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Multiple superconducting phases in a nearly ferromagnetic system

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The nearly ferromagnetic superconductor UTe_2 shows several intriguing phenomena, including an extraordinary reinforcement of superconductivity in very strong magnetic fields. Here we show that pressure tunes the system to a more correlated state and probable magnetic order. The superconducting critical temperature is also strongly enhanced, reaching almost 3 K, a new record for U-based heavy fermion superconductors. Most spectacularly under pressure we find a transition within the superconducting state, putting UTe_2 among the very rare systems having multiple superconducting phases. UTe_2 under pressure is a treasure trove of several of the most fascinating phenomena in unconventional superconductivity and may well be a keystone in their understanding.

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In most superconductors, the superconducting order parameter is *s*-wave, meaning it has the same symmetry as the crystal lattice. However, in the ever-expanding family of unconventional superconductors, which includes such disparate members as high- T_C cuprates and pnictides, organic superconductors, and heavy fermions, the order parameter can assume a number of different symmetries, usually lower than the lattice symmetry. This opens the intriguing possibility that a given system could in principle exhibit different order parameters, each one being selected by changing an external variable such as temperature or magnetic field. This scenario does in fact exist, but is extremely rare, having been really established only in superfluid ^3He ¹ and in two superconductors: UPt_3 ^{2,3} and thorium-doped UBe_{13} ⁴. Here we show that pressure can induce this phenomenon in UTe_2 .

The recently discovered superconductivity in the heavy fermion system UTe_2 ⁵ shows several unusual properties, the most spectacular being reentrant superconductivity when magnetic fields as high as 60 T are applied in specific directions^{6,7}. Another intriguing property is the temperature dependence of the specific heat. Indeed, in all samples a large residual term, of $\sim 50\%$ of the normal state specific heat, seems to remain as the temperature approaches zero^{5,8}. UTe_2 is a paramagnetic heavy fermion system, although its ground state seems to lie close to ferromagnetism^{5,8}. The crystal structure is orthorhombic with an extremely anisotropic magnetic susceptibility at low temperatures, and a pronounced easy axis along the *a*-axis. When a magnetic field is applied along the hard *b*-axis, UTe_2 exhibits a metamagnetic transition at ~ 35 T^{9,10} and most spectacularly shows field-enhanced superconductivity up to this field⁶ and reentrant superconductivity at even higher fields for specific angles of the applied field⁷. In this, it shares some characteristics of the known ferromagnetic superconductors URhGe and UCoGe, which exhibit metamagnetism and enhanced or reentrant superconductivity when a magnetic field is applied perpendicular to the easy axis^{11,12}. However, in these systems the enhancement of superconductivity is related to the suppression of ferromagnetism with a transverse field so the case of UTe_2 is quite different. Field-enhanced superconductivity in a system without ferromagnetism offers further conceptual challenges and may well shed new light on this fascinating phenomenon. The remarkable properties and especially the superconductivity of these systems are undoubtedly linked to the proximity of a magnetic instability, and pressure is a powerful parameter to tune this proximity. Pressure can either push the system away from the instability, or,

more interestingly, move the system closer to and even cross the magnetic instability. In the vast majority of cases, superconductivity under pressure is studied by resistivity measurements. Zero resistance is the best-known characteristic of a superconductor, and is also easy to measure. But it is not necessarily the best probe, as zero resistance can be obtained with filamentary superconductivity or superconducting impurities and it is not proof of bulk superconductivity. Furthermore, once the resistance is zero, resistivity measurements are essentially blind to any further change of state that may take place inside the superconducting state. For this reason, the essential part of this study is obtained from calorimetry measurements, performed under pressure in a diamond anvil cell (see Methods). This technique, although not quantitative, gives information that is directly related to the specific heat of the sample. The calorimetry study is complemented by resistivity measurements that give extra information about the superconducting and normal states.

In this report, we show that the superconductivity is extremely sensitive to hydrostatic pressure as a tuning parameter and that UTe_2 is probably another example of multiple superconducting phases. The superconducting state found at zero pressure is monotonously depressed with pressure, but a second superconducting state is found to emerge as pressure is increased. Pressure increases the splitting between the two transitions and the high-temperature superconducting transition reaches nearly 3 K, a new record for a U-based heavy fermion superconductor. Pressure also drives the system towards a more correlated state, with evidence for a strong enhancement of the electronic effective mass. At a critical pressure of ~ 1.7 GPa both superconducting states are suppressed and a new order parameter, probably magnetic, is found.

