration on the behavior of $H_{c2}$ along the $c$ axis in the paramagnetic state, or even when the exchange field becomes small. At least it is quite surprising that in the
assumed paramagnetic domain above $p_c$, where spin fluctuations have decreased, we do not recover the common variation of $H_{c2}(T)$, i.e. a negative curvature.

In a very simple picture it follows from the upward curvature of $H_{c2}$ that the pair breaking effect is reinforced with increasing temperature or reduced by magnetic field. A possible candidate for this pair breaking effect would be spin flip scattering. It was shown that the suppression of spin flip scattering with magnetic field can be responsible for a positive curvature in $H_{c2}$ curves [17]. Theories about spin flip scattering were developed for $s$ and $d$ wave paired states, but may be valid for equal spin Cooper pairs.

In fig. 3 we have shown that the upward curvature of $H_{c2}$ persists and gets even stronger far above the critical pressure in the paramagnetic state. For a second order phase transition a strong enhancement of the critical ferromagnetic fluctuations is expected at the critical pressure. Here this enhancement is rather weak. In the generic phase diagram of an itinerant ferromagnet close to the quantum critical point the ferromagnetic transition changes from second to first order and under magnetic field a wing structure appears [25]. Therefore we may expect that the shape of the $(p, T, H)$ phase diagram of UCoGe shows a wing structure similar to those of the heavy fermion ferromagnet UGe$_2$ [19] or UCoAl [20]. The increase of $H_{c2}$ with pressure above $p_c$ might be the consequence of such a ferromagnetic wing structure. In this case the critical region would be shifted to higher pressure under magnetic field. So it might explain why $H_{c2}(p)$ has its maximum far above the critical pressure $p_c$. The strong upward curvature in the paramagnetic phase may be related to the wing structure. Ferromagnetic fluctuations would be enhanced under magnetic field due to the proximity of the critical region which is shifted to a non-zero field region and provokes the strong upward curvature. However, recently it has been noticed that the critical region may be much more complicated and inhomogeneous phases may occur at the border of the itinerant ferromagnetism [24][23].

In a simple framework the effective mass $m^* = m_B + m^{**}$ is the sum of the renormalized band mass $m_B$ and the correlated mass $m^{**}$. According to the orbital limit $H_{c2}$ is proportional to $(m^*T_{sc}^2)$. Superconductivity is sensitive to the ratio between correlation mass and effective mass $\lambda = m^{**}/m_B$ [21]. The open question is the field and pressure dependence of $m_B$. So the upward curvature may also results from an opposite behavior of the band mass and the correlation mass under magnetic field.

5. Conclusion

The temperature and pressure variation of the upper critical field of UCoGe was measured in the configuration $H \parallel c$. An upward curvature was observed in the temperature dependence of $H_{c2}$ at ambient pressure. This upward curvature is surprisingly enhanced with pressure. These results may be explained by a model calculation of the upper critical field dominated by the damping of ferromagnetic fluctuations under field along the $c$ axis [16]. The upward curvature may be due to a pair breaking effect reduced by the field or enhanced by the temperature. The challenge is now to identify this effect. A further possible explanation of the observed effect may be given by the ferromagnetic wing structure in magnetic field due to the first order nature or an even more complex phase diagram close to $p_c$ [24][23]. Unfortunately, up to now it was not possible to observe directly a magnetic phase where $T_{sc}$ overcomes the Curie temperature and the wing structure might be inside the superconducting dome. In this case it would be difficult to follow the ferromagnetic wings. Similar measurements of $H_{c2}$ with magnetic field along the $a$ or the $b$ axis are needed to fully understand the upper critical field of UCoGe.

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References

[1] Saxena S S, Agarwal P, Ahilan K, Grosche F M, Haselwimmer R K W, Steiner M J, Pugh E J, Walker I R, Julian S R, Mouthou P, Lonzarich G G, Huxley A, Sheikin I, Braithwaite D and Flouquet J 2000 Nature 406 587
