Pressure Dependence of the Magnetization in the Ferromagnetic Superconductor UGe_2

C. Pfleiderer[1] and A. D. Huxley[2]

[1]Physikalisches Institut, Universität Karlsruhe, Wolfgang-Gaede-Strasse 1, D-76128 Karlsruhe, Germany
[2]Départment de Recherche Fondamentale sur la Matière Condensée, SPSMS, CEA Grenoble, 38054 Grenoble Cedex 9, France

(Received 22 February 2002; published 16 September 2002)

We report measurements of the pressure dependence of the low-temperature magnetization that show that the two pressure induced magnetic transitions in UGe_2 are of first order. Further, the pressure dependence of the uniform susceptibility relative to the superconducting transition is not as expected if the latter is driven by the proximity to a ferromagnetic quantum critical point. Our data instead suggest that the superconducting pairing could be associated with a sharp spike in the electronic density of states that is also responsible for the lower pressure magnetic transition.

DOI: 10.1103/PhysRevLett.89.147005 PACS numbers: 74.70.Tx, 71.27.+a, 75.50.Cc

The possible coexistence of superconductivity and ferromagnetism, although considered as a theoretical possibility for idealized weak itinerant ferromagnets over 20 years ago [1,2] has only recently been demonstrated to occur experimentally [3–5]. The theoretical calculations assumed the superconductivity to be mediated by an abundance of low-energy small–wave-vector magnetic excitations. These excitations become prevalent near a ferromagnetic quantum critical point (QCP), that is at the value of the pressure (or another control parameter) at which a second order transition is driven to zero temperature and at which the longitudinal magnetic susceptibility becomes singular. More recent theoretical work suggests that in an isotropic material a coupling between transverse and longitudinal excitations should give a much higher superconducting transition in the ferromagnetic state [6]. The presence of crystalline anisotropy has also been considered, and was shown to circumvent the depression of the superconducting critical temperature exactly at the QCP itself [7].

For UGe_2 it has already been established that in the limit of zero temperature the transition from ferromagnetism to paramagnetism as the pressure is increased through $p_c \approx 15.8$ kbar is first order [8]. This transition therefore does not correspond to a QCP. However, at lower pressures the temperature dependence of the magnetization shows a sharp change at a pressure dependent temperature $T_x(p)$ well below the Curie temperature. T_x decreases with p and vanishes at $p_x \approx 12.2$ kbar. The superconducting transition temperature, T_s, and superconducting coupling parameter are largest at pressures close to p_x [9]. This would be naturally explained in the spirit of the above theory if T_x were to correspond to a second-order transition, with p_x a QCP for this transition. A detailed explanation along these lines has indeed been proposed [10] in which T_x is identified with the formation of a simultaneous charge and spin density wave (CSDW). Theoretically the formation of a CSDW would lead to a change in the temperature evolution of the magnetic moment, as well as an enhancement of the longitudinal magnetic susceptibility [10] similar to that near to a simple ferromagnetic QCP. Although band structure calculations [11,12] indicate that a spin-majority Fermi-surface sheet could become nested as a function of the magnetic polarizaton, a necessary condition for a CSDW to arise, extensive neutron diffraction studies [13] have as yet failed to detect any static order due to a CSDW.

In this Letter we establish for the first time that the low T ordered moment (i.e., the ferromagnetic order parameter) and therefore a first-order derivative of the free energy changes abruptly at p_x. Thus there is unambiguously a first-order transition between two ferromagnetic phases at p_x and therefore no QCP. We will refer to the high pressure phase as FM1 and the low pressure phase as FM2.

Although the low-field low-temperature uniform longitudinal susceptibility undergoes a large change between FM1 and FM2, we show that it is almost pressure independent within each phase and is thus not correlated with $T_s(p)$ far away from p_x. Above p_x the transition FM1 $\rightarrow$ FM2 can be induced by a magnetic field. We find that the field at which the transition occurs, H_x, depends on p but the magnetic polarizaton at H_x is only weakly p dependent. This shows that the FM1 $\rightarrow$ FM2 transition occurs at a particular spin splitting between the majority and minority spin bands as would occur when the Fermi-energy passes through a sharp maximum in the electronic density of states for one spin direction. If virtual excitations to states at this maximum were also associated with the superconducting pairing mechanism a pairing spectrum peaked at finite energy (in the extreme limit an Einstein spectrum) would result. We show that this provides a natural relationship between T_s in zero field and the field necessary to induce the transition between the two magnetic phases for $p > p_x$. Thus the pressure dependence of T_s, which was the motivation for previously supposing that there was a QCP at p_x, can be explained without invoking a QCP.

