Superconductivity on the border of itinerant-electron ferromagnetism in UGe₂

S. S. Saxena*†‡, P. Agarwal*, K. Ahilan*, F. M. Grosche*‡, R. K. W. Haselwimmer*, M. J. Steiner*, E. Pugh*, I. R. Walker*, S. R. Julian*, P. Monthoux*, G. G. Lonzarich*, A. Huxley§, I. Sheikin§, D. Braithwaite§ & J. Flouquet§

* Department of Physics, Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, UK † Materials Science Centre, University of Groningen, Nigenborgh 4, 9747AG, The Netherlands § Département de Recherche Fondamentale sur la Matière condensée - SPSMS, CEA Grenoble, 17 Av. des Martyrs, Grenoble 38054, France

The absence of simple examples of superconductivity adjoining itinerant-electron ferromagnetism in the phase diagram has for many years cast doubt on the validity of conventional models of magnetically mediated superconductivity. On closer examination, however, very few systems have been studied in the extreme conditions of purity, proximity to the ferromagnetic state and very low temperatures required to test the theory definitively. Here we report the observation of superconductivity on the border of ferromagnetism in a pure system, UGe₂, which is known to be qualitatively similar to the classic d-electron ferromagnets. The superconductivity that we observe below 1 K, in a limited pressure range on the border of ferromagnetism, seems to arise from the same electrons that produce band magnetism. In this case, superconductivity is most naturally understood in terms of magnetic as opposed to lattice interactions, and by a spin-triplet rather than the spin-singlet pairing normally associated with nearly antiferromagnetic metals.

The origin of the remarkable stability or rigidity of structures in nature is a question of universal interest. The stability of simple systems of particles, such as atoms and molecules, is now described in great detail in terms of elementary quantum mechanics. But in the more complex systems that are of interest in condensed-matter physics and beyond, our understanding of rigidity is often incomplete or lacking entirely. A particularly dramatic example of largescale quantum rigidity is the phenomenon of superconductivity, which is thought to arise from the emergence of an attractive interaction that in some sense overwhelms the usual Coulomb repulsion between pairs of electrons. In the standard model due to Frohlich and to Bardeen, Cooper and Schrieffer (BCS), the crucially important attraction arises naturally from the indirect effects of the deformable underlying crystalline lattice of ions¹. This BCS picture, in which electrons form bound pairs as a result of lattice interactions, is now believed to account well for the great majority of known superconductors. But there is a growing number of metallic compounds, including the high-transition-temperature superconductors, in which superconductivity appears anomalous and where the precise mechanism of electron pairing remains controversial.

Soon after the advent of the BCS theory, alternative models for electron pairing were proposed that relied directly on subtle dynamical effects of the electrons themselves, in a static, nondeformable lattice. An effective attraction between pairs of electrons, or more precisely of fermion quasiparticles near the Fermi surface, arises—at first sight, paradoxically—from the cooperative effects of a collection of electrons that mutually repel each other as they move over the underlying static lattice of ions. In contrast to the bare Coulomb repulsion, which is independent of the electron spin, a part of the complex interaction between the quasiparticles can depend on the relative orientation of the spins and thus of the magnetic moments of the carriers^{2–17}. In the simplest case of nearly ferromagnetic metals—that is, metals on the verge of undergoing a transition to a ferromagnetic state at low temperatures—pairs of quasiparticles with parallel spins can attract while pairs with antiparallel spin tend to repel. When magnetic interactions in this example dominate over other types of quasiparticle interactions, parallel-spin quasiparticles tend to form pairs that must necessarily be in odd-parity orbitals. This can lead to spin-triplet, magnetically mediated, superconductivity^{2–8}. The effective magnetic interaction that we are describing is not the familiar dipole–dipole interaction in the theory of electromagnetism, which is relativistic and usually weak, but is a consequence of the Coulomb interaction itself, together with the subtle effects of quantum correlations; hence this effective magnetic interaction can be relatively strong, and potentially important for superconductivity in some cases.

Current theory suggests that this type of superconductivity is most likely to occur in metals that satisfy at least the following three conditions. First, they should be close to the border of ferromagnetism, either in a strongly paramagnetic or a weakly ferromagnetic state at low temperature where the longitudinal magnetic susceptibility that enters the magnetic interaction potential is strong and where magnetic interactions overwhelm other competing interactions. The balance between competing interactions, and also between the pair-forming and pair-breaking tendencies of magnetic interactions themselves, is a delicate one; superconductivity may not arise at all in some cases, or may exist in practice only over a very narrow range in a control parameter, such as hydrostatic pressure, used to tune the system towards the border of ferromagnetism. Second, the specimens selected must be of sufficiently high purity that the carrier mean free paths (due to either spin-dependent or spin-independent scattering mechanisms) exceed the typical dimensions of the spin-triplet pair states, that is, the characteristic superconducting coherence length. In contrast to conventional superconductors, these states are anisotropic in space and thus can be very sensitive to impurity scattering that tends to be essentially isotropic and strongly pair-breaking. Third, most candidate materials available, even at optimal lattice density and in their ideally pure states, may have to be cooled to the millikelvin temperature range to exhibit a spin-triplet form of magnetically

[‡]Present address: MPI Chemische Physik fester Stoffe, Bayreuther Str. 40, 01189 Dresden, Germany (E.M.G.); Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK (S.S.S.).

mediated superconductivity. Numerical analyses suggest that the superconducting transition temperature T_{SC} due to magnetic interactions is normally much lower for the spin-triplet pairing appropriate for our case of a nearly ferromagnetic metal, than for the spin-singlet even-parity pairing expected, for example, for the more complex case of a nearly antiferromagnetic metal at least in two dimensions¹⁸. (This assumes that the parameters entering the model are otherwise the same.) A key difference between these two cases is that an important factor defining the magnetic interaction potential—namely, the expectation of the inner product of the spins of the interacting quasiparticles—is three times stronger in magnitude for the spin-singlet state than for the spin-triplet state. This peculiar quantum property holds only for particles, such as the fermions of interest here, with spin quantum number of one-half in normal isotropic space.

