Re-entrant hidden order at a metamagnetic quantum critical end point

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Magnetization measurements of URu\(_2\)Si\(_2\) in pulsed magnetic fields of 44 T reveal that the hidden order phase is destroyed before appearing in the form of a re-entrant phase between \(\approx 36\) and 39 T. Evidence for conventional itinerant electron metamagnetism at higher temperatures suggests that the re-entrant phase is created in the vicinity of a quantum critical end point.

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Recent studies of itinerant electron magnetism in strongly correlated \(d\)- and \(f\)-electron metals have shown that metamagnetism gives rise to a new class of field-induced quantum phase transition \([1, 2]\). Sr\(_3\)Ru\(_2\)O\(_7\), CeRu\(_2\)Si\(_2\) and UP\(_3\) \([3]\) are all considered examples of systems that could possess a quantum critical end point, in which a notional line of first order phase transitions terminates at zero rather than finite temperature \([1]\). Here we propose that URu\(_2\)Si\(_2\) may be the first example of a system in which thermodynamic instabilities associated with the end point give rise to an ordered phase at high magnetic fields and low temperatures \([4]\). This behaviour is reminiscent of the creation of superconductivity in the vicinity of an antiferromagnetic quantum critical point in zero field \([2]\). We show that the presence of multiple magnetic transitions in URu\(_2\)Si\(_2\) at low temperatures \([4, 7]\) can be ascribed to re-entrant phenomena arising from the interplay between itinerant electron metamagnetism (IEM) and the hidden order (HO) parameter recently attributed to orbital antiferromagnetism \([7]\).

URu\(_2\)Si\(_2\) belongs to a class of strongly correlated metals in which \(f\)-electrons, rather than being localised and giving rise to magnetism, develop a distinctly itinerant character \([10]\). Coulomb interactions cause the quasiparticle effective masses to be heavily renormalised, making the energetic rewards for forming ordered ground-states substantially higher than in normal metals \([11, 12]\). Indeed, in addition to forming the HO phase at \(T_o \approx 17.5\) K \([9]\), URu\(_2\)Si\(_2\) becomes superconducting at \(T_c \approx 1.2\) K \([10]\). The presence of an itinerant \(f\)-electron Fermi surface \([13, 14]\) also furnishes URu\(_2\)Si\(_2\) with the essential preconditions for IEM \([12]\), by which the \(f\)-electrons revert to a localised behaviour upon their alignment in strong magnetic fields. IEM is considered to account for the increase in the magnetization by \(\approx 1\) \(\mu_B\) per U atom at magnetic fields between \(\approx 35\) T and \(\approx 40\) T, although the existence of multiple magnetic transitions has remained controversial \([4, 7]\). Recent observations that local moment antiferromagnetism occurs within a minority phase that is destroyed by fields in excess of 15 T \([4, 10]\) call for a re-examination of the bulk high magnetic field phenomena in URu\(_2\)Si\(_2\).

In this paper, we show that multiple transitions in URu\(_2\)Si\(_2\) can be explained by a scenario in which the magnetic field first destroys the HO phase before creating a new field-induced re-entrant phase \([4]\) in the vicinity of the metamagnetic transition (see Fig. 1 for a phase diagram). IEM is accompanied by a pronounced asymmetry between the occupancy of itinerant spin-up and spin-down \(f\)-electron states \([7]\), brought on by the sudden population of the spin-up component as it sinks below the Fermi energy \(\varepsilon_F\) at a magnetic field \(B_M\). Magnetization measurements reveal that the magnetic field-induced phase is accompanied by the opening of a gap in the spin-up \(f\)-electron band at \(B_M\). We argue that such a gap could be compatible with a spin-singlet order parameter that breaks translational symmetry, of which the orbital antiferromagnetic (OAF) phase (recently proposed by Chandra \textit{et al.} \([4]\) to explain the origin of the HO) is one such example.