Two different single crystals cut by spark erosion from larger crystals grown by the Czochralski technique were studied. The larger was a cylinder of diameter 2.4 mm and

 © 2002 The American Physical Society

length 5 mm parallel to the easy magnetic a axis, while the smaller was a plate also parallel to this axis (stuck to a small washer to fix its orientation in the pressure cell). Other parts of the larger crystal had previously been studied and found to have residual resistivity ratios of order 100 (current parallel to the b axis) [3,14]. The larger sample was also confirmed to become superconducting under pressure in a separate ac susceptibility measurement [15]. Here we do not distinguish further between the two samples since they gave equivalent results, with only small differences in the widths of the various transitions. The dc magnetization was measured with a nonmagnetic Cu:Be clamp cell using a methanol:ethanol (1:4) pressure transmitting medium in a commercial vibrating sample magnetometer (VSM). The pressure was determined from the superconducting transition of Sn. The empty pressure cell generated a very weakly T and H dependent background contribution that was smaller than 2% of the signal from the larger sample in the ferromagnetic state at low temperature. The data shown have been corrected for this background. The samples were seen to float freely within the pressure medium before and after the experiment, while their orientation was constrained by the bore of the Teflon sealing capsule in the cell (internal diameter 2.5 mm, external diameter 3.0 mm). Systematic errors in the overall calibration of the magnetometer when using the pressure cell mean that the absolute accuracy in determining the magnetization is only about 5%. We have therefore scaled the data for each sample by a constant factor close to unity to give the correct ordered moment of 1.5 μ_B/formula at zero pressure. The experimental error in measuring the relative changes of magnetization is in contrast much smaller, and smaller than the size of the data points used in the various figures.

In Fig. 1 the temperature dependence of the ordered magnetic moment, $\mu(T)$, in zero field at different pressures is shown (we use the symbol μ for the ordered moment extrapolated to zero field and M more generally for the magnetization in a field). A clear change in the T dependence of $\mu(T)$ occurs at T_x, as has been previously reported [14,16], where T_x decreases with increasing p and disappears as $p \rightarrow p_x \approx 12.2$ kbar. While T_C can be conveniently defined as the point where $-dM/dT$ is a maximum in a small field (we used 0.02 T) the determination of T_x is slightly more subjective and is taken as the position of a local maximum of $d\mu/dT$. The resulting p versus T phase diagram for UGe_2 constructed from the present measurements is shown in Fig. 2(a) along with $T_s(p)$ taken from Ref. [14]. T_x cannot be assigned from the present magnetization measurements below ~6 kbar. The position of a peak in the temperature derivative of the resistivity reported by Oomi *et al.* [17] can, however, be used to extend the T_x line to give $T_x \approx 30$ K at $p = 0$. In the following we focus on the pressure dependence of the magnetization at low T.

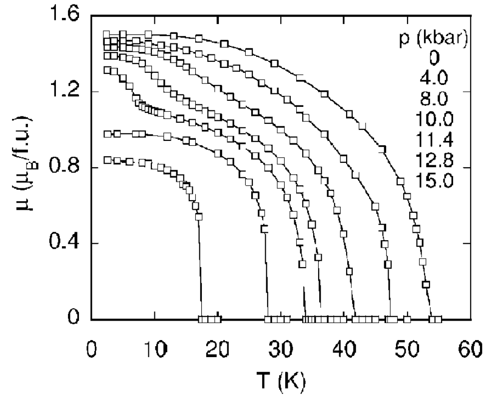

FIG. 1. Temperature dependence of the ordered ferromagnetic moment, μ, in the limit of zero field, deduced in the usual way from measured hysteresis loops. Curves correspond from top to bottom to the pressures indicated in the top right corner of the frame. The error bars are much smaller than the symbols.

The p dependence of the low T ordered moment μ at 2.3 K is shown in Fig. 2(b). Striking features are the abrupt changes of $\mu(p)$ on crossing p_x and p_c, respectively. This is the main new result. It shows that the transition from FM2 $\rightarrow$ FM1 at p_x is a first-order transition in the limit of $T = 0$, and confirms that the transition from the ferromagnetic state FM1 to the paramagnetic phase at p_c is also first order [8,14,18].

