Acute enhancement of the upper critical field for superconductivity approaching a quantum critical point in URhGe

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When a pure material is tuned to the point where a continuous phase-transition line is crossed at zero temperature, known as a quantum critical point (QCP), completely new correlated quantum ordered states can form^{1–7}. These phases include exotic forms of superconductivity. However, as superconductivity is generally suppressed by a magnetic field, the formation of superconductivity ought not to be possible at extremely high field⁸. Here, we report that as we tune the ferromagnet, URhGe, towards a QCP by applying a component of magnetic field in the material's easy magnetic plane, superconductivity survives in progressively higher fields applied simultaneously along the material's magnetic hard axis. Thus, although superconductivity never occurs above a temperature of 0.5 K, we find that it can survive in extremely high magnetic fields, exceeding 28 T.

The formation of superconducting states close to magnetic quantum critical points (QCPs) first came to prominence in studies of the antiferromagnets CeCu₂Ge₂ (ref. 1), CeIn₃ and CePd₂Si₂ (ref. 2) and more recently CeRhIn₅ (refs 5,6). Theoretically, on tuning a material towards a QCP by application of pressure or magnetic field, the strength of the magnetic fluctuations that potentially bring about superconductivity increases. On approaching a ferromagnetic QCP, longitudinal magnetic fluctuations promote the formation of unconventional spin-triplet superconductivity^{9–11}. Theories predict *d*-wave superconductivity close to QCPs involving antiferromagnetic states. The evolution of the superconductivity as a material is tuned more closely to the field or pressure of the underlying QCP depends on the balance between the weights of fluctuations that are pair forming and pair breaking¹². The critical temperature for superconductivity, T_s , is predicted to saturate⁹ or may even decrease¹⁰. Experimentally, superconducting states in antiferromagnetic systems have been more extensively studied than in ferromagnets and measurements show T_s to have a dome-shaped pressure dependence crossing the underlying QCP^{2,5}. However, recent work indicates that the pressure–temperature phase diagrams of antiferromagnetic systems might be more complex than previously thought^{5,6,13}. As it is difficult to vary pressure continuously at low temperature, other properties of the superconducting state, such as the critical field to suppress superconductivity in CePd₂Si₂ (ref. 14), lack measurements at a sufficient number of pressures to discern how they vary approaching a QCP. For URhGe, the QCP is ferromagnetic rather than antiferromagnetic and can

be approached by applying a magnetic field, which can be swept continuously.

In zero magnetic field, URhGe undergoes a ferromagnetic transition at $T_{\text{Curie}} = 9.5$ K. Below this temperature, the ordered magnetic moment is aligned parallel or antiparallel to the crystallographic *c* axis¹⁵. Magnetic fields that correspond to opposite directions of the *c*-axis moment are separated in low magnetic field by a plane of first-order transitions that is crossed when the *c*-axis component of the magnetic field changes sign. The temperature above which this first-order transition plane ceases defines a line along which the phase transition is continuous. The schematic temperature–field phase diagram for fields throughout the *bc* plane is shown in Fig. 1. Close to a field of 12 T, the plane of first-order transitions bifurcates into two spurs. There is a corresponding tricritical point (TCP) at low, but non-zero temperature, where the continuous-transition line splits. The points where the continuous-transition line falls to zero temperature are QCPs⁴. URhGe is also close to a quantum TCP; such a point would occur if the TCP could be suppressed to zero temperature by varying some further tuning parameter. The phase diagram in Fig. 1 resembles that predicted theoretically for a ferromagnet driven towards a QCP (ref. 16) with the magnetic field applied along the *b* axis in the present case replacing pressure in the theoretical diagram.

A consequence of the low-temperature TCP is that fluctuations develop with a similar strength at very low temperature at all points along the length of the first-order spurs. Experimentally, this is manifested by the peaks in the temperature-dependent part of the resistivity having comparable amplitudes crossing the weak first-order transition at different points⁴. Closer than 0.25 T to the spurs, there is an additional sharp peak in the normal-state resistivity, which is independent of temperature (up to 800 mK). We have attributed this effect to extra scattering of charge carriers from magnetic domains that form close to the first-order transition⁴. As the temperature is reduced to 50 mK, superconductivity occurs both in low applied magnetic fields below 2 T and at high applied fields engulfing the QCPs and the regions where the first-order transition is weak (Fig. 1). The magnitude of the fields at which superconductivity occurs relative to the superconducting transition temperature in both the low-field and high-field pockets implies that equal spins must be paired in contrast to opposite spin pairing in conventional superconductivity^{4,17}.

Figure 1 The schematic field-temperature phase diagram of URhGe for magnetic fields in the bc plane. The magnetization changes discontinuously on crossing the first-order transition surface shaded green that ends at finite temperature along a continuous-transition line. The continuous-transition line bifurcates at a TCP and falls to zero temperature at QCPs shown as blue points. The low-temperature resistivity (of the sample studied) at 50 mK for fields in the bc plane is shown by the colour shading on the lower face of the axis frame: yellow corresponding to a resistivity of approximately $5 \mu\Omega$ cm and red to superconductivity (zero resistivity).

