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Anisotropy and pressure dependence of the upper critical field of the ferromagnetic superconductor UGe₂

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The temperature and pressure dependence of the upper critical field, H_{c2} , of the ferromagnetic superconductor UGe₂ is reported for fields applied along all three crystallographic axes. For fields parallel to the easy magnetic a axis, the relationship between an unusual reentrant behavior of H_{c2} and a field-induced transition associated with a change in the electronic density of states is reviewed. For transverse field directions a significant evolution in the behavior of H_{c2} with pressure is found. As the pressure is decreased the dependence of H_{c2} on temperature for fields along the crystal's c axis acquires a positive curvature that extends from the critical temperature, T_c , down to almost the lowest temperature measured ($T_c/10$) where H_{c2} exceeds the usual weak coupling paramagnetic and orbital limits.

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UGe₂ is a material in which superconductivity and itinerant ferromagnetism have been demonstrated to coexist^{1,2} in single crystals with a Curie temperature that is substantially higher than the superconducting transition temperature ($T_c \sim 0.8$ K and $T_{\text{Curie}} \sim 35$ K at 12 kbar). Both states exist over a relatively wide range of applied pressures (11–16 kbar) and apparently disappear simultaneously at an upper critical pressure, $P_c \sim 16$ kbar.

Theoretically, the possibility of finding spin triplet superconductivity near to a ferromagnetic quantum critical point was pointed out a long time ago.^{3,4} However, the original scenario considered the divergence of magnetic fluctuations close to a second order quantum critical point, while it has been argued^{2,5} that in UGe₂ the ferromagnetic transition becomes first order just below P_c . Although the associated ferromagnetic fluctuations could still favor the formation of spin triplet Cooper pairs, the maximum interaction obtainable is less than for a second order transition. Experiments, however, suggest that there could be a further phase boundary within the ferromagnetic state.^{5,6} Even though the associated order parameter has not been identified, the observation that the superconducting critical temperature is highest near to the critical pressure ($P_x \sim 12.5$ kbar) necessary to suppress the transition suggests that it is intimately related to the pairing interaction. Whatever the pairing mechanism, odd-parity spin triplet superconductivity might be expected since the spin majority and minority Fermi-surfaces are very strongly split in the ferromagnetic state. Since time invariance is already broken by the ferromagnetic order, the superconducting state is necessarily nonunitary,⁷ as suggested from measurements of the flux-flow resistivity.² This situation resembles that of the A1 phase of superfluid ³He. Differences from the case of ³He are expected since apart from the obvious change to charged particles, the presence of a strong crystal field can modify the orientation and symmetry of the order parameter.

Measurements of the upper critical field provide a powerful method to help confirm whether the superconductivity is

indeed spin triplet. Such measurements have previously led to a hypothesis of triplet pairing in the heavy fermion superconductor UPt₃ (Refs. 8–11) and in the quasi-one-dimensional organic compound (TMTSF)₂PF₆.^{12,13}

Our measurements of H_{c2} along the three principal crystallographic directions of orthorhombic UGe₂ cover three different pressure ranges: just below and above P_x and close to P_c . The measurements were performed in a nonmagnetic piston-cylinder-type pressure cell. H_{c2} was determined from the electrical resistivity measured by the usual four terminal method with an ac current (10 or 100 μ A at 11.7 Hz) passed along the length of the sample and parallel to its a axis. H_{c2} was taken as the midpoint of the superconducting transition crossed by either changing the field (several examples are shown in the inset of Fig. 1) or the temperature; our conclusions do not depend on the particular choice of criteria, except for $H\parallel c$ at 15 kbar, where there was a strong dependence on field history, which will be described later. H_{c2} was measured parallel to the a and b directions at exactly the same pressures without heating the cryostat above liquid helium temperatures. To make measurements with $H\parallel c$ it was necessary to warm the cryostat and pressure cell to room temperature and change the magnet. This thermal cycle gave rise to a small change of pressure (determined by measuring the superconducting transition of a tin strip in the cell prior to application of any field), that was sufficient to change slightly the superconducting critical temperature of the UGe₂ crystal. To facilitate comparison we therefore show in Fig. 1 the temperature dependence of the upper critical field normalized to T_c for all three principal orientations of the magnetic field plotted against T/T_c for the three distinct pressure ranges considered. The corresponding values of T_c are given in the figure caption, while the zero field transitions have been reported elsewhere.¹⁴