Although there is growing evidence for the existence of magnetically mediated superconductivity in general, most of the information gathered thus far concerns the more complex examples of spin-singlet pairing normally associated with metals on the border of antiferromagnetism as opposed to ferromagnetism^{9–32}. Magnetic pairing of quasiparticles similar to the simplest kind we consider above is indeed thought to be relevant to superfluidity in liquid ³He (refs 3–7), but the corresponding phenomena in the nearly ferromagnetic metals has been more elusive. The lack of success in many searches for this simplest kind of magnetically mediated superconductivity, starting with early work on the nearly ferromagnetic metal Pd, has cast doubt on the validity of the theory of magnetically mediated superconductivity as it is presently formulated; even the possibility of a purely electronic mechanism for superconductivity in general has been doubted.

On closer examination, however, we find that very few itinerantelectron ferromagnets studied to date have been prepared in a sufficiently pure state, or have been 'tuned' (for example, by hydrostatic pressure) to be sufficiently close to the border of ferromagnetism, or cooled to sufficiently low temperatures, to provide a definitive check of the predictions of theory (for a recent review, see ref. 33). Recent analyses suggest that Pd, long considered the archetypal incipient ferromagnet, is in fact too far removed from the border of ferromagnetism to be a prime candidate for magnetically mediated superconductivity (P.M. and G.G.L., unpublished results). Low-temperature ferromagnets such as $ZrZn_2$ have not yet been prepared in sufficiently pure form to be expected to exhibit this kind of superconductivity, even near the critical



Figure 1 The magnetization and inverse magnetic susceptibility of UGe₂. Results (in SI units) are shown for a single crystal at ambient pressure along the easy axis in an applied field of 0.1 T. Left inset, the orthorhombic unit cell (the easy axis, or *a*-axis, is along the horizontal in the page); right inset, the typical form of the hysteresis loop at 4.5 K along the easy axis. The specimens used for magnetization and a.c. susceptibility measurements (Fig. 3) have demagnetizing factors along the easy axis of less than 0.2. *M*, magnetization; *H*, magnetic field; *B*₀, applied magnetic induction.

pressure where the Curie temperature $T_{\rm C}$ vanishes³⁴. In one promising system, MnSi, in which the three conditions mentioned above have apparently been met, the anticipated odd-parity spin-triplet superconductivity is not in fact observed^{35,36}. However, this material may fail to meet another and more subtle criterion needed for spintriplet pairing. In particular, its B20 crystal structure lacks the inversion symmetry required to guarantee that equal-spin states of opposite momentum are degenerate and hence effectively coupled by magnetic interactions. In the nearly magnetic metal Sr₂RuO₄, evidence for spin-triplet superconductivity is mounting, but its connection with ferromagnetic spin susceptibility and the even stronger antiferromagnetic spin susceptibility inferred from neutron-scattering experiments has not yet been clarified³⁷⁻³⁹. In RuSr₂GdCu₂O₈ superconductivity and magnetism with a ferromagnetic component appear to coexist at least in different layers of the layered perovskite structure^{40,41}. However, superconductivity is thought to arise from spin-singlet *d*-wave pairing in the nearly antiferromagnetic CuO₂ layers, and not from spin-triplet pairing associated with the ferromagnetism of the RuO₂ layers. Finally, in the system of nominal composition Y₉Co₇ very weak ferromagnetism and some form of superconductivity may coexist^{42,43}, but complex metallurgical properties, strong impurity scattering and ambiguities concerning the character of the magnetic electrons and the superconducting carriers have hampered progress in understanding. We note that measurements of the heat capacity, residual resistivity and upper critical field suggest that the carrier mean free path in Y₉Co₇ is considerably smaller than the superconducting coherence length. This would seem to preclude in this case the possibility of spin-triplet magnetically mediated superconductivity.

We conclude that an unequivocal example of magnetically mediated superconductivity connected with the simplest case of ferromagnetism, as opposed to the more complex case of antiferromagnetism, is still lacking. Here we report the results of a search for this kind of superconductivity in the low-temperature metallicferromagnet UGe₂, which appears to satisfy the various requirements mentioned above and which may not have the disadvantages of the systems described thus far. Our study may also test the alternative models of ferromagnetism and superconductivity of refs 44 and 45.