The field dependence of the magnetization at 2.3 K for different p is shown in Fig. 3. For pressure $p > p_x$ a large increase of nearly 50% in the magnetization is observed at a field H_x (H_x, defined as the field at which dM/dH has a local maximum is plotted as a function of pressure in Fig. 2(c)). For $p > p_c$ the magnetization undergoes a second increase at a lower field H_m corresponding to the transition from the paramagnetic phase to FM1. Interestingly, the uniform susceptibility given by the slope dM/dH has almost constant values independent of the pressure within each phase; in the FM1 phase it is greater than in the FM2 phase but less than in the paramagnetic state above p_c

The existence of metamagnetic behavior just above p_x is in itself an indication that the transition between the two magnetic phases is first order. We now consider further the transitions at H_x. Hysteresis loops of the dc magnetization in low fields show that the sample is already monodomain in a field of 0.02 Tesla and therefore no hysteresis would normally be expected at much higher fields of several Tesla. However, we observe hysteresis of a few mT (not visible on the scale of Fig. 3) at both H_m and H_x in careful measurements. The evidence for such hysteresis at H_x and H_m is demonstrated unambiguously by comparing the present data to measurements of the ac susceptibility χ_{ac}. In the inset of Fig. 3, χ_{ac} is shown as a function of field in the vicinity of H_x at a pressure of 15.7 kbar (from Ref. [14]). The amplitude of the peak in the ac susceptibility at 3 K is smaller than the derivative

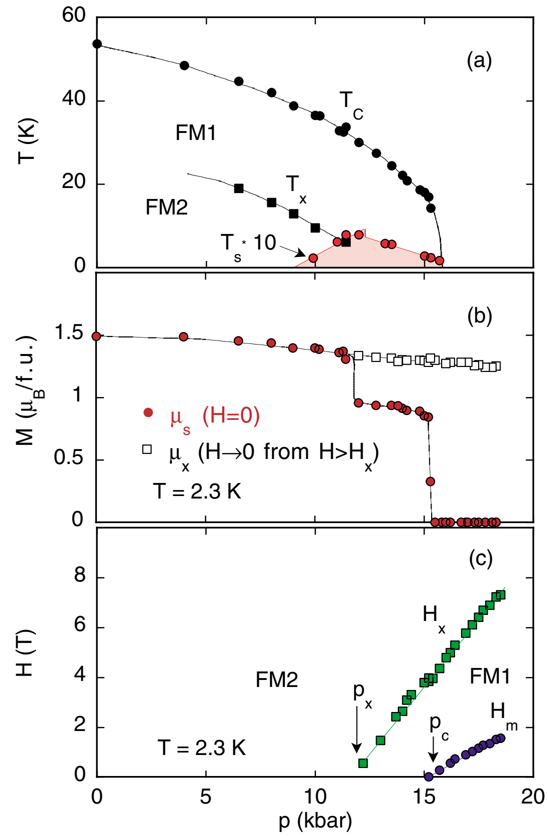

FIG. 2 (color online). (a) The p versus T phase diagram of UGe_2. T_C is the Curie temperature and T_x is defined in the text. T_s is the superconducting temperature (onset) from Ref. [14]. The lines through the data points are a guide to the eye, noting that T_s might change discontinuously at p_x and p_c. (b) The pressure dependence of μ in zero field at 2.3 K, (full circles). The moment obtained by extrapolating the data from above H_x to zero field (squares) is also shown when this is different. (c) the pressure evolution of the fields H_x and H_m of metamagnetic transitions (at which dM/dH has a local maximum) at 2.3 K.

of the uniform magnetization dM/dH at H_x, despite the fact that the peak in the ac measurement is slightly sharper than the dc transition width. Further, dM/dH at H_x, decreases with increasing T, whereas the amplitude of the peak in χ_{ac} increases with T (at least up to 5 K). This shows that the ac measurement traces minor hysteresis loops in the vicinity of H_x that become wider at lower T. The same result is also found for the transition at H_m [8]. The observation of hysteresis supports our previous conclusion that the transition between the FM1 and FM2 phases is first order at low temperature; for a first-order transition a phase can exist metastably in a limited region beyond that in which it is thermodynamically stable.