Results

Calorimetry measurements. In Fig. 1a, we show the specific heat of a large crystal (8 mg) measured at ambient pressure compared to the measurement on the small crystal in the diamond anvil cell at zero pressure. For the large sample, the superconducting transition appears as a sharp step at 1.45 K. The sample is therefore homogeneous even though the critical temperature is slightly lower than some reported values. The low temperature side shows that C/T extrapolates to a residual value of $\sim 60\text{--}70$ mJ mol⁻¹ K⁻² as is found systematically in UTe_2 . As we mentioned already, the measurement in the diamond anvil cell is not

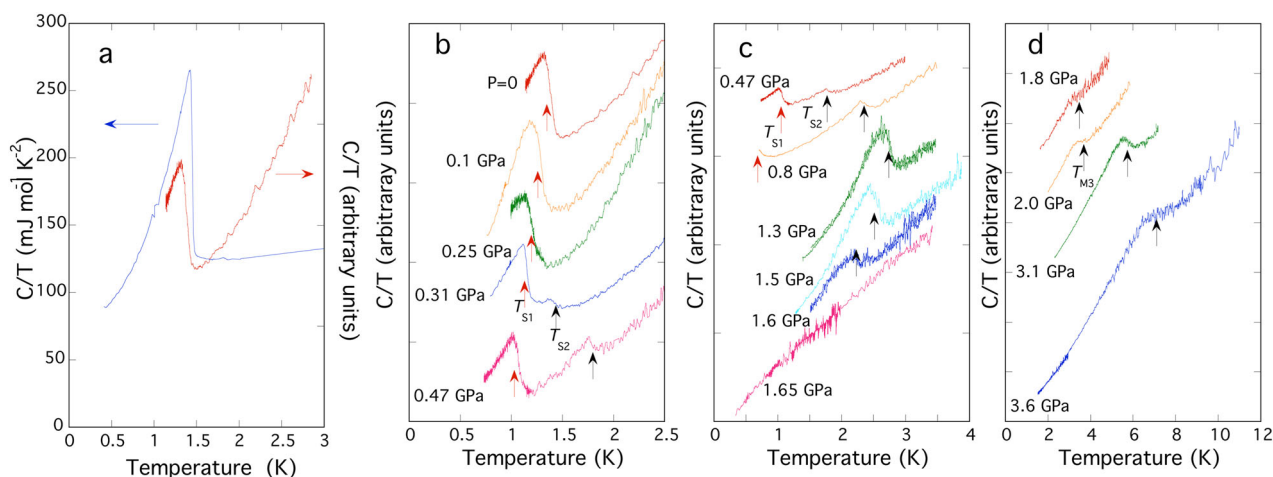


Fig. 1 Calorimetry results. Graph **a** shows the specific heat of a large sample from the same batch measured at ambient pressure and the measurement at zero pressure in the diamond anvil cell. Graphs **b**, **c**, and **d** show measurements under pressure in different pressure ranges. The curves have been shifted vertically for clarity.

quantitative. Still the transition shows up as a sharp anomaly, with a similar aspect to the ambient pressure measurement. It is slightly broadened due to some very small pressure, some degradation of the sample during the setup of the cell, or the measurement technique. The transition appears on top of a temperature-dependent background. In Fig. 1, graphs b, c, and d show the evolution of the temperature dependence of C/T for three pressure ranges. At low pressure, the anomaly at T_{S1} corresponding to the superconducting transition clearly shifts to lower temperatures. Above 0.3 GPa, a second anomaly appears at a higher temperature, labeled T_{S2} . T_{S2} then continues to increase with pressure. Figure 1c shows the intermediate pressure range: T_{S1} continues to decrease and falls below the lowest measurable temperature (~ 0.5 K) above 0.8 GPa. Above 1 GPa the anomaly corresponding to T_{S2} becomes much more pronounced. T_{S2} continues to increase, reaching a maximum of ~ 2.8 K at 1.3 GPa, then decreases rapidly, and the anomaly disappears above 1.5 GPa. However, as shown in Fig. 1d, as pressure is further increased above 1.8 GPa, a new anomaly appears at a temperature labeled T_{M3} , of ~ 3.5 K. This last anomaly, initially weak, becomes more pronounced with pressure and moves to higher temperatures.