Band magnetism in UGe₂

UGe₂ crystallizes congruently from the melt into an orthorhombic structure with full inversion symmetry^{46,47}. Single crystals grow readily in the stoichiometric state, and specimens with unusually high purity can be prepared. In the best cases residual resistivities as



Figure 2 The temperature–pressure phase diagram of UGe₂. T_c denotes the Curie temperature and T_{SC} the superconducting transition temperature; the latter is determined from the 50% drop in resistivity, and the former from the cusp in the resistivity or the peak in the a.c. susceptibility (see for example, refs 50 and 53). (We note that the T_{SC} values are scaled by a factor of 10. For T_c versus pressure, see also ref. 49. The dashed and solid lines serve only to connect the data.)

low as 0.2 μΩ cm have been reported, corresponding in a conventional analysis to carrier mean free paths of several thousand ångstroms (ref. 47). Ferromagnetic order is observed below a $T_{\rm C}$ that starts at 53 K at ambient pressure and falls monotonically towards absolute zero at a critical pressure p_c of 1.6 to 1.7 GPa, that is, at a pressure readily accessible with conventional piston and cylinder cells⁴⁸⁻⁵⁰. The magnetic transition near p_c appears to be of first order⁵⁰, as was also noted earlier in the case of MnSi (ref. 35). This implies that the longitudinal susceptibility that enters the magnetic interaction potential cannot be tuned with pressure to arbitrarily high values at low temperatures. Thus, the conditions for magnetically mediated superconductivity need not be fulfilled at any pressure. As we shall see, however, superconductivity does indeed occur at least in the millikelvin range in a narrow pressure window in the ferromagnetic state just below p_c .

In the uranium compounds known as 'heavy-fermion systems' the 5f levels are highly localized and, in conjunction with other more delocalized levels with which they couple, give rise to gapless fermion excitations characterized by effective masses approaching (in extreme cases) that of protons or neutrons. In UGe₂, however, the 5f electrons are more itinerant than in many heavy-fermion systems, and Fermi-surface studies together with heat capacity measurements suggest that they behave more like the 3d electrons in the traditional itinerant-electron ferromagnets such as Fe, Co and Ni (refs 47, 51, 52). We stress that the magnetization in the latter ferromagnets arises from an exchange spin splitting of conduction electron bands, and not from a polarization of strictly localized electrons as is the case, for example, in the ferromagnetic metal Gd. UGe₂ differs from the 3d metals mainly in having a stronger spinorbit interaction that leads to an unusually large magnetocrystalline anisotropy. The magnetization in UGe₂ is held along the easy magnetic axis by an anisotropy field of the order 100 T which is two or more orders of magnitude greater than in the 3d metals. As discussed in ref. 18, this anisotropy may be particularly favourable for magnetic pairing in the triplet channel. The important point is that, in the best available specimens, the f electrons give rise to fully itinerant quasiparticles with sufficiently long mean free paths to allow, at least at very low temperatures and near p_c , for the possibility of spin-triplet magnetically mediated superconductivity of the type considered here.



Figure 3 The a.c. susceptibility χ of UGe₂ at high pressure. The main figure shows the typical form of χ along the easy axis in the Earth's magnetic field and at approximately 1.3 GPa. The rapid drop of the susceptibility upon entering the ferromagnetic state is consistent with the behaviour of the initial susceptibility observed in hysteresis measurements at ambient pressure (see Fig. 1 right inset). This behaviour is most naturally understood in terms of the abnormally strong magnetic anisotropy that characterizes this material. The interpretation of the observed drop of the susceptibility in the superconducting state (inset; at approximately 1.5 GPa) must take account of the presence of the internal field due to the ferromagnetic magnetization that is expected to lead to a spontaneous vortex lattice even in the absence of an external applied magnetic field^{43,62}. The existence of an internal field of the order of 0.1 to 0.2 T is consistent with our measurements of the upper critical field versus temperature.

Measurements of UGe₂

The magnetization and susceptibility versus temperature of UGe₂ at ambient pressure and in an applied field of 0.1 T are shown in Fig. 1, for the easy axis of magnetization. Also shown, as insets to this figure, are the unit cell and a typical hysteresis loop with the applied field along the easy axis. The form of the hysteresis curve and the difference in the magnetization along the easy and hard axes are consistent with an extremely strong magnetocrystalline anisotropy as discussed earlier. We note that one effect of this anisotropy is to greatly suppress at the centre of the hysteresis loop the initial susceptibility, which is governed by the way in which magnetic domains can grow or rotate. We also note that the data of Fig. 1 for the easy axis yield a moment per U atom of $\sim 1.4 \,\mu_B$ within a single domain at low temperatures and low fields, but an effective paramagnetic moment above $T_{\rm C}$ of ~2.7 $\mu_{\rm B}$. This difference in values, and in particular a ratio of the paramagnetic to the ordered moment that is greater than unity, is widely observed in the 3d magnetic metals and is consistent with the picture of band or itinerant electron magnetism that is supported by Fermi-surface studies in UGe₂.

The central result of our studies is the temperature–pressure phase diagram presented in Fig. 2. It shows that $T_{\rm C}$ falls monotonically with increasing pressure, drops precipitously above 1.5 GPa, and appears to vanish at a critical pressure $p_{\rm c}$ of between 1.6 to 1.7 GPa (ref. 50). Values of $T_{\rm C}$ determined from the resistivity, a.c and d.c. magnetization measurements and elastic neutron scattering, are found to be consistent within experimental error. As in a previous study in MnSi, but now to a more extreme degree, the magnetic transition versus temperature becomes first order just below $p_{\rm c}$ but the critical temperature versus pressure nevertheless appears to remain continuous⁵⁰.

In a narrow range in pressure below p_c and thus within the ferromagnetic state, we observe a sudden and complete loss of resistivity in the millikelvin temperature range below $T_{\rm SC}$ (Fig. 2). The abrupt loss of resistivity just below p_c appears to be a robust property of UGe₂, and has been observed in all of the eight samples that we have investigated. The survival of bulk ferromagnetism below $T_{\rm SC}$ has been confirmed directly via elastic neutron scattering measurements (A. Huxley *et al.*, manuscript in preparation). Also, we note that the a.c. susceptibility below $T_{\rm SC}$ tends to the limit of -1 (in SI units), as expected in the presence of bulk superconductivity (Fig. 3). The upper critical field B_{c2} is in excess of 3 T near the maximum of $T_{\rm SC}$ and is much higher than normally expected for





conventional superconducting inclusions⁵³. Moreover, the apparent initial gradient $\partial H_{c2}/\partial T$ near T_{SC} is anomalously large, and consistent with that expected in the presence of an internal field arising from ferromagnetic order.

