Pressure dependence of the magnetization in the ferromagnetic superconductor UGe$_2$

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The recent discovery that superconductivity occurs in several clean itinerant ferromagnets close to low temperature magnetic instabilities naturally invites an interpretation based on a proximity to quantum criticality. Here we report measurements of the pressure dependence of the low temperature magnetisation in one of these materials, UGe$_2$. Our results show that both of the magnetic transitions observed in this material as a function of pressure are first order transitions and do not therefore correspond to quantum critical points. Further we find that the known pressure dependence of the superconducting transition is not reflected in the pressure dependence of the static susceptibility. This demonstrates that the spectrum of excitations giving superconductivity is not that normally associated with a proximity to quantum criticality in weak itinerant ferromagnets. In contrast our data suggest that instead the pairing spectrum might be related to a sharp spike in the electronic density of states that also drives one of the magnetic transitions.

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The possible co-existence of superconductivity and ferromagnetism, although considered as a theoretical possibility for idealised 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-wavevector 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, which is present only in the ferromagnetic phase, 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 magnetisation 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 [10,11] indicate that a spin-majority Fermi-surface sheet could become nested as a function of the magnetic polarisation, a necessary condition for a CSDW to arise, extensive neutron diffraction studies [3] 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 $T_x(p)$ 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, $H_z$, depends on $p$ but the magnetic polarisation at $H_z$ 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 length 5 mm parallel to the easy magnetic \( a \)-axis, while the smaller was a plate also parallel to this axis (glued 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) \[12\]. The larger sample was also confirmed to become superconducting under pressure in a separate a.c. susceptibility measurement \[13\]. 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 d.c. magnetization was measured with a non-magnetic 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 constant factor close to unity to give the correct ordered moment of 1.5 \( \mu_B \) at zero pressure. The experimental error in measuring the relative changes of magnetisation 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, \( \mu \), 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.

![Graph showing temperature dependence of ordered ferromagnetic moment](image)

**FIG. 1.** Temperature dependence of the ordered ferromagnetic moment, \( \mu \), 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.

In Fig. 2. (a) The \( p \) versus \( T \) phase diagram of UGe\(_2\). \( T_C \) is the Curie temperature and \( T_s \) 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 \( \mu \) in zero field at 2.3 K. (full circles). The moment obtained by extrapolating the data from above \( H_s \) to zero field (squares) is also shown when this is different. (c) The pressure evolution of the fields \( H_s \) and \( H_m \) of metamagnetic transitions (at which \( dM/dH \) has a local maximum) at 2.3 K.

![Graph showing magnetic phase diagram and hysteresis loops](image)

**FIG. 2.** (a) The \( p \) versus \( T \) phase diagram of UGe\(_2\). \( T_C \) is the Curie temperature and \( T_s \) is defined in the text. \( T_c \) 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 \( \mu \) in zero field at 2.3 K. (full circles). The moment obtained by extrapolating the data from above \( H_s \) to zero field (squares) is also shown when this is different. (c) The pressure evolution of the fields \( H_s \) and \( H_m \) of metamagnetic transitions (at which \( dM/dH \) has a local maximum) at 2.3 K.
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 d.c. magnetisation in low fields show that the sample is already mono-domain 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 a.c. susceptibility, $\chi_{ac}$. In the inset of Fig. 3, $\chi_{ac}$ is shown as a function of field in the vicinity of $H_x$ at a pressure of 15.7 kbar (from reference [14]). The amplitude of the peak in the a.c. susceptibility at 3K is smaller than the derivative of the uniform magnetisation $dM/dH$ at $H_x$, despite the fact that the peak in the a.c. measurement is slightly sharper than the d.c. transition width. Further, $dM/dH$ at $H_x$ decreases with increasing $T$, whereas the amplitude of the peak in $\chi_{ac}$ increases with $T$ (at least up to 5K). This shows that the a.c. 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 $T_x(p)$ 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 $T_x$ can be estimated as $T_x = \theta e^{-\gamma g/\Delta^2}$, where $\Delta$ is that part of the linear temperature dependence of the normal state electronic heat capacity, $\gamma$, associated with the excitations responsible for pairing [11-20]. $\theta$ 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 ($\omega$) and wavevector transfer ($q$) [20]. For such a spectrum and conventional $p$- and $\omega$-independent mode-mode coupling $\Delta^2$ is directly related to the $T$ dependence of $\mu^2$ 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). $\Delta^2$ 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 $g$ or $\omega$. However, experimentally $\gamma$ is known to be almost

![Graph](image)

**FIG. 3.** The field dependence of the easy-axis magnetisation 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 magnetisation 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 a.c. susceptibility in S.I. units measured as a function of field at 15.7 kbar at 1 K and 3 K.

The $p$ dependence of the low $T$ ordered moment $\mu$ 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→FM1 at $p_x$ is a first-order transition in the limit of $T \approx 0$, and confirms that the transition from the ferromagnetic state FM1 to the paramagnetic phase at $p_c$ is also first order [14-18].

The field dependence of the magnetisation 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 magnetisation 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 magnetisation 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 $p_x$
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 magnetisation 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-polarisation. 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 $\Delta$ 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 a 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 \[20\] 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 \[13\] and so we briefly examine whether these can be reconciled with a sharp peak in the D.O.S. 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 renormalisations 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 behaviours 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 non-conventional superconductivity) can emerge in strongly correlated electron systems due to a much wider range of circumstances than has hitherto been supposed.

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