Specific heat alone is not sufficient to identify the phases that appear. By continuity, there seems little doubt that T_{S1} corresponds to the superconducting phase seen at ambient pressure. We will show that the effect of magnetic field on this transition is also consistent with ambient pressure measurements. In order to gain information on T_{S2} and T_{M3} , we compare them with resistivity measurements.

Resistivity measurements. Figure 2 shows $\rho(T)$ curves for different pressures. The superconducting critical temperature increases with pressure, reaching a maximum of ~ 2.7 K between 0.5 and 1 GPa. This implies that the anomaly seen at T_{S2} in the specific heat is also due to superconductivity. On increasing the pressure, the superconducting critical temperature in the resistivity experiment starts to decrease, consistent with the specific heat results.

At higher pressure no superconductivity is seen, and a new anomaly appears. This corresponds to the temperature T_{M3} in the specific heat measurement and shows up as a small increase in resistivity as the temperature decreases. The most likely explanation is the appearance of magnetic order, although this remains to be checked by other types of measurements. The phase diagrams obtained from resistivity and specific heat are compared in Fig. 3. The spacing of the pressure points for the resistivity experiment was too large to resolve the initial decrease of the critical temperature. Also the calorimetry experiment found a strong increase in T_{M3} at high pressure, which was not observed in the resistivity, possibly because the maximum pressure was not high enough. There is also a slight difference in the pressure scale between the two experiments, probably due to the different pressure conditions (see Methods), but the two phase diagrams are consistent.

More information can be gained from the normal state temperature dependence of the resistivity, which can be fitted with a Fermi liquid behavior of the form $\rho = \rho_0 + AT^2$. In the right-hand graph of Fig. 2, we show the pressure dependence of the pre-factor A (Supplementary Fig. 2 and Supplementary note 2). The value of A can be approximately related to the electronic effective mass, usually obeying an empirical relationship¹³ $A^{1/2} \approx m^*$, although this relation may break down close to an instability. A increases quite significantly with pressure by a factor 6 between 0 and 1 GPa, and has a maximum ~ 1.3 GPa consistent with the existence of a critical pressure in this region.

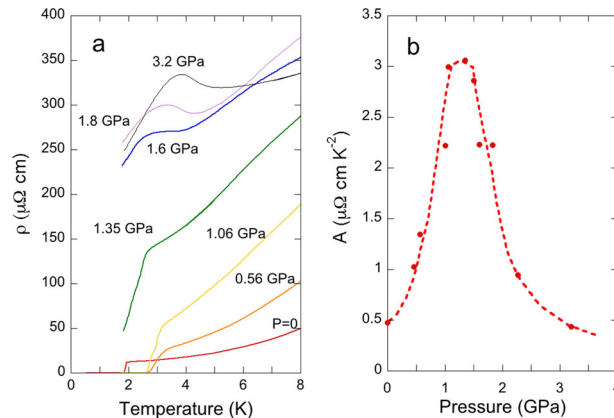


Fig. 2 Resistivity under pressure. Graph **a** shows the temperature dependence of the resistivity, ρ , for different pressures. Graph **b** shows the pressure evolution of the pre-factor A obtained by fitting the temperature dependence of the resistivity with a Fermi liquid law of the form $\rho = \rho_0 + AT^2$, where ρ_0 is a constant residual term. The dashed line is a guide for the eye.