We discuss first H_{c2}^a , the upper critical field for a field applied parallel to the easy magnetization direction (a axis), since a field applied along this direction gives rise to distinct

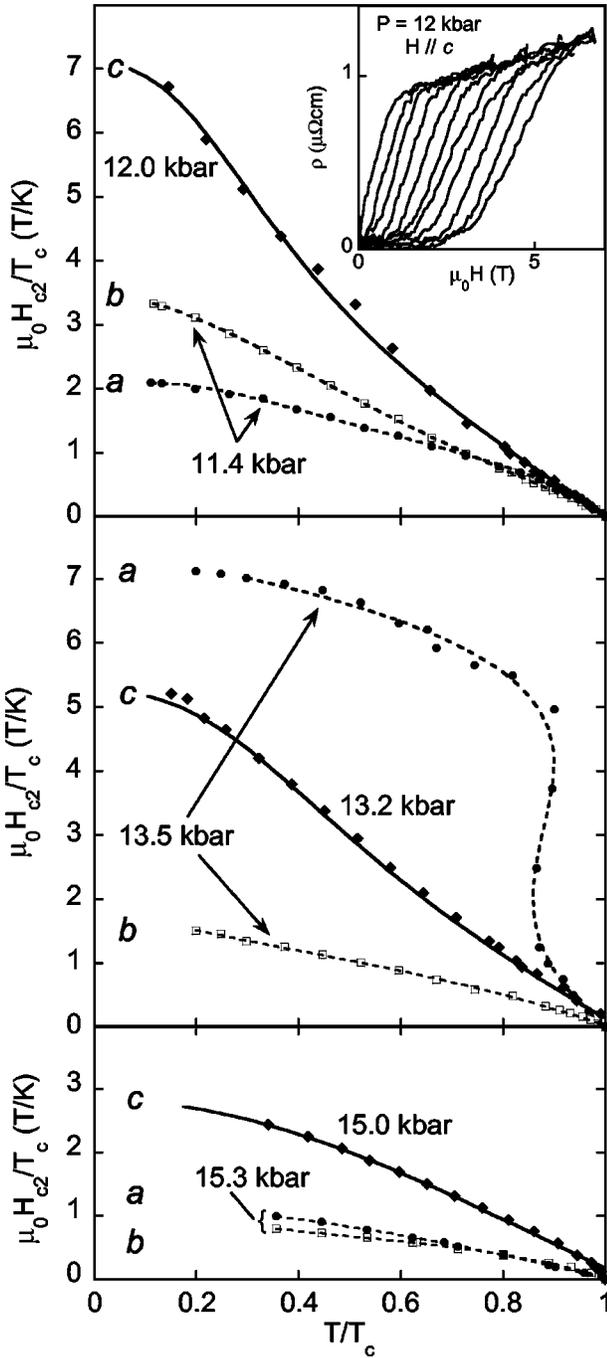


FIG. 1. The upper critical field normalized to the superconducting transition temperature, H_{c2}/T_c , is plotted against temperature, T/T_c , for applied fields parallel to the *a*, *b*, and *c* axes. The top and middle panels are, respectively, for pressures slightly below and above P_x , while the bottom panel corresponds to a pressure close to the critical pressure to suppress ferromagnetism (and superconductivity), P_c . The critical temperatures at the various pressures are 0.757, 0.685, 0.466, 0.403, 0.258, and 0.225 K at 11.4, 12, 13.2, 13.5, 15, and 15.3 kbar. The solid lines are fits to a strong coupling calculation described in the text, while the dashed lines serve only to associate the points. The inset shows resistivity versus magnetic field at temperatures from 0.1 to 0.6 K in steps of 0.05 K at 12 kbar for $H\parallel c$.