As shown in Fig. 4 the resistivity $\rho(T)$ above T_{SC} , but typically below 10 K, is roughly described by an expression of the form $\rho(T) = \rho_0 + AT^2$, where ρ_0 is the residual resistivity and A is the quadratic coefficient. This is the form expected from the mutual scattering of fermion quasiparticles within the standard theory of metals; that is, within Fermi-liquid theory when the direct contribution to $\rho(T)$ from scattering of quasiparticles by well-defined collective modes such as phonons and magnons can be neglected. The parameters ρ_0 and A are not constants but vary with crystallographic direction and, more importantly, with pressure⁴⁸. As shown in the lower inset of Fig. 4, the quadratic coefficient A has a very pronounced pressure dependence and rises rapidly at a pressure $p_x \approx 1.2$ GPa close to (but distinctly below) p_c and within the narrow pressure window where superconductivity is indeed observed. We note that as p_x is approached, the range of validity of the above Fermi-liquid form of the resistivity collapses⁴⁸. The temperature variation of $\rho(T)$ is then more usefully described by a temperature exponent well below the Fermi liquid value of 2 (at low T but above T_{SC}).

Naively, we expect the quasiparticle interactions and thus A to be strongest at the critical boundary separating ferromagnetism and paramagnetism; namely, at p_c rather than at the lower pressure p_x , as observed. There is, however, evidence for the existence of a cross-over anomaly within the ferromagnetic state whose characteristic temperature T_x collapses towards zero also near p_x . This anomaly shows up typically as a weak and broad maximum in the temperature derivatives of the resistivity and the magnetization versus temperature⁴⁸. It also appears in the temperature dependence of the thermal expansion coefficient⁴⁹. We discuss below the possible origin of this cross-over phenomenon or transition, and its potential relevance to the pressure dependence of A and to superconductivity.

Role of magnetic interactions

We begin by comparing our findings with the predictions of the simplest model of magnetic interactions for an itinerant-electron ferromagnet. First, we consider more fully the facts that support the itinerant-electron picture, which is crucial to our discussion. Band structure analyses show that magneto-oscillatory phenomena observed in UGe₂ can be understood in terms of conventional energy bands calculated in the hypothetical paramagnetic state but with the Fermi level for majority and minority spins separated by an exchange splitting of the order of 70 meV (refs 47, 51). (This conclusion was based on an early determination of the crystal structure but is also expected to hold for the slightly revised crystal structure given in refs 54, 55.) The required exchange splitting is not large enough to entirely fill the majority spin bands; thus the Fermi surface consists of both majority and minority spin sheets. The masses of the quasiparticle excitations on these sheets of the Fermi surface are of the order of 20 times the bare electron mass or lower, and are consistent with the observed linear coefficient of the heat capacity of 35 mJ per mol U per K² (ref. 47). These findings, and the high ratio of the effective paramagnetic moment to the ordered moment discussed above, are generally consistent with the behaviour that has long been associated with the traditional itinerantelectron ferromagnets.

Within the simplest itinerant-electron model, the longitudinal magnetic susceptibility that enters the magnetic interaction potential for spin-triplet pairing is expected to have a maximum in the low-temperature limit around p_c . This should lead to a maximum in the resistivity coefficient *A* at p_c , as already noted, and spin-triplet magnetically mediated superconductivity over a narrow pressure range both above and below p_c . If the magnetic transition is strictly continuous or second order, a narrow dip in T_{SC} may arise very close

to p_c where pair formation tends to be frustrated by strong quasiparticle damping^{7,18}. This dip is expected to be suppressed, however, when the transition is first order. Moreover, when the transition is strongly first order, as is the case in UGe₂, the pressure range in which $T_{\rm SC}$ is finite is expected to contract and even collapse entirely on one or both sides of p_c .

The observation of superconductivity on just one side of p_c is not necessarily inconsistent with this model, but the shift in the peak in A versus pressure from p_c to the lower pressure p_x associated with the cross-over anomaly, would seem to require a more elaborate picture. A possible explanation for these features that is still broadly consistent with a spin-triplet magnetic interaction model is offered by a rather special property of the majority electron sheet of UGe₂. In the hypothetical paramagnetic state, the Fermi surface consists of an electron sheet having a quasi-cylindrical shape and a more complex hole sheet enclosing an equal volume to that of the electron sheet. In the presence of the exchange splitting, the minority-spin electron surface contracts greatly while the majority-spin counterpart expands towards the boundaries of the Brillouin zone and is cut off by the zone walls on two sides. What remains of the majority electron surface are two large and roughly parallel sheets reminiscent of a quasi-one-dimensional (quasi-1D) system⁵¹. Because the other sheets of the Fermi surface remain essentially three-dimensional, the resistivity and magnetic susceptibility are not expected to show quasi-1D behaviour. However, the 'hidden' quasi-1D character associated with the large and potentially important electron sheet of the majority Fermi surface can have profound consequences. In particular, the strong nesting of this sheet is expected to lead to very strong magnetic interactions between carriers of the same spin, peaked periodically in space at distances defined by the inverse of the nesting wave vector.