Figure 2a shows examples of the magnetization \(M\) of URu\(_2\)Si\(_2\) measured in pulsed magnetic fields of up to 44 T at several different temperatures. The data are obtained using a wire-wound sample-extraction magnetometer in which the sample is inserted or removed from the detection coils in-situ. While the experimental curves in Fig. 2a appear similar to those measured by other groups \([4, 7]\), the phase diagram obtained in Fig. 1 upon extracting the positions of the maxima in the differential susceptibility \(\chi = \mu_0 \partial M / \partial B|_B\) at different temperatures is markedly different. In a recent study, Jaime \textit{et al.} \([4]\) noted that the magnetocaloric effect can cause severe variations in sample temperature in pulsed magnetic field experiments if the sample cannot exchange heat with the bath as the magnetic field \(B\) changes. This effect is particularly serious if the sample is too large, a poor thermal diffusivity isolates the sample, or if the field is swept too rapidly. Adequate isothermal equilibrium in pulsed magnetic fields could, however, be achieved by using a long-pulse magnet (with a field decay constant of \(\approx 0.25\) s) combined with a sample thickness of \(\approx 150\) \(\mu_m\) \([4]\). It is by making such provisions in the present study that we obtain a phase diagram that agrees more closely with specific heat measurements in static magnetic fields \([4]\).

The existence of IEM of a similar type to that observed
in Sr$_2$Ru$_2$O$_{7}$ [17], UPt$_3$ [18, 19] and CeRu$_2$Si$_2$ [20, 21] is evidenced at temperatures above $\approx 6$ K in Fig. 2a by the presence of a single broad maximum in $\chi$. The dashed line in Fig. 1 indicates that the location of this feature at $B_M \approx 37.9$ T does not change significantly with temperature. The rapid increase in the $\chi$ at $B_M$ with decreasing temperature, shown in Fig. 2a (filled squares), implies that the jump in $M$ sharpens with decreasing temperature. Such behaviour is consistent with the existence of a first order critical end point at a field $B_M$ at temperatures well below 6 K that is broadened by thermal fluctuations at higher temperatures [2]. Rather than diverging indefinitely, however, the maximum in $\chi$ vanishes below $\approx 6$ K on entering the field-induced ordered phase recently identified in specific heat measurements [2]. The fact that $B_M$ occurs within the field-induced ordered phase implies that fluctuations associated with the metamagnetic critical end-point could play a role in its formation. Strong fluctuations in the vicinity of quantum critical points can cause metals to become highly susceptible to order as a means of lowering energy [1, 2]. A well known example is provided by the creation of superconductivity in the vicinity of an antiferromagnetic quantum critical point [3].

The intense magnetic fields combined with formation of the field-induced phase below $\approx 6$ K in URu$_2$Si$_2$ make the process of identifying whether the critical end point would otherwise terminate at $T = 0$ less certain than with Sr$_2$Ru$_2$O$_{7}$ [1], UPt$_3$ [18, 19] or CeRu$_2$Si$_2$ [20, 21]. This normally requires evidence for non-Fermi liquid behaviour. Fortunately, the transition in the specific heat $C$ at $\approx 5$ K [4] appears to be first order (being of $\approx 0.25$ K in width, albeit without observable hysteresis), implying that the region above $\approx 6$ K is free from thermal fluctuations of the field-induced HO parameter. This region can therefore be investigated for non-Fermi liquid effects associated with IEM [5]. Transport studies are thus far incomplete, presently yielding only a broad maximum in the magnetoresistance at $B_M$ [5]. The strongly divergent behaviour of $\chi$ above $\approx 6$ K in Fig. 2a together with the approximately linearly decreasing variation in $C/T$ with $T$ [4] at $\approx 38$ T could, nevertheless, be possible indications of non-Fermi liquid behaviour. Similar types of behaviour in other $f$-electron systems have been ascribed to the presence of a non-Fermi liquid [4].