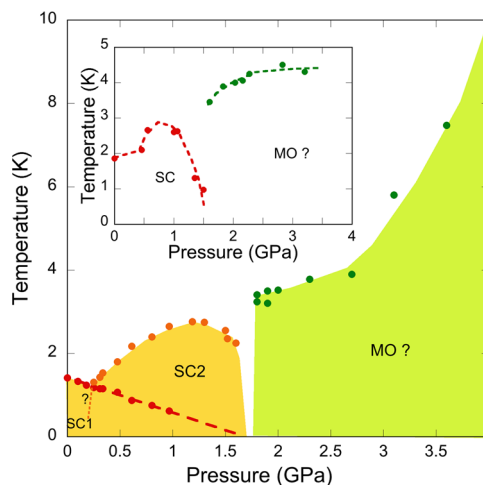


Fig. 3 Pressure-temperature phase diagram of UTe_2 . The main graph is obtained from specific heat measurements, showing the two superconducting phases, labeled SC1 and SC2, as well as the probably magnetically ordered high-pressure phase, labeled MO. The inset shows the similar phase diagram obtained from resistivity. Due to the wider spacing of the pressure points, the resistivity experiment was not able to resolve the initial decrease of the critical temperature. No transition was detected below T_{S1} at low pressure, but the phase diagram may contain a hidden phase boundary, as indicated by the question mark.

At higher pressures A decreases, although its determination is less reliable because of the influence of the magnetic transition, meaning that A has to be determined at higher temperatures. We note that this enhancement of A by a factor 6 is quite similar to that found under high fields at the metamagnetic transition⁹. The evolution of m^* with field has also been estimated from magnetization and magnetocaloric measurements and an enhancement of a factor 1.5–2 is found at the metamagnetic transition^{10,14}. Interestingly, it was shown that this enhancement of m^* , if due to an increase of the pairing strength, would imply an increase in the superconducting critical temperature to ~ 3 K^{10,14}, quite similar to the value we find under pressure.

Measurements under magnetic field. The effect of an applied field is also instructive. We could apply a field only along the

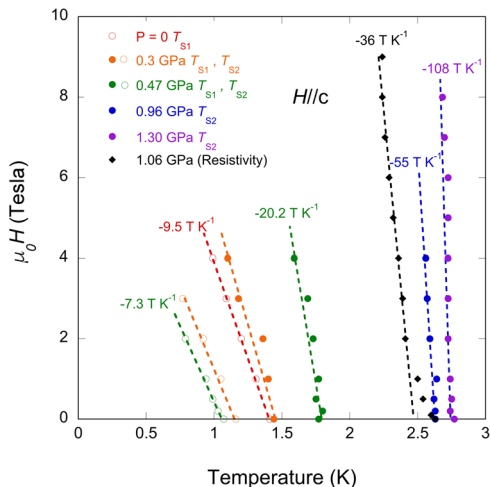


Fig. 4 Upper critical field. Field dependences of the two superconducting transition temperatures for different pressures. The open and closed symbols represent, respectively, the upper transition at T_{S2} and the lower one at T_{S1} . The field dependence of the superconducting transition measured by resistivity at 1.06 GPa is also included for comparison.

c -axis. In Fig. 4, we show the field dependence of the different transitions at several pressures. The slope of the upper critical field H_{C2} at zero pressure is consistent with previous reports^{5,8}. At low pressure this slope decreases slightly, scaling with T_{S1} as expected. When the second transition appears at T_{S2} , its field dependence is initially quite similar to that of T_{S1} . However, as pressure is increased the slope becomes extremely steep. The resistivity measurement also reveals a large increase in the slope. In a simple picture, the slope should scale with both the critical temperature and the square of the effective mass. The first term gives a factor of 1.8. Using the relation $m^* \approx A^{1/2}$ would imply an enhancement of a factor 6 for m^{*2} . This is the right order of magnitude to explain the increase by more than a factor 10 of the slope of H_{C2} ; however, this method probably largely overestimates the enhancement of m^* . Another approach is to reproduce the enhancement of T_{S2} between 0.3 and 1.3 GPa by an increase of the strong coupling constant λ . The expected slope can then be calculated by taking into account the renormalization of m^* due to the increase of λ . This accounts quite well for the increase of the slope of H_{C2} up to 1 GPa; however, it only accounts for $\sim 50\%$ of the maximum measured slope of over 100 T K^{-1} (Supplementary Fig. 1 and Supplementary note 1).