This quasi-1D model would tend to drive a transition from a simple ferromagnetic state to a modulated ferromagnetic state once a well defined exchange splitting, and hence quasi-1D character of the majority electron surface, has developed. This may provide a natural explanation of the cross-over anomaly T_x observed well below $T_{\rm C}$ and $p_{\rm c}$. The peak in A at $p_{\rm x}$ instead of $p_{\rm c}$ would then be due to the fact that the ferromagnetic transition is strongly first order and magnetic interactions associated with it are relatively weak, while the lower transition (or a tendency) to a modulated structure is more nearly second order and hence produces strong magnetic interactions and pronounced quasiparticle scattering at low temperatures. As in the simplest model, this more elaborate scheme is also expected to lead naturally to spin-triplet magnetically mediated superconductivity. This is because the interactions are strong only between quasiparticles on the quasi-1D sheet of the Fermi surface which are all of the same spin, and also because the form of the magnetic interaction potential itself favours p-wave pairing. We stress that our model is not the same in detail as that applied to the quasi-1D organic compounds⁵⁶ in which the transport properties are highly anisotropic and metallic magnetism tends to be suppressed, nor is it the same as that traditionally applied to ³He (refs 2-6)—in which the Fermi surface is a simple sphere, and magnetic interactions are non-oscillatory in space.

For completeness we also consider briefly the viability of alternative available models for the superconductivity that we observe. Recently, the coexistence of ferromagnetism and spin-singlet *s*-wave superconductivity has been reconsidered^{44,45}. Singlet pairing cannot be ruled out when the exchange splitting of the Fermi surface is extremely small; for example, at the very border of a magnetic transition of second order at low *T*. In UGe₂, however, the ferromagnetic transition is strongly first order and the exchange splitting is large. Also, the proposed *s*-wave model does not seem to provide a natural interpretation of the cross-over anomaly, the pressure dependence of *A*, nor for the absence of superconductivity under similar conditions in the ferromagnetic states of several closely related metals. (We note that the deleterious effects of impurities or breakdown of inversion symmetry that were discussed for odd parity or *p*-wave pairing do not extend in the same degree to *s*-wave pairing.)

The mechanisms that we have considered thus far are electronic in nature, and involve the lattice vibrations only in an indirect way-for example, in possibly modifying the phenomenological parameters that enter the quasiparticle interaction potential. A more direct role of the dynamics of the lattice for pairing in an itinerant-electron ferromagnet cannot be ruled out, but it would probably be considerably more exotic than in the traditional BCS model. In the band model appropriate to UGe₂, pairing in the spinsinglet channel is unlikely because states of opposite momentum and of opposite spin on the exchange-split Fermi surface are only rarely (if ever) degenerate. At least in the presence of inversion symmetry, on the other hand, states of opposite momentum and of the same spin are always degenerate, and can readily be paired en masse in a zero-total-momentum state. Thus, it would seem that mechanisms for superconductivity based in some way more directly on lattice dynamics must also, as with the case of magnetic interactions, be compatible with spin-triplet odd-parity pairing.

As previously noted, the kind of superconductivity that we consider here can only arise if the carrier mean free path *l* exceeds the characteristic superconducting coherence length, ξ . From the measured upper critical field, we estimate in the standard way that ξ is of the order of 200 Å for pressures near p_x . This is indeed substantially lower than *l*, which is estimated from values of ρ_0 of our samples to be of the order of or greater than 1,000 Å (ref. 57). It is also of interest to examine whether the above order of magnitude of ξ is indeed reasonable within the itinerant-electron model we are considering. In the standard description, ξ depends on T_{SC} , the typical quasiparticle mass and the typical Fermi wavevector. The latter is not sensitive to pressure and can be estimated from zero pressure properties, but the quasiparticle mass is expected to vary strongly with pressure and is more difficult to determine. Within the magnetic interactions model the principal contribution to this mass is due to self interactions, which are naturally also connected to the quasiparticle scattering rate and hence to A. This leads to a simple connection between A and the linear coefficient of the heat capacity γ , which is proportional to the average of the quasiparticle mass. The connection can be expressed roughly as $A/\gamma^2 \approx 10 \,\mu\Omega \,\mathrm{cm} \,\mathrm{K}^{-2}$ per $(J \mod^{-1} K^{-2})^2$, provided neither γ nor A are too close to singularities⁵⁸. This formula is found to be obeyed in order of magnitude by many metals on the border of magnetic order⁵⁹. It is also obeyed approximately by UGe2 at ambient pressure, and from the known pressure variation of A, it suggests that the typical quasiparticle mass near p_x may be nearly two orders of magnitude greater than the bare electron mass. This leads us to a value of ξ in the itinerant model of a few hundred ångstroms and thus of the same order of magnitude as that determined from critical field measurements. We note that at least near p_{xy} the estimated characteristic quasiparticle mass in UGe₂ is approaching in magnitude that of heavy-fermion metals such as UPt₃ (refs 21, 60). In contrast to UGe₂, however, UPt₃ and the other known heavy-fermion superconductors are either paramagnetic or antiferromagnetic in their normal non-superconducting states. For this and the other reasons given above, UGe2 appears to be unique among the known superconductors.