phenomena in the normal state, and therefore requires special consideration. In zero magnetic field, just below 12.5 kbar there is a sharp transition in the temperature dependence of the normal state resistivity and magnetization at a temperature T_x [the latter is also visible in a neutron study at 9.5 kbar where $T_x=15$ K (Ref. 2)]. T_x decreases with pressure and vanishes at $P_x=12.5$ kbar. There is a drop in the residual resistivity (20%), a sharp (300%) increase in the coefficient of the temperature dependent part of the resistivity (which remains quadratic at low temperature),² and an increase in the low temperature specific heat¹⁵ as P is increased through P_x . These observations suggest that the transition marks a change of phase to a state with a higher electronic density of states above P_x . Although such observations are consistent with the suppression of a supposed charge- and spin-density wave (CDW/SDW) with pressure and temperature, no direct evidence (e.g., from neutron scattering) has been forthcoming to confirm such a hypothesis. A CDW necessary implies a SDW because of the large splitting between the majority and minority spin Fermi surfaces in the ferromagnetic state. In this case an increase of magnetization $M(T)$ below T_x could arise as a result of mode coupling between M , $S_{\vec{Q}}$ and $N_{-\vec{Q}}$, where $S_{\vec{Q}}$ and $N_{-\vec{Q}}$ are the SDW and CDW order parameters with ordering vectors $\pm\vec{Q}$.¹⁶

The consequence of the above for H_{c2}^a follows from the sensitivity of T_x to a magnetic field parallel to the easy axis. The superconducting transition temperature is determined in a strong coupling formalism by the spectral weight of the Bose excitations that mediate the pairing interaction. In a simplistic model this can be described by a Lorentzian function, $g^2F(\omega)=\lambda(\Gamma/\pi)[\omega/(\omega^2+\Gamma^2)]$. Then the number of parameters is reduced to three: a coupling constant, λ , the Fermi energy (assumed constant), and the Lorentzian width, Γ , of this distribution. With the approximation that the same Bose spectrum accounts for the entire renormalization of the electronic density of states from its value at $P=0$, $\lambda(P)$ is simply proportional to the excess specific heat as a function of pressure, which can be estimated from the data of Tateiwa *et al.*¹⁵ $\Gamma(P)$ is then determined by the pressure dependence $T_c(P)$. To understand the measurements of $H_{c2}^a(T)$, P is replaced by $\tilde{P}(H)$ obtained by inverting the relationship $T_x(P,H)=T_x(\tilde{P},0)$ based on the field dependence of T_x at 11 and 13.5 kbar and the zero field dependence of $T_x(P)$.¹⁴ The upper critical field at different pressures can then be calculated. The results of such a calculation are shown in Fig. 2. Despite the oversimplistic nature of the above analysis, it is apparent that the strange discontinuous behavior of H_{c2}^a for P just above P_x seen experimentally at 13.5 kbar is qualitatively reproduced by this model and can therefore be attributed to a rapid evolution of Γ and λ as the critical condition $T_x\rightarrow 0$ is approached by application of a field. By the same argument there should also be an enhancement of H_{c2}^a at 15.3 kbar ($P>P_x$) and a small depression of H_{c2}^a at 11.4 kbar ($P<P_x$) at low temperatures. This is consistent with the data: at 11.4 kbar $H_{c2}^a(T)$ has a stronger curvature than $H_{c2}^b(T)$ at low temperature and $H_{c2}^a(0)<H_{c2}^b(0)$, whereas the situation is reversed at 15.3 kbar. No evolution

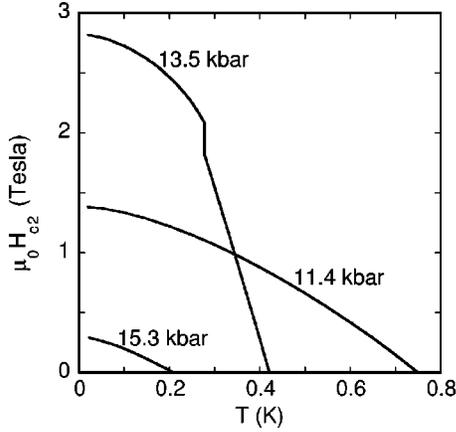


FIG. 2. $H_{c2}(T)$ for $H||a$ calculated taking into account the field/pressure dependence of the transition at T_x (see text for explanation).

of T_x for applied fields perpendicular to the easy axis has been observed and the above considerations therefore will not modify the form of H_{c2} for these directions.