Changes in the value of $M$ through the transitions provides clues as to the nature of the ordered phase. For $B \lesssim 25$ T, $M$ is weakly dependent on temperature, exhibiting a predominantly Pauli paramagnetic response typical of heavy Fermi liquids [11, 12]. This, together with specific heat measurements above the ordering temperature [2] and de Haas-van Alphen measurements below the ordering temperature [13, 14], unambiguously establishes the existence of a heavy Fermi liquid with an effective Fermi energy of order 10 meV. In the itinerant $f$-electron picture, a heavy Fermi liquid results from mixing of the $f$-electrons with regular conduction electron states [11]. When IEM occurs, the spin-up component of the Fermi energy shifts by the Zeeman interaction to energies just below $\varepsilon_F$ at $B_M$ (see Fig. 3), causing $M$ to undergo a dramatic increase by as much as 1 $\mu_B$ per $f$-electron atom [15]. As a result, $f$-electrons that were mostly itinerant below $B_M$ become mostly aligned and localised at fields above $B_M$. The field $B_M$ corresponds to a situation where the Fermi energy $\varepsilon_F$ intersects the middle of the spin-up $f$-electron band causing it to be half occupied. This leads to an approximately temperature independent $M$ at $B_M$ (see Fig. 2a) but with $\chi$ increasing dramatically with decreasing temperature (see Fig. 2c). The continuation of the temperature independence of $M$ at $B_M$ below $\approx 6$ K, accompanied by an abrupt reduction in $\chi$, indicates that the field-induced ordered phase stabilises a situation where approximately half of the $5f$-electrons become localised while the other half remain itinerant. This type of behaviour implies the existence of a charge gap in the spin-up itinerant $5f$-electron band at $\varepsilon_F$, at $B_M$.

The formation of a charge gap in the spin-up $5f$-electron band is consistent with the existence of a singlet order parameter that breaks translational symmetry. Order parameters of the charge-density wave [22] and OAF [3] type both possess this essential property; the latter also breaks time reversal symmetry. They both involve singlet pairing of quasiparticles at a characteristic translational wavevector $Q$, where $Q$ is determined by details of the Fermi surface topology [3, 22, 23]. In fact, regardless of the pairing symmetry, any singlet order parameter that involves spatial variations in charge density, or relative charge densities between one or more electron channels, will lead to such a gap. Order parameters of this type are also amenable to the possibility of re-entrant behaviour (see below). Evidence that the HO and field-induced HO phases have a common origin may be provided by the field and temperature dependence of $\chi$. The transition into the HO phase is devoid of any pronounced features in the temperature and field dependence of $\chi$ at fields below $\approx 34$ T, while those into the field induced phase exhibit similar behaviour over a narrow interval between 36.8 and 37.1 T (see Fig. 1). Furthermore, all transitions into (or out of) both phases evolve into ones that are first order in the limit $T \to 0$, although actual magnetic hysteresis remains undetected. First order transitions give rise to pronounced maxima in $\chi$ and/or magnetocaloric heating as the field is swept. Some degree of similarity between the low and high magnetic field phases is also apparent in the temperature dependence of the $\chi$ in Fig. 2a at 34.0 T and 37.9 T respectively.