Spin-triplet superconductivity

We have observed superconductivity in the ferromagnetic state of the itinerant-electron system UGe₂. Superconductivity exists only within a narrow window in lattice density, close to a critical density where ferromagnetism disappears. Our findings have been discussed in relation to a quasiparticle interaction model that favours a spintriplet magnetically mediated form of superconductivity. This differs from the simplest model of magnetic pairing in a ferromagnet in that it includes the effects of a quasi-1D majority-spin sheet of the Fermi surface embedded in an otherwise threedimensional Fermi surface that characterizes this material. The effects of nesting of this majority-spin sheet can account qualitatively for the observed cross-over anomaly, and leads naturally to magnetic pairing in the spin-triplet channel in a narrow range of density within the ferromagnetic side of the critical density. Because the ferromagnetic state out of which superconductivity arises is well described within a band model, even more exotic models than the one we propose here would seem to require spin-triplet pairing. UGe₂ provides us with a new, and apparently unique, example of superconductivity in the field of magnetism.

Note added in proof: Additional theoretical analyses relevant to this work may be found in refs 63 and 64. For a more complete discussion of the background of this research and of our initial search for magnetically mediated superconductivity in UGe₂ see ref. 65.

Methods

Samples of UGe₂ were grown by radio-frequency induction melting in water-cooled copper crucibles in ultrahigh-vacuum chambers. Details of the preparation techniques will be given elsewhere. The crystals selected for investigation have residual resistivities as low as a few tenths of 1 μ Ω cm. Also, no secondary chemical phases have been detected by electron-probe microanalysis with a resolution of 100 nm and accuracy of 1% of composition.

Measurements of the electrical resistivity and a.c. magnetic susceptibility in both aligned crystals and polycrystalline samples have been performed as a function of temperature, magnetic field and pressure. The pressure cells are of the piston-cylinder type made of non-magnetic alloys suitable for efficient cooling to the low millikelvin range and for the application of external magnetic fields⁶¹. The pressure-transmitting fluids employed within these cells are those found to produce hydrostatic pressures of adequate homogeneity for sensitive quantum oscillation studies, for example. The low millikelvin range was reached by means of a dilution refrigerator (Oxford Instruments) or a demagnetization refrigerator (Cambridge Magnetic Refrigeration) with base temperatures of 20 mK and 100 mK, respectively, in the presence of a standard pressure cell. Studies as function of magnetic field were made with a 6-T superconducting magnet attached to the dilution refrigerator system.

The resistivity was measured by the conventional four-terminal method using common mode voltage rejection circuitry and low-temperature transformers to allow adequate voltage sensitivity with the very low excitation powers needed for acceptable levels of sample heating. The a.c. magnetic susceptibility was studied with similar electronics and in zero d.c. external magnetic field by means of a miniature niobium modulation coil wound on top of a compensated copper pick-up coil mounted as an assembly inside the pressure cells. The sample temperature was measured in the millikelvin range by means of calibrated RuO₂ or carbon resistance thermometers thermally anchored to the outside of the Au-plated pressure cells and also anchored to the samples directly via copper leads passing into the pressure cells. To ensure that the samples and thermometers were at the same temperature, very slow rates of sweep were employed (typically 1 mK min⁻¹ below 1 K), and it was checked that up and down temperature ramps gave equivalent results independent of the excitation power in the ranges of interest.

Finally, bulk magnetization measurements as function of magnetic field and temperature were performed by means of a SQUID magnetometer (Quantum Design) with a base temperature of 2 K. A miniature pressure cell only 8 mm in diameter was constructed out of high-purity non-magnetic BeCu to allow high-precision measurements to be extended from atmospheric pressure to applied pressures of up to approximately 1.5 GPa. Elastic neutron scattering measurements have also been carried out in this temperature and pressure range. Details of these measurements will be given elsewhere.

Received 19 May; accepted 27 June 2000.

- Bardeen, J., Cooper, L. N. & Schrieffer, J. R. Theory of superconductivity. *Phys. Rev.* 108, 1175–1204 (1957).
- Brueckner, K. A., Soda, T., Anderson, P. W. & Morel, P. Level structure of nuclear matter and liquid ³He. *Phys. Rev.* 118, 1442–1446 (1960).
- Nakajima, S. Paramagnon effect on the BCS transition in ³He. Prog. Theor. Phys. 50, 1101–1109 (1973).
- Brinkman, W. F., Serene, J. W. & Anderson, P. W. Spin-fluctuation stabilization of anisotropic superfluid states. *Phys. Rev. A* 10, 2386–2394 (1974).
- Leggett, A. J. A theoretical description of the new phases of liquid ³He. *Rev. Mod. Phys.* 47, 331–414 (1975).
- Levin, R. & Valls, O. T. Strong-coupling theory of superfluid transition temperatures for paramagnon models: application to ³He. *Phys. Rev. B* 17, 191–200 (1978).
- Fey, D. & Appel, J. Coexistence of p-state superconductivity and itinerant ferromagnetism. *Phys. Rev.* B 22, 3173–3182 (1980).
- Hirsch, J. E. Attractive interaction and pairing in fermion systems with strong on-site repulsion. *Phys. Rev. Lett.* 54, 1317–1320 (1985).
- Miyake, K., Schmitt-Rink, S. & Varma, C. M. Spin-fluctuation mediated even-parity pairing in heavyfermion superconductors. *Phys. Rev. B* 34, 6554–6556 (1986).
- Scalapino, D. J., Loh, E. Jr, Hirsch, J. E. d-wave pairing near a spin-density-wave instability. *Phys. Rev.* B 34, 8190–8192 (1986).
- Millis, A. J., Sachdev, S. & Varma, C. M. Inelastic-scattering and pair breaking in anisotropic and isotropic superconductors. *Phys. Rev. B* 37, 4975–4986 (1988).