After accounting for the particular effects described in the previous paragraph, the upper critical field H_{c2}^c is seen to be systematically larger than H_{c2}^b and H_{c2}^a (except perhaps very close to T_c). In our data the critical field refers to the applied field which could differ from the total field by up to $\mu_0 M < 0.2$ T where M is the sample magnetization in the ferromagnetic state. The additional field due to the magnetization of the sample can explain an apparent down turn of the H_{c2} curves for $H < 0.1$ T close to T_c . To avoid this complication all of the following analysis is restricted to data in fields above 0.1 T, where to a first approximation the difference between the applied field and total field can be neglected. The anisotropy of dH_{c2}/dT close to T_c can then be expressed in terms of an anisotropic effective mass tensor $m_{ij}^{-1} \propto \langle v_{fi} v_{fj} \Delta^2 \rangle$ such that $(dH_{c2}/dT)_x / (dH_{c2}/dT)_y \propto \sqrt{m_{yy}} / \sqrt{m_{xx}}$ (where x, y are principal axis). The mass tensor depends on the Fermi-surface averages (denoted by $\langle \rangle$) of different components of the Fermi velocity (v_{fi}) and the superconducting gap, Δ . The Fermi-velocity anisotropy can eventually be estimated experimentally from the anisotropy of the temperature dependence of the resistivity in the normal state measured with currents along different crystal directions. From our data the limiting value of dH_{c2}/dT (deduced from the data above 0.1 T) decreases strongly with pressure. The corresponding coherence lengths, ξ , at 11–12 kbar are all within 25% of 100 Å. While the anisotropy between the a and b axis is hardly changed at 15 kbar, a marked anisotropy with respect to the c axis becomes apparent with $\xi_a \approx 210$ Å, $\xi_b \approx 140$ Å, and $\xi_c \approx 700$ Å. In addition at 15 kbar there is a large hysteresis in H_{c2}^c on cycling the field. Figure 3 shows that the transition to the normal state in increasing field occurs at a lower field than in a decreasing field. This suggests that there are differently oriented magnetic domains with different apparent H_{c2} 's depending on the relative alignment of the applied field to the domain direction. In the low pressure magnetic structure the ordered moments are, however, locked to the a axis, whereas the

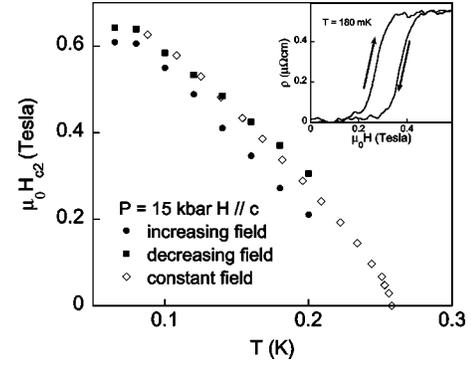


FIG. 3. $H_{c2}(T)$ for $H||c$ at 15 kbar determined from both temperature and field sweeps is shown to illustrate the unusual hysteresis (see the inset) between the increasing and decreasing field measurements.

hysteresis for $P \approx P_c$ occurs only for $H||c$. This hints that close to P_c , a rotation of the moment in the ac plane might occur. The unusual hysteresis is not visible for the other field directions or at lower pressure.