Figure 2 shows our measurements with magnetic fields applied at different angles, γ , from the b axis towards the a -axis direction. The persistence of the sharp peak in the resistivity at 500 mK shows that the magnetic transition remains weakly first order as γ is increased. The field at which the peak occurs, $H_R(\gamma)$, is well modelled by the relation $H_R(\gamma) = H_R(0)/\cos(\gamma)$. For fields in the ab plane, the transition field is therefore crossed when the component of the field along the b axis is equal to $H_R(0)$ (henceforth abbreviated to H_R) irrespective of the a -axis field. Application of a magnetic-field component parallel to the a axis for a constant b -axis field therefore enables us to determine the critical field for superconductivity at a constant offset from the QCP.

Within our experimental resolution, the angular dependence of the critical field for the low-field pocket of superconductivity at low temperature (denoted as H_{sc1}) follows the standard form for an anisotropic superconductor (with an orbitally limited critical field),

$$H_{sc1}(\gamma) = \frac{\phi_0}{2\pi\xi_c} \frac{1}{\sqrt{\xi_a^2 \cos^2(\gamma) + \xi_b^2 \sin^2(\gamma)}}. \quad (1)$$

$H_{sc1a} = \phi_0/2\pi\xi_c\xi_b = 2.53$ T, $H_{sc1b} = \phi_0/2\pi\xi_c\xi_a = 2.07$ T and $H_{sc1c} = \phi_0/2\pi\xi_b\xi_a = 0.69$ T are the critical fields along each of the crystal axes for the sample studied, determined in separate more accurate measurements (ξ_a , ξ_b and ξ_c are the coherence lengths along the different crystal axes and ϕ_0 is the flux quantum). An equation of the same form as equation (1) is derived from the Ginzburg–Landau theory and is applicable in general close to the zero-field superconducting transition temperature, T_s . For URhGe, we can specialize to the case of no Pauli limiting¹⁷. In the absence of Pauli limiting, although the coherence lengths depend on temperature, for simple ellipsoidal Fermi surfaces (which can account for an anisotropic electronic effective mass) and isotropic s -wave superconductivity, the anisotropy of the coherence lengths and the angular dependence of the critical field is unchanged at low temperature from that close to T_s (ref. 18). Different factors can cause the anisotropy to be temperature dependent or modify the angular dependence of the critical field at low temperature¹⁹.

Figure 2 Resistivity measurements for fields rotated through different angles, γ , from the b axis in the ab plane. **a**, The measured resistance plotted against magnetic field for fields applied at $\gamma = 10^\circ$ at 45 and 500 mK and also at 45 mK for $\gamma = 66^\circ$, which is the geometric inset with the highest onset field for superconductivity. The geometric inset defines the angle, γ , of the field direction relative to the directions of the a and b crystal axes. **b**, The critical fields for superconductivity at 45 mK (at which the resistance has fallen to half its normal-state value) measured at different γ . Superconductivity occurs below H_{sc1} and between H_{sc2} and H_{sc3} . H_R denotes the field at which the magnetic transition occurs, determined from the position of a sharp peak in the resistance at 500 mK. **c**, The same critical fields plotted in the H_a – H_b plane.

(1) Strong coupling gives only minor modifications in case studies where detailed calculations have been made²⁰. (2) Anisotropy of the superconducting order parameter can give changes of anisotropy with temperature of the order of 20% (refs 17,21). However, numerical calculations for different non-conventional superconductors¹⁸ suggest that equation (1) continues to provide a good phenomenological description of the angular dependence of the critical field at low temperature. (3) Special choices of Fermi-surface geometry can lead to major modifications of the form of the angular dependence at low temperature¹⁹. As the

Figure 3 The magnetic-field dependence of the geometric average of the coherence length, assuming constant anisotropy. The quantity plotted is proportional to the inverse-square coherence length, $\phi_0/2\pi\xi^2$, given in units of magnetic field where $\xi = (\xi_a\xi_b\xi_c)^{1/3}$ is the geometric mean of the coherence lengths along the different axes, calculated assuming a constant anisotropy. The vertical line marks the magnetic field at which the rotation transition occurs. The other straight line gives the value of $\phi_0/2\pi\xi^2$ below which the critical field for superconductivity for $H \parallel b$ is less than the applied field along the b axis; superconductivity cannot occur when $\phi_0/2\pi\xi^2$ falls below this line. The coherence length, ξ , changes from 143 Å for the data point closest to the origin to 44 Å for the data point with the highest value of $\phi_0/2\pi\xi^2$. The line through the data points is to guide the eye.

angular dependence we have measured is standard, the Fermi surface in URhGe cannot have such a special geometry in low magnetic field.