- Bickers, N. E., Scalapino, D. J. & White, S. R. Conserving approximations for strongly correlated electron-systems - Bethe-Salpeter equation and dynamics for the two-dimensional Hubbard-model. *Phys. Rev. Lett.* 62, 961–964 (1989).
- Pines, D. Spin excitations and superconductivity in cuprate oxide and heavy electron superconductors. *Physica B* 163, 78–88 (1990).
- Moriya, T., Takahashi, Y. & Ueda, K. Antiferromagnetic spin fluctuations and superconductivity in 2dimensional metals - a possible model for high-T_c oxides. J. Phys. Soc. Jpn 52, 2905–2915 (1990).
- Monthoux, P., Balatsky, A. V. & Pines, D. Towards a theory of high temperature superconductivity in the antiferromagnetically correlated cuprate oxide. *Phys. Rev. Lett.* 67, 3448–3451 (1991).
- Bulut, N., Hone, D. W., Scalapino, D. J. & Bickers, N. E. Knight-shifts and nuclear-spin-relaxation rates for 2-dimensional models of CuO₂. Phys. Rev. B 41, 1797–1811 (1990).
- Schrieffer, J. R., Wen, X. G. & Zhang, S. C. Spin-bag mechanism of high-temperature superconductivity. *Phys. Rev. Lett.* 60, 944–947 (1988).
- Monthoux, P. & Lonzarich, G. G. ρ-wave and d-wave superconductivity in quasi-two-dimensional metals. *Phys. Rev. B* 59, 14598–14605 (1999).
- Steglich, F. et al. Superconductivity in the presence of strong Pauli paramagnetism: CeCu₂Si₂. Phys. Rev. Lett. 43, 1892–1896 (1979).
- Ott, H. R., Rudigier, H., Fisk, Z. & Smith, J. L. UBe₁₃—an unconventional actinide superconductor. Phys. Rev. Lett. 50, 1595–1598 (1983).
- 21. Stewart, G. R. Heavy-fermion systems. Rev. Mod. Phys. 56, 755-787 (1984).
- 22. Fisk, Z. et al. Heavy-electron metals: new highly correlated states of matter. Science 239, 33-42 (1988).
- 23. Jerome, D. Organic Conductors (Dekker, New York, 1994).
- 24. Ishiguro, T. & Yamaji, K. (eds) Organic Conductors Ch. 3 (Springer, Berlin, 1990).
- Jaccard, D., Behnia, K. & Sierro, J. Pressure-induced heavy fermion superconductivity of CeCu₂Ge₂. *Phys. Lett. A* 163, 475–480 (1992).
 Grosche, F. M., Julian, S. R., Mathur, N. D. & Lonzarich, G. G. Magnetic and superconducting phases
- 20. Grosting F. M., Junan, G. K., Walmar, N. D. & Lonzarten, G. G. Magnetic and superconducting phase in CePd₂Si₂. *Physica B* 224, 50–52 (1996).
- Movshovich, R. et al. Superconductivity in heavy-fermion CeRh₂Si₂. Phys. Rev. B 53, 8241–8244 (1996).
- Julian, S. R. et al. The normal states of magnetic d and f transition metals. J. Phys. Condens. Matter 8, 9675–9688 (1996).
- Walker, I. R., Grosche, F. M., Freye, D. M. & Lonzarich, G. G. The normal and superconducting states of Celn₃ near the border of antiferromagnetic order. *Physica C* 282–287, 303–306 (1997).
- Fukuyama, H. Electronic phase transition tuned by pressure: superconductivity and antiferromagnetism. Rev. High Pressure Sci. Technol. 7, 465–468 (1998).
- Mathur, N. D. *et al.* Magnetically mediated superconductivity in heavy fermion compounds. *Nature* 394, 39–43 (1998).
- Jourdan, M., Huth, M. & Adrian, H. Superconductivity mediated by spin fluctuations in the heavyfermion compound UPd₂Al₃. *Nature* 398, 47–49 (1999).
- Lonzarich, G. G. in *Electron* (ed. Springford, M.) Ch. 6 (Cambridge Univ. Press, Cambridge, 1997).
 Grosche, F. M., Pfleiderer, C., McMullan, G. J., Lonzarich, G. G. & Bernhoeft, N. R. Critical behaviour
- of ZrZn₂. *Physica B* **206 & 207**, 20–22 (1995). 35. Pfleiderer, C., McMullan, G. J., Julian, S. R. & Lonzarich, G. G. Magnetic quantum phase transition in
- MnSi under hydrostatic pressure. *Phys. Rev. B* 55, 8330–8338 (1997).
 36. Thessieu, C. Pfleiderer, C. & Flouquet, J. Thermodynamical study under hydrostatic pressure of MnSi.
- Inesseu, C. Fneiderer, C. & Fiouquet, J. Thermodynamical study under hydrostatic pressure of MnSi Physica B 239, 67–70 (1997).
- Maeno, Y. *et al.* Superconductivity in a layered perovskite without copper. *Nature* 372, 532–534 (1994).
- 38. Rice, T. M. An analogue of superfluid ³He. Nature 396, 627-629 (1998).
- Sidis, Y. et al. Evidence for incommensurate spin fluctuations in Sr₂RuO₄. Phys. Rev. Lett. 83, 3320– 3323 (1999).
- Tallon, J. et al. Coexisting ferromagnetism and superconductivity in hybrid rutheno-cuprate superconductors. IEEE Trans. Appl. Supercond. 9, 1696–1699 (1999).
- Pickett, W. E., Weht, R. & Shick, A. B. Superconductivity in ferromagnetic RuSr₂GdCu₂O₈. *Phys. Rev. Lett.* 83, 3713–3716 (1999).
- Kolodziejczyk, A., Sarkissian, B. V. B. & Coles, B. R. Magnetism and superconductivity in a transition metal compound: Y₄Co₃. *J. Phys. F* 10, L333–L337 (1980).
- Sinham K. P. & Kakani, S. L. Magnetic Superconductors: Recent Developments (Nova Science, New York, 1989).