The curves for $H_{c2}^c(T)$ are also remarkable at lower pressures, where they show an unusual positive curvature and $H_{c2}^c(0)$ is much larger than given by both the usual weak coupling orbital and paramagnetic limits. A positive curvature of H_{c2} has been seen in other superconductors, notably in $Tl_2Ba_2CuO_6$,¹⁷ the organic material $(TMTSF)_2PF_6$,¹² and in the heavy fermion material UBe_{13} ,¹⁸ although for $Tl_2Ba_2CuO_6$ H_{c2} does not exceed the paramagnetic limit. $(TMTSF)_2PF_6$ is extremely anisotropic and the low dimensionality of the Fermi surface in high fields is proposed to lead to the reestablishment of superconductivity above the usual orbital limit,¹³ although the exact form of the H_{c2} curve is predicted to be very sensitive to the purity of the material.¹⁹ The anisotropy of H_{c2} manifest in this and other organic materials where such explanations might be appropriate is however orders of magnitude larger than for UGe_2 . Therefore, the explanation put forward for a strong curvature of H_{c2} in the cubic material UBe_{13} appears more relevant to the present case. Thomas *et al.*¹⁸ proposed that in UBe_{13} the coupling parameter, λ , is extremely large which can give both a large $H_{c2}(0)$ and a positively curved temperature dependence. H_{c2} is required (as observed in UBe_{13}) to saturate and have negative curvature in this model at very low temperatures in contrast to the models that apply to $(TMTSF)_2PF_6$. The details of the form of H_{c2} calculated for strong coupling depend upon the details of the Bose spectrum and its coupling to the electrons to give pairing. However, a simple estimate in an extreme case where the spectrum is a δ function at finite frequency (an alternative form to the Lorentzian centered at zero frequency considered earlier) can be made as outlined by Bulaevskii²⁰ in terms of the coupling parameter λ , where $\lambda \ll 1$ again gives the weak coupling BCS result.²¹ In such a model with isotropic pairing the form of the curve is completely determined by the low temperature limiting field and the slope of the upper critical field near to T_c . The calculation should be regarded as illustrative, serving only to give a taste of the forms that might be

compatible with a more realistic pairing spectrum and taking into account the gap anisotropy. With Bulaevskii's model²⁰ we cannot explain the anisotropy of H_{c2} along different directions (different λ 's would be required) but we can get an idea of how the coupling strength might evolve with pressure. Fits to the experimental data for H_{c2}^c are shown in Fig. 1 as solid lines, where the values of the parameter λ are 14, 7, and 1.7 at 12, 13.2, and 15 kbar, respectively. Unlike for UBe_{13} , good fits are obtained only if Pauli limiting is excluded in the calculation as would be appropriate for certain spin triplet states. For a spin triplet state another higher limiting field occurs due to the effect of the field on the nonzero orbital momentum of the Cooper pairs,²² $H_l = (m^*/m)H_p$ (where m^*/m is the effective electron mass compared to the bare mass and H_p is the usual paramagnetic limiting field). Since m^*/m is at least 3 in the pressure range where superconductivity is observed, this limit is not quite attained.

In this paper we have reported that at pressures close to P_x , H_{c2} significantly exceeds both paramagnetic and orbital limits that would apply to a weakly coupled *s*-wave superconductor. Further, the paramagnetic limit appears to be exceeded even for the case of strong coupling. The lack of paramagnetic limitation argues against a hypothesis of *s*-wave pairing, but is consistent with triplet superconductiv-

ity. Although the necessary parameters to make complete calculations including the anisotropy of the pairing interaction and Fermi surface are not yet established, it is clear that the positive curvature of $H_{c2}(T)$ contrasts with the usual dependence for phonon mediated superconductors, but bears some similarity with the heavy fermion material UBe_{13} , suggestive of a very strong coupling. Thus although phonons could still contribute to pairing, other pairing interactions related to the strongly correlated electronic state might play a significant role. The strength of the coupling appears to decrease rapidly and the coherence lengths become larger as one moves away from the critical pressure P_x . This relationship is most dramatically demonstrated by the results for H_{c2}^a where the extreme sensitivity of T_x to field parallel to the easy axis gives rise to an unusual reentrant behavior of the superconductivity. Finally, close to P_c an unusual hysteresis of H_{c2}^c occurs, which suggests that the applied field required to suppress superconductivity is different in differently oriented magnetic domains. The observation of hysteresis only for fields parallel to the *c* axis, however, is surprising given the Ising-like anisotropy of the magnetism at lower pressures where the ordered moments have been shown to be aligned to the *a* axis.

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