Discussion

In Fig. 3, we show the general phase diagram obtained from the specific heat measurements. The strong enhancement of the critical temperature, with T_{S2} reaching 2.8 K, makes UTe_2 the heavy fermion superconductor with the highest critical temperature among all U-based systems known so far, and close to the highest value for Ce-based heavy fermion systems¹⁵. Significantly higher values have only been found in neptunium- and plutonium-based heavy fermion compounds^{16,17}. The most striking result is the discovery of two superconducting states with different transition temperatures. As stated in the introduction, this is an extremely rare phenomenon. Both transitions are visible in the specific heat, implying that they are bulk phases. Pressure has opposite effects on the two transition temperatures: T_{S1} decreases while T_{S2} increases with increasing pressure. The effect of field applied along the c -axis on the two transitions, while initially similar, becomes also quite different at high pressure. These different pressure and field effects are strong arguments for the two phases

being of quite different nature. In general, multiple superconducting transitions are expected from either lifting the degeneracy of a multicomponent order parameter, or from two different irreducible representations of the space group that are accidentally nearly degenerate. In UPt_3 , the consensus nowadays favors the former^{18–21}. However, as UTe_2 is orthorhombic, all irreducible representations are one-dimensional so only the second scenario is allowed. This however means that the two transitions should cross rather than split, as suggested by the dotted line in Fig. 3, even though within the experimental sensitivity no indication of T_{S2} lying below T_{S1} is seen at low pressure. In this, UTe_2 resembles more the case of UBe_{13} , where a small amount of doping with thorium apparently splits the single superconducting transition into two separate transitions^{22,23}, but in fact an observed change in the order parameter implies the existence of a fourth line in the phase diagram even though no transition has been directly detected^{22,24,25}. We have sketched a similar line with a question mark in Fig. 3.

There is another intriguing possibility that allows only two superconducting phases. Generally, a phase diagram with three second-order transition lines meeting at a single multicritical point is thermodynamically forbidden except for the very special case where the specific heat jump on one of the lines vanishes at the crossing point and the slopes of the other two lines are identical (Supplementary Fig. 6 and Supplementary note 6). In fact, in our results the jump in C/T at T_{S2} is initially very small, and the slope of T_{S1} is identical on both sides of the meeting point, so possibly the solution with just three phase boundaries could be thermodynamically allowed here. Nevertheless, the small initial amplitude in the jump in C/T at T_{S2} is puzzling, and seemingly difficult to reconcile with a bulk transition between normal and superconducting states. This is in contrast to the cases of UBe_{13} or UPt_3 , where the two specific heat jumps have similar amplitudes. The steep slope of H_{C2} implies that the effective mass of the electrons condensing at T_{S2} is large, and so should contribute to the specific heat. We cannot exclude that for some reason only part of the sample volume is initially superconducting at T_{S2} , but the fact that the transition is detected in the specific heat means that this fraction is non-negligible. This could be due for example to a structural phase transition. We see no clear evidence for this (Supplementary Fig. 5 and Supplementary note 5), but a detailed study of the structure of UTe_2 under pressure is highly desirable. At higher pressure, the jump in C/T at T_{S2} becomes larger than that for T_{S1} at ambient pressure, and taking into account the probable increase in m^* their amplitudes are quite similar (Supplementary Fig. 4 and Supplementary note 4). Other results also point to the phase at T_{S2} being an intrinsic property of UTe_2 : The normal state resistivity shows that the appearance of the high pressure phase at T_{M3} and the strong increase of A are fully bulk effects, and the superconducting phase at T_{S2} is related to both of these, disappearing at the onset of the high-pressure phase, and with the maximum of T_{S2} being associated with the maximum of A . The small jump in C/T may be related to the still unsolved question of the residual specific heat at ambient pressure, which has been attributed to a non-unitary superconducting state, with only half of the electrons condensing⁵. While this point still has to be established, a possible explanation for our result could be that under pressure at T_{S2} an even smaller fraction of the electrons initially condenses, concerning just a small part of the Fermi surface, which progressively increases with pressure. An almost gapless order parameter could also be responsible for a reduced jump in C/T . What we miss at this stage is a complete entropy balance, which cannot be estimated because the AC calorimetry technique used here is not quantitative, and the measurements do not extend to low enough temperatures.