The present sample has a residual resistance ratio, $RRR = 50$. RRR is the ratio of the electrical resistance at room temperature divided by the normal-state resistance at low temperature and is proportional to the defect-limited electronic mean-free-path. The critical field, H_{sc1} , of the present sample is larger than that for a previously studied lower quality sample with $RRR = 20$ (ref. 17). The ratio $H_{sc1c}(RRR = 50)/H_{sc1c}(RRR = 20) \approx H_{sc1a}(RRR = 50)/H_{sc1a}(RRR = 20) = 1.34$ is approximately equal to the ratio of the square of the superconducting transition temperatures of the two samples in zero magnetic induction $(T_s(RRR = 50)/T_s(RRR = 20))^2 = (0.278 \text{ K}/0.237 \text{ K})^2 = 1.37$ (ref. 17). This result is the theoretical expectation for a non-conventional superconducting state. The ratio $H_{sc1b}(RRR = 50)/H_{sc1b}(RRR = 20) = 1.70$ is 30% larger than the above ratio; this is discussed further below in the light of our study of the high-field superconductivity.

The angular dependence of the critical fields, H_{sc2} and H_{sc3} , bounding the pocket of superconductivity at high magnetic field is quite different from the angular dependence of the critical field for the low-field superconducting pocket. The field window over which the high-field superconductivity occurs extends to either side of $H_R(\gamma)$ up to the highest applied field of 28 T (Fig. 2b). The main result of our study is that the component of the critical field parallel to the a axis plotted as a function of H_b (that serves to tune the material through the underlying magnetic transition) shows a strong upward curvature as H_b approaches H_R (Fig. 2c).

The measured angular dependence of H_{sc1} shows that the Fermi-surface geometry does not give rise to a stronger anisotropy

of the critical field than that which can be accounted for with an anisotropic effective mass. Assuming that the Fermi-surface geometry does not change radically with field, the strong field dependence of the critical fields H_{sc2} and H_{sc3} visible in Fig. 2 indicates that the magnitude of the coherence length changes as H_b is tuned through H_R . The geometric average of the coherence length, $\xi = (\xi_a\xi_b\xi_c)^{1/3}$, deduced from the measured H_{sc1} , H_{sc2} and H_{sc3} , assuming a fixed coherence length anisotropy, is shown in Fig. 3 (see the Methods section). Remarkably, the low-field and high-field superconducting regions seem to lie on the same smoothly varying curve. Furthermore, the stronger than T_s^2 increase of H_{sc1b} with sample quality can now be understood to result from an increase of $\phi_0/2\pi\xi^2$ as the low-field pocket of superconductivity extends to cover higher b -axis magnetic fields in better samples, reinforced by a reduction in the effectiveness of pair breaking by impurities owing to a shortening of the coherence length.

Theoretically, $1/\xi \approx k_B T_s / \hbar v_f$, where T_s is the hypothetical superconducting temperature in the absence of magnetic field and v_f is the Fermi velocity, inversely proportional to the effective electron quasi-particle mass for a fixed Fermi surface ($\hbar$ is the Planck constant divided by 2π and k_B is the Boltzmann constant). The increase of $1/\xi$ as the b -axis field approaches H_R requires that the effective mass and/or T_s increase strongly. For field applied along the b axis, the measured critical temperature for superconductivity has an upward curvature when plotted against field approaching H_R (ref. 4). Furthermore, the measured critical temperature reaches a higher value in the high-field superconducting pocket than found at zero field⁴. These measurements suggest that most of the increase of $1/\xi$ approaching H_R over the field range measured can be accounted for by an increase of T_s . Theoretically, T_s depends exponentially on different contributions to the effective mass¹², so this is not inconsistent with more modest changes of the effective mass. At a QCP driven by local physics, the effective quasi-particle mass diverges, as observed in YbRh₂Si₂ (refs 22,23). An increase of the effective mass is also predicted for a QCP in an itinerant ferromagnet²⁴ and observed approaching magnetic-field-induced transitions in several materials^{25,26}.

In other materials where QCPs have been studied experimentally under high magnetic field, including URu₂Si₂ (ref. 3) and Sr₃Ru₂O₇ (refs 7,27), new states that are not superconducting have been found. Our work on URhGe shows that superconductivity can, however, survive to much higher magnetic fields close to a QCP than previously thought likely⁸.

METHODS

The measurements were made with the M9 resistive magnet at the Grenoble High Magnetic Field Laboratory (GHMFL) in a top-loading dilution refrigerator equipped with a low-temperature rotator. The sample studied is the same as that used for resistivity measurements for fields in the bc plane⁴ and is a small bar aligned with its c axis parallel to the axis of rotation (within 1°). Current was passed along the length of the sample (b axis). The position of 90° rotation with the field parallel to the a axis was determined so that $H_R(\gamma)$ changed symmetrically for positive and negative rotations of γ away from this direction.

The fixed value for the coherence length anisotropy used to estimate the geometrically averaged coherence length from the measured values of the critical fields H_{sc1} , H_{sc2} and H_{sc3} was chosen to give the measured critical field anisotropy of H_{sc1} of the sample with $RRR = 20$ with a constant value of $\phi_0/2\pi\xi^2$. Symmetry imposes that the leading field dependence of $\phi_0/2\pi\xi^2$ is quadratic around zero field. The lower quality sample has a lower value of H_{scb1} than the higher quality sample and the change of $\phi_0/2\pi\xi^2$ from its zero-field value at H_{scb1} is small enough to be neglected.

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Competing financial interests

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