- Blagoev, K. B., Engelbrecht, J. R. & Bedell, K. S. Effect of ferromagnetic spin correlations on superconductivity in ferromagnetic metals. *Phys. Rev. Lett.* 82, 133–136 (1999).
- Krachev, N. I., Blagoev, K. B., Bedell, K. S. & Littlewood, P. B. Coexistence of superconductivity and ferromagnetism in ferromagnetic metals. Preprint cond-mat/9911489 at (http://xxx.lanl.gov) (1999; cited 30 Nov. 1999).
- Menovsky, A., de Boer, F. R., Frings, P. H. & Franse, J. J. M. in *High Field Magnetism* 189 (ed. Date, M.) (North-Holland, Amsterdam, 1983).
- 47. Satoh, K. et al. de Haas-van Alphen effect in UGe2. J. Phys. Soc. Jpn. 61, 1827-1828 (1992).
- Oomi, G., Kagayama, T. & Onuki, Y. Critical electron scattering in UGe₂ near the magnetic phase transition induced by pressure. J. Alloys Compounds 271–273, 482–485 (1998).
- Nishimura, K., Oomi, G., Yun, S. W. & Onuki, Y. Effect of pressure on the Curie temperature of singlecrystal UGe₂. J. Alloys Compounds 213, 383–386 (1994).
- Huxley, A., Sheikin, I. & Braithwaite, D. Metamagnetic behaviour near the quantum critical point in UGe₂. *Physica B* 284 & 288, 1277–1278 (2000).
- Yamagami, H. & Hasegawa, A. Fermi surface of the ferromagnetic heavy-electron compound UGe₂. *Physica B* 186–188, 182–184 (1993).
- Lonzarich, G. G. Band structure and magnetic fluctuations in ferromagnetic or nearly ferromagnetic metals. J. Magn. Magn. Mater. 45, 43–53 (1984).
- 53. Agarwal, P. Magnetism and superconductivity in heavy fermion metals. Thesis, Univ. Cambridge (2000).
- 54. Oikawa, K., Kamiyama, T., Asano, H., Onuki, Y. & Kohgi, M., Crystal structure of UGe₂. *J. Phys. Soc. Jpn* **65**, 3229–3232 (1996).
- Boulet, P. et al. Crystal and magnetic structure of the uranium digermanide UGe₂. J. Alloys Compounds 247, 104–108 (1997).
- Lee, I. J., Naughton, M. J., Danner, G. M. & Chaikin, P. M. Anisotropy of the upper critical field in TMTSF₂ PF₆. *Phys. Rev. Lett.* 78, 3555–3558 (1997).
- Mackenzie, A. P. et al. Extremely strong dependence of superconductivity on disorder in Sr₂RuO₄. Phys. Rev. Lett. 80, 161–164 (1998).
- Takimoto, T. & Moriya, T. Relationship between resistivity and specific heat in heavy electron systems. Solid State Commun. 99, 457–460 (1996).
- Kadowaki, K. & Woods, S. B. Universal relationship of the resistivity and specific heat in heavyfermion compounds. *Solid State Commun.* 58, 507–509 (1986).
- Taillefer, L. & Lonzarich, G. G. Heavy-fermion quasiparticles in UPt₃. Phys. Rev. Lett. 60, 1570–1573 (1988).
- Walker, I. R. Nonmagnetic piston-cylinder pressure cell for use at 35 kbar and above. *Rev. Sci. Instrum.* 70, 3402–3412 (1999).
- Bulaevskii, L. N., Buzdin, A. I., Panjukov, S. V. & Kulic, M. L. Coexistence of superconductivity and magnetism: theoretical predictions and experimental results. *Adv. Phys.* 39, 175 (1985).
- Roussev, R. & Millis, A. J. Quantum critical effects on transition temperature of magnetically mediated p-wave superconducitivity. Preprint cond-mat/0006208 at (http::xxx.lanl.gov) (2000; cited 26 May 2000).
- Ohmi, T. & Machida, K. Nonunitary superconducting state in UPt³. Phys. Rev. Lett. 71, 625–628 (1993).
- 65. Saxena, S. S. Magnetic and superconducting phases of heavy fermion compounds. Thesis, Univ. Cambridge (1998).

Acknowledgements

We thank in particular S. V. Brown and also F. Beckers, K. S. Bedell, K. B. Blageov, D. M. Broun, P. Coleman, D. Forsythe, C. D. Frost, D. E. Khmelnitskii, P. B. Littlewood, A. J. Millis, P. Niklowitz, T. T. M. Palstra, D. Pines, C. Pfleiderer, K. Sandeman, A. J. Schofield and A. Tsvelik for discussions. The work was supported in part by the Cambridge Research Centre in Superconductivity, the UK EPSRC, the Paul Instrument Fund of the Royal Society, the Cambridge Newton Trust and the Commonwealth Scholarship Commission. The work performed in Grenoble was supported by the CEA Direction des Sciences de la Matière.

Correspondence and requests for materials should be addressed to G.G.L. (e-mail: ltp-secretary@phy.cam.ac.uk).