We now discuss the maximum of $T_s(p)$ near p_x, which was previously the main motivation to suppose that p_x marked a QCP. We focus on the FM1 phase (i.e. $p_x < p < p_c$) where the superconducting transitions are much sharper. For ferromagnetically mediated pairing T_s can

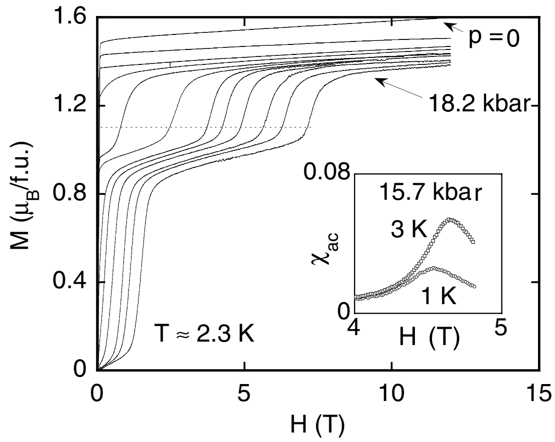

FIG. 3. The field dependence of the easy-axis magnetization at 2.3 K for various pressures. The broken line passes through the points H_x at which dM/dH has a local maximum. The magnetization at H_x is almost independent of pressure and suggests that the transition FM1 → FM2 occurs at a fixed value for the splitting between spin-majority and minority bands. Curves correspond from top to bottom to $p = 0, 6.5, 9.0, 11.1, 12.8, 13.8, 15.3, 15.5, 16.0, 16.7, 17.3,$ and 18.2 kbar. The inset shows the ac susceptibility in SI units measured as a function of field at 15.7 kbar at 1 and 3 K.

be estimated as $T_s = \theta\, e^{-\gamma/g\Delta\gamma}$, where $\Delta\gamma$ is that part of the linear temperature dependence of the normal state electronic heat capacity, γ, associated with the excitations responsible for pairing [19,20]. θ is the characteristic energy of these excitations and g the effectiveness of this pairing channel (we consider the superconductivity to be non s-wave with $g < 1$ and constant). In the usual description of itinerant ferromagnetism the spectrum of longitudinal magnetic excitations is assumed to be a Lorentzian peaked at zero energy (ω) and wave vector transfer (q) [20]. For such a spectrum and conventional q- and ω-independent mode-mode coupling $\Delta\gamma$ is directly related to the T dependence of μ^2 at low T. Our experiment shows that the temperature dependence is much weaker for pressures just above p_x than just below p_c (Fig. 1). $\Delta\gamma$ is therefore expected to increase significantly with p even though the static longitudinal susceptibility defined as dM/dH (Fig. 3) is experimentally almost independent of p between p_x and p_c. The latter point could still be reconciled with a Lorentzian spectrum if the width of the spectrum increases either in q or ω. However, experimentally γ is known to be almost constant between p_x and p_c [21] and thus $T_s(p)$ would also increase with p if superconductivity was indeed due to a Lorentzian spectrum of excitations. This is in stark contrast with the observed decrease of T_s with p. Thus a simple spectrum of longitudinal magnetic excitations of the type usually considered near a ferromagnetic QCP cannot account for our experimental observations.

In the following we outline a mechanism that qualitatively explains the observed pressure dependence of T_s

consistently with first-order transitions at p_x and H_x. The mechanism is based on our observation that the FM1 → FM2 transition occurs at a constant magnetization independent of the pressure. This strongly suggests that the transition takes place when the Fermi-energy crosses a sharp maximum in the electronic density of states (DOS) for one spin-polarizaton. In the FM1 phase an applied field parallel to the easy magnetic axis leads to an additional Zeeman splitting between the majority and minority spin bands, which drives the Fermi-energy through this maximum. $\mu_B H_x$ is then proportional to the energy of the maximum in the DOS relative to the Fermi-energy in zero field. If we suppose that the superconducting pairing involves virtual excitations that access the same feature in the DOS the pairing strength and therefore $\Delta\gamma$ decrease strongly as the feature becomes more remote from the Fermi-surface. Thus, for example, the decrease of T_s with p in the FM1 phase is naturally linked with the increase in H_x. Further support that the excitations responsible for pairing have a spectrum peaked at a finite energy proportional to H_x comes from the measured upper critical field for fields along the c-axis (i.e., perpendicular to the easy-axis). It has previously been shown that the temperature dependence of the upper critical field is well modelled by a strong coupling calculation assuming an Einstein spectrum for the pairing interaction [9]; the position of the peak in the spectrum obtained by fitting the measured upper critical field to this model increases with pressure in the FM1 phase as we have described.