The other main point is that pressure clearly drives UTe_2 toward a more strongly correlated state. The strong increase of the A coefficient implies a large enhancement of the effective mass. At the same time, superconductivity becomes much more robust to a magnetic field applied along the c -axis, more than expected from the increase in the strong coupling constant: the most likely explanation for this anomalous slope is that the magnetic field actually enhances the pairing strength as seen in the ferromagnetic superconductors^{26,27}, and in UTe_2 at zero pressure. So far, a spectacular reinforcement of superconductivity has been found in UTe_2 for a field applied along the b -axis, and also at a specific angle in the b - c plane^{6,7}. Our results suggest that under pressure field-reinforced superconductivity probably also exists for a field along the c -axis. At a pressure of 1.7 GPa, no clear anomaly corresponding to any transition is seen down to ~ 0.5 K, the lowest temperature measured; however, at 1.8 GPa a new phase transition is observed at ~ 3.5 K, probably due to a magnetically ordered state. This is seen in both the specific heat and in the resistivity. The precise nature of this phase is an important question that we cannot resolve from the present data, although there are some clues. The resistivity shows a small but clear increase as temperature decreases through this phase transition. Such a phenomenon is usually associated with a change in the Fermi surface due to a change in the periodicity of the unit cell. It is therefore possible that the ordered state is not a simple ferromagnetic order. We can speculate that if some modulated structure develops, it could be responsible for the suppression of ferromagnetic fluctuations, and of superconductivity. Clearly, this initial study calls for many future experiments to determine the structure, the absolute values of specific heat, and the nature of the probable magnetic order in UTe_2 at high pressure.

UTe_2 is a fascinating system that challenges many of our ideas about heavy fermion superconductivity. Clearly, its high-pressure properties are no exception. UTe_2 under pressure combines two of the most intriguing effects found in unconventional superconductors, namely, multiple superconducting order parameters and field-reinforced superconductivity. These are associated with the highest known superconducting critical temperature for a U -based heavy fermion system. The similarities in the reinforcement of correlations and superconductivity with pressure and with high magnetic field are quite striking. Under pressure we find a field reinforcement of superconductivity for the field applied along the c -axis, whereas at ambient pressure this effect was only seen for field applied closer to the b -axis. This suggests a change in the magnetic and/or electronic anisotropies, and we can expect spectacular effects under combined pressure and very high fields. Further studies to obtain a more complete picture are now under way and will no doubt improve our understanding of these phenomena and perhaps bring more exciting discoveries.

Methods

Single crystals of UTe_2 were grown by vapor transport, as described in more detail here⁸. The ambient pressure specific heat was measured in a commercial device (Quantum Design PPMS). The AC calorimetry was measured in a diamond anvil cell²⁸. The transmitting medium was argon, ensuring very hydrostatic conditions in this pressure range. The sample was heated by a laser diode at a frequency of 637 Hz and its temperature oscillations were measured with a Au/Au:Fe thermocouple. The pressure was tuned in situ in a dilution refrigerator and measured with the ruby luminescence scale. Because of the heat load from the laser diode the lowest temperature achievable was ~ 0.5 K. The resistivity was measured in a modified Bridgman cell with ceramic anvils as described here²⁹. The pressure-transmitting medium was Fluorinert. The sample for resistivity came from a different batch and showed a higher transition temperature. Measurements were performed down to 1.8 K in a commercial device (Quantum Design PPMS). In both experiments a magnetic field of up to 8 or 9 T could be applied along the c -axis of the crystal. The theoretical analysis of thermodynamically allowed phase diagrams is performed by the Taylor expansion of the Gibbs free energy in the vicinity of the multicritical point in the P - T diagram. The evaluation of the pressure evolution of the slopes vs. the critical temperatures uses the same

strong coupling model as used for $UCoGe$ ²⁷ (Supplementary Fig. 3 and Supplementary note 3).

Data availability

The data that support the findings of this study are available from the corresponding author on reasonable request.

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Author contributions

D.B. supervised the project and performed the calorimetry measurements with F.H. M.V. performed the resistivity measurements with G.K. G.L. grew the single crystals with input from D.A. D.B. and M.V. analyzed the data. J.-P.B. performed the strong coupling analysis. M.E.Z. performed the thermodynamic analysis. D.B. wrote the manuscript with input from M.V., J.F., J.-P.B., A.P., M.E.Z., G.K., and D.A.

Competing interests

The authors declare no competing interests.

Additional information

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