Reentrant Hidden Order at a Metamagnetic Quantum Critical End Point

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Magnetization measurements of URu_2Si_2 in pulsed magnetic fields of 44 T reveal that the hidden order phase is destroyed before appearing in the form of a reentrant phase between ≈ 36 and 39 T. Evidence for conventional itinerant electron metamagnetism at higher temperatures suggests that the reentrant 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 a new class of field-induced quantum phase transition [1,2]. $Sr_3Ru_2O_7$, $CeRu_2Si_2$, and UPt_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_2Si_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 behavior is reminiscent of the creation of superconductivity in the vicinity of an antiferromagnetic quantum critical point in zero field [5]. We show that the presence of multiple magnetic transitions in URu_2Si_2 at low temperatures [6–8] can be ascribed to reentrant phenomena arising from the interplay between itinerant electron metamagnetism (IEM) and the hidden order (HO) parameter recently attributed to orbital antiferromagnetism [9].

URu_2Si_2 belongs to a class of strongly correlated metals in which f electrons, rather than being localized and giving rise to magnetism, develop a distinctly itinerant character [10]. Coulomb interactions cause the quasiparticle effective masses to be heavily renormalized, making the energetic rewards for forming ordered groundstates substantially higher than in normal metals [11,12]. Indeed, in addition to forming the HO phase at $T_0 \approx 17.5$ K [9], URu_2Si_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_2Si_2 with the essential preconditions for IEM [15], by which the f electrons revert to a localized behavior 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 ≈ 35 T and ≈ 40 T, although the existence of multiple magnetic transitions has remained controversial [6–8]. Recent observations that local moment antiferromagnetism occurs within a minority phase that is destroyed by fields in excess of 15 T [9,16] call for a reexamination of the bulk high magnetic field phenomena in URu_2Si_2.

In this Letter, we show that multiple transitions in URu_2Si_2 can be explained by a scenario in which the magnetic field first destroys the HO phase before creating a new field-induced reentrant 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 [15], brought on by the sudden population of the spin-up component as it sinks below the Fermi energy ε_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 *et al.* [9] to explain the origin of the HO) is one such example.

Figure 2(a) shows examples of the magnetization M of URu_2Si_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

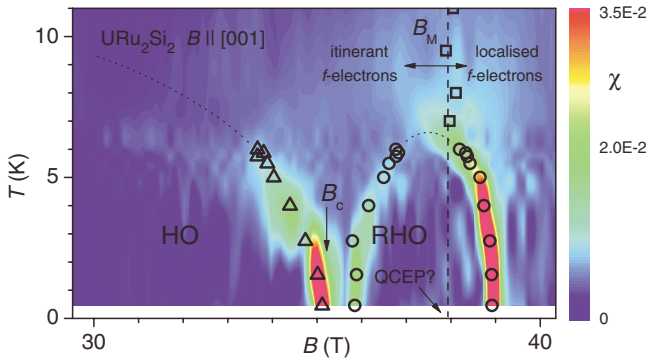

FIG. 1 (color). The $B > 30$ T versus T phase diagram of URu_2Si_2 combined with a color intensity plot of χ measured at many different temperatures. Square, triangle, and circle symbols mark B_M and transitions into and out of the HO and RHO hidden order (RHO) phases, respectively. The curved dotted lines depict the continuation of the phase boundaries revealed by specific heat and transport studies [4].

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which the sample is inserted or removed from the detection coils *in situ*. While the experimental curves in Fig. 2 appear similar to those measured by other groups [6–8], the phase diagram obtained in Fig. 1 upon extracting the positions of the maxima in the differential susceptibility $\chi = \mu_0 \partial M/\partial B|_T$ at different temperatures is markedly different. In a recent study, Jaime *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 ≈ 0.25 s) combined with a sample thickness of $\simeq 150$ μm [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 $Sr_3Ru_2O_7$ [17], UPt_3 [18,19], and $CeRu_2Si_2$ [20,21] is evidenced at temperatures above ≈ 6 K in Fig. 2(b) by the presence of a single broad maximum in χ. 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 χ at B_M with decreasing temperature, shown in Fig. 2(c) (filled squares), implies that the jump in M sharpens with decreasing temperature. Such behavior 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 χ vanishes below