Spectroscopic measurements capable of detecting a sharp peak in the DOS have not yet been reported. Quantum oscillation measurements as a function of p were however recently published [18] and so we briefly examine whether these can be reconciled with a sharp peak in the DOS. The striking feature in the quantum oscillation data is that the electronic masses of all the detected orbits are much higher in the FM1 phase than in the FM2 phase (some frequencies remain similar while others differ substantially). Large mass renormalizations in heavy fermion materials are usually attributed to a Kondo-like mechanism where narrow f-electron bands in the pure ordered system play the roles of the Kondo impurity states lying just below the Fermi-energy in the original Kondo analysis [22]. Assuming a similar mechanism is responsible for the large effective masses observed in the FM1 phase of UGe_2 the much smaller masses in the FM2 phase require a destruction of the mechanism. Such a destruction would indeed occur if the Fermi level were to cross one of the narrow bands responsible for the resonant mass enhancement.

To conclude, we note that understanding the emergence of new physical behaviors close to quantum criticality represents one of the central themes in contemporary studies of correlated electron physics. The case of a ferromagnetic QCP is particularly important since the order parameter is directly measurable by macroscopic techniques. However, we have shown that although superconductivity in UGe_2 is intimately related to a proximity to a magnetic phase transition there is no quantum criticality associated with the suppression of this transition to zero temperature at pressure p_x. The implication is that new ground states (in this case nonconventional superconductivity) can emerge in strongly correlated electron systems due to a much wider range of circumstances than has hitherto been supposed.

Discussions with J. Flouquet, H. v. Löhneysen, G. G. Lonzarich, V. Mineev, K. Miyake, E. Ressouche, A. Rosch, J. P. Sanchez, K. Sandeman, M. Uhlarz, and R. Vollmer are gratefully acknowledged. We also acknowledge financial support from the Deutsche Forschungsgemeinschaft, CEA–Direction des Sciences de la Matière, and the European Science Foundation FERLIN program.

[1] V. L. Ginzburg, Sov. Phys. JETP **4**, 153 (1957).
[2] D. Fay and J. Appel, Phys. Rev. B **22**, 3172 (1980).
[3] S. Saxena *et al.*, Nature (London) **406**, 587 (2000).
[4] C. Pfleiderer *et al.*, Nature (London) **412**, 58 (2001).
[5] D. Aoki *et al.*, Nature (London) **413**, 613 (2001).
[6] T. R. Kirkpatrick *et al.*, Phys. Rev. Lett. **87**, 127003 (2001).
[7] R. Roussov and A. J. Millis, Phys. Rev. B **63**, 140504 (2001).
[8] A. D. Huxley *et al.*, Physica (Amsterdam) **284B–288B**, 1277 (2000).
[9] I. Sheikin *et al.*, Phys. Rev. B **64**, 220503(R) (2001).
[10] S. Watanabe and K. Miyake, cond-mat/0110492; Physica (Amsterdam) **312B–313B**, 115 (2002).
[11] A. B. Shick and W. E. Pickett, Phys. Rev. Lett. **86**, 300 (2001).
[12] H. Yamagami, Physica (Amsterdam) **186B–188B**, 182 (1993).
[13] J. Flouquet *et al.*, J. Phys. Soc. Jpn. (Suppl. A) **70**, 14 (2001).
[14] A. D. Huxley *et al.*, Phys. Rev. B **63**, 144519 (2001).
[15] R. Vollmer, Ph.D. thesis, University of Karlsruhe, 2002.
[16] N. Tateiwa *et al.*, J. Phys. Soc. Jpn. **70**, 2876 (2001)
[17] G. Oomi *et al.*, Physica (Amsterdam) **206B & 207B**, 515 (1995).
[18] T. Terashima *et al.*, Phys. Rev. Lett. **87**, 166401 (2001); T. Terashima *et al.*, Phys. Rev. B **65**, 174501 (2002); R. Settai *et al.*, J. Phys. Condens. Matter **14**, L29 (2002).
[19] A. J. Millis *et al.*, Phys. Rev. B **37**, 4975 (1988).
[20] G. G. Lonzarich, in *Electron*, edited by M. Springford (Cambridge University Press, Cambridge, United Kingdom, 1997), Chap. 6.
[21] N. Tateiwa *et al.*, J. Phys. Condens. Matter **13**, L17 (2000).
[22] J. Kondo, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic Press, New York, 1969), Vol. 23, p. 183.