In order to understand how re-entrance of the HO parameter can occur, it is instructive first to consider the density of electronic states (DOS) within the ordered phase for $B < 35$ T, which has received most attention thus far [1, 2]. Figures 4a-e show a schematic of the evolution of the total DOS with $B$ with (black lines) and without (red lines) ordering. At $B = 0$ (Fig. 4a, red line), the spin-up and and spin-down Fermi surfaces are degenerate. The introduction of a periodic charge
potential $V(r \cdot Q_0)$ within the HO phase must therefore result in the independent formation of band gaps for both spins (black line). This process is only efficient at minimising the energy of the systems if a significant part of the DOS is gapped at $\varepsilon_F$. The introduction of $B$ in Fig. 3, however, causes the energies of the spin-up and spin-down bands to split, leading to spin-up and spin-down Fermi surfaces of different sizes. The efficiency by which $V(r \cdot Q_0)$ can gap both spins therefore becomes progressively worsened as $B$ is increased, leading to the weakening of the gap and, eventually, to its destruction in a manner analogous to that of reaching the Pauli limit of a singlet superconductor [2, 21, 22]. A previous magneto-resistance study has provided experimental evidence for weakening of the gap in fields of $\approx 25$ T [27]. Ultimately, the ordered phase must be destroyed at a critical field $B_c \leq B_M$, whereupon the spin-up and spin-down Fermi surfaces become extremely asymmetric. The effect of $B$ on translational symmetry-breaking spin singlet order parameters has been extensively modeled using mean field theory [24, 25, 26]. One possibility is that the transition evolves into one that is first order that terminates at a critical field $B_c = \Delta_0/\sqrt{2gH_B}$. Figure 3 depicts the density of electronic states at $E_F$, where the spin-up and spin-down $f$-electrons states have become clearly resolved and singlet gap formation is no longer favoured. Upon estimating the size of the order parameter using the BCS relation $2\Delta_0 = 3.52k_BT_o$ [24, 26] and inserting free electron parameters for the spin $g = \frac{1}{2}$ and $g$-factor $g = 2$, we obtain $B_c \approx 32$ T. This is of comparable order to the first-order-like transition at $T_o$ (27) obtained from the current measurements (see Fig. 1), as well as the specific heat [24]. Given that the product $sg\mu_B$ in $f$-electron systems can depart from the free electron value [11], this agreement may be merestilus. However, a further prediction of mean field theory is that both the transition temperature $T_o(B)$ as a function of $B$ [24] and the critical field $B_c(T)$ as a function of $T$ [26] intersect the axes in a perpendicular manner, and both can be expanded in series of even powers of $B$ and $T$ respectively. A plot of $T_o^2$ versus $B^2$ should therefore yield a line that intercepts both axes in an approximately linear fashion. This is confirmed in Fig. 3 upon making such a plot with actual URu$_2$Si$_2$ data. An interesting situation then develops in the vicinity of $B_M$, enabling the realisation of a re-entrant hidden order (RHO) phase with a modified translational vector $Q_\perp$. Strong magnetic fluctuations at $B_M$ can be associated with the vanishingly small energy that separates spin-up electrons in localised and itinerant states at $\varepsilon_F$ [24]. This, combined with the weak dispersion of the spin-up $f$-electron band, makes the system especially vulnerable to forming an ordered phase. Ordering is especially easy to realise if the periodic potential $V(r \cdot Q_\perp)$ becomes comparable to the bandwidth of the spin-up $f$-electrons, because it will succeed at gapping the entire spin-up density of $f$-electron states regardless of the value of $Q_\perp$ and regardless of the absence of well defined spin-up momentum quantum numbers. A significant amount of energy is gained by opening such a gap at $\varepsilon_F$ and this would then account for the observed narrow gap in the spin-up $f$-electron band (see Fig. 3) [1]. The value of $Q_\perp$ need only be optimised to match the topology of the spin-down Fermi surface, which continues to be present at $B_M$. Once $B > B_M$, ordinary Fermi liquid behaviour is expected to be restored, but with the spin-up $f$-electrons being fully polarized as depicted in Fig. 3. The pronounced asymmetry between spins in the Fermi liquid makes the formation of an ordered phase unlikely, enabling the emergence of a Schottky anomaly in the specific heat [24].

In summary, we present $M$ data which shows that the HO parameter is first destroyed by Zeeman splitting in a magnetic field but then restored in a re-entrant phase [3]. The $T$ and $B$ dependence of $\chi$ reveals that IEM plays a role in its re-entrance, possibly indicating that HO is restored in response to magnetic fluctuations in the vicinity of a metamagnetic quantum critical end point [1, 2]. If true, this could be the first observation of the creation of an ordered phase in the vicinity of a magnetic field-induced quantum critical end point. We propose the existence of separate HO and RHO phases characterised by a common spin-singlet translational symmetry-breaking order parameter with slightly different translational vectors $Q_0$ and $Q_\perp$.

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FIG. 1: The $B > 30$ T versus $T$ phase diagram of URu$_2$Si$_2$ combined with a colour intensity plot of $\chi$ measured at many different temperatures. Square, triangle and circle symbols mark $B_M$ and transitions into and out of the HO and RHO hidden order (RHO) phases respectively. The curved dotted lines depict the continuation of the phase boundaries revealed by specific heat and transport studies [4].

FIG. 2: a, $M$ of URu$_2$Si$_2$ at several different temperatures for $B$ applied along the $c$-axis. b, $\chi$ in the vicinity of $B_M$ at several temperatures above the re-entrant ordering temperature. c, $\chi$ at $B_M$ and at $B \approx 34$ T as a function of $T$.

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FIG. 3: a-e Schematics of the evolution of the total DOS in URu$_2$Si$_2$ with $B$ (as indicated) before (red lines) and after (black lines) formation of the HO or RHO phases. Prior to ordering, mixing between conduction electron states and $f$-electron states gives rise to a large “Abrikosov-Suhl resonance”-like feature [11]. f A plot of the transition temperature squared $T_o^2$ versus the magnetic field squared $B^2$ taken from specific heat and transport data in Reference [4].
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