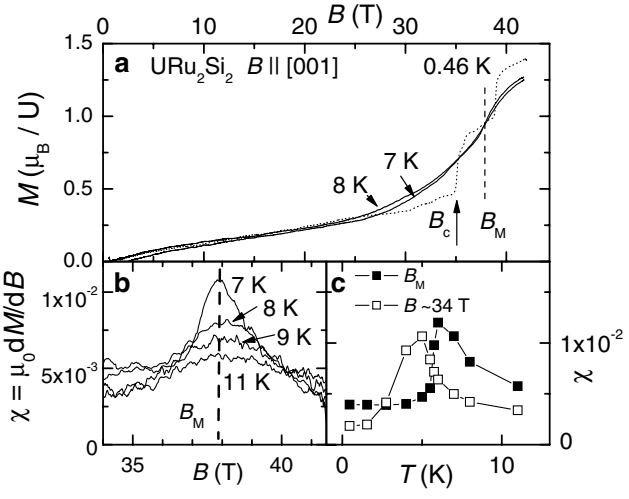

FIG. 2. (a) M of URu_2Si_2 at several different temperatures for B applied along the c axis. (b) χ in the vicinity of B_M at several temperatures above the reentrant ordering temperature. (c) χ at B_M and at $B \approx 34$ T as a function of T.

≈ 6 K on entering the field-induced ordered phase recently indentified in specific heat measurements [4]. The fact that B_M occurs within the field-induced ordered phase implies that fluctuations associated with the metamagnetic critical end point [1] 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 [5].

The intense magnetic fields combined with formation of the field-induced phase below ≈ 6 K in URu_2Si_2 make the process of identifying whether the critical end point would otherwise terminate at $T = 0$ less certain than with $Sr_3Ru_2O_7$ [1], UPt_3 [18,19], or $CeRu_2Si_2$ [20,21]. This normally requires evidence for non-Fermi liquid behavior. Fortunately, the transition in the specific heat C at ≈ 5 K [4] appears to be first order (being of ≈ 0.25 K in width, albeit without observable hysteresis), implying that the region above ≈ 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 [3]. Transport studies are thus far incomplete, presently yielding only a broad maximum in the magnetoresistance near B_M [4]. The strongly divergent behavior of χ above ≈ 6 K in Fig. 2(c) together with the approximately linearly decreasing variation in C/T with T [4] at ≈ 38 T could, nevertheless, be possible indications of non-Fermi liquid behavior. Similar types of behavior in other f-electron systems have been ascribed to the presence of a non-Fermi liquid [3].

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 [4] 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 itinerant f-electron band is shifted by the Zeeman interaction to energies just below ε_F at B_M [see Fig. 3], causing M to undergo a dramatic increase by as much as 1 μ_B per f-electron atom [15]. As a result, f electrons that were mostly itinerant below B_M become mostly aligned and localized at fields above B_M. The field B_M corresponds to a situation where the Fermi energy ε_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. 2(a)] but with χ increasing dramatically with decreasing temparture [see

Fig. 2(c)]. The continuation of the temperature independence of M at B_M below ≈ 6 K, accompanied by an abrupt reduction in χ, indicates that the field-induced ordered phase stabilizes a situation where approximately half of the $5f$ electrons become localized while the other half remain itinerant. This type of behavior implies the existence of a charge gap in the spin-up itinerant $5f$-electron band at ε_F, at B_M.

The formation of a charge gap in the spin-up $5f$-electron band is consistent with the existence of a spin-singlet order parameter that breaks translational symmetry. Order parameters of the charge-density wave [22] and OAF [9] type both possess this essential property; the latter also breaks time reversal symmetry. They both involve singlet pairing of quasiparticles at a characteristic translational wave vector $\mathbf{Q}$, where $\mathbf{Q}$ is determined by details of the Fermi surface topology [9,22,23]. 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 reentrant behavior (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 χ. The transition into the HO phase is devoid of any pronounced features in the temperature and field dependence of χ at fields below ≈ 34 T, while those into the field-induced phase exhibit similar behavior 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 $T \rightarrow 0$, although actual magnetic hysteresis remains undetected [4]. First order transitions give rise to pronounced maxima in χ and/or magnetocaloric heating as the field is swept [4]. Some degree of similarity between the low and high magnetic field phases is also apparent in the temperature dependence of χ in Fig. 2(c) at 34.0 and 37.9 T, respectively.

In order to understand how reentrance 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 the most attention thus far [1,9]. Figures 3(a)–3(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. 3(a) red line], the spin-up and and spin-down Fermi surfaces are degenerate. The introduction of a periodic charge potential $V(\mathbf{r} \cdot \mathbf{Q}_0)$ within the HO phase must therefore result in the independent formation of band gaps for both spins (black line). This process is efficient only at minimizing the energy of the systems if a significant part of the DOS is gapped at ε_F. The introduction of B in Fig. 3(b), 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 $V(\mathbf{r} \cdot \mathbf{Q}_0)$ can gap

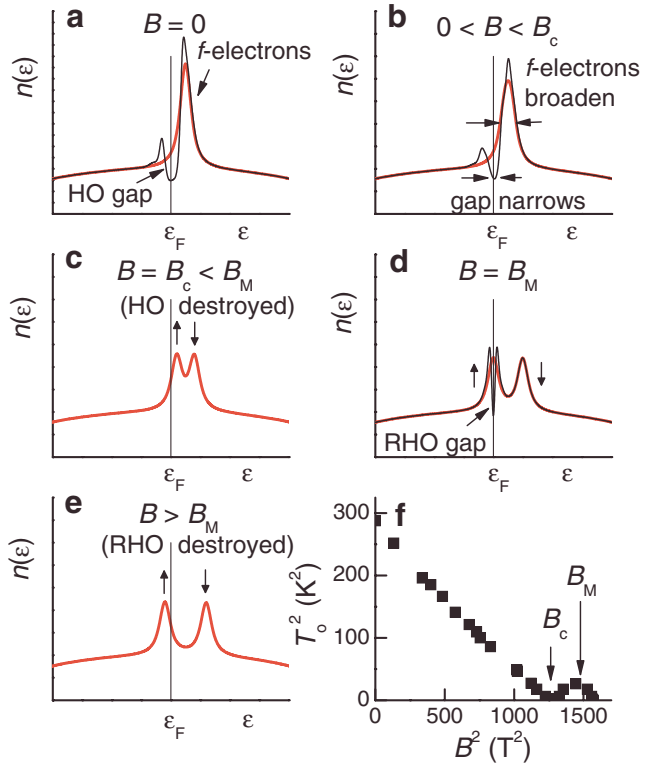

FIG. 3 (color). (a)–(e) Schematics of the evolution of the total DOS in URu_2Si_2 with B (as indicated) before (red lines) and after (black lines) formation of the HO or RHO phases. Prior to ordering, mixing between conduction electron states and f-electron states gives rise to a large "Abrikosov-Suhl resonance"-like feature [11]. (f) A plot of the transition temperature squared T_0^2 versus the magnetic field squared B^2 taken from specific heat and transport data in Ref. [4].

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 [24–26]. A previous magnetoresistance study has provided experimental evidence for weakening of the gap in fields of ≈ 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 [24–26]. One possibility is that the transition evolves into one that is first order that terminates at a critical field $B_c = \Delta_0/\sqrt{2}\sigma g \mu_B$. Figure 3(c) depicts the density of electronic states at B_c where the spin-up and spin-down f-electrons states have become clearly resolved and singlet gap formation is no longer favored. Upon estimating the size of the order parameter using the BCS relation $2\Delta_0 = 3.52 k_B T_0$ [24,26] and inserting free electron parameters for the spin $\sigma = \frac{1}{2}$ and g-factor $g = 2$, we obtain $B_c \approx 32$ T. This is of comparable order to the first orderlike transition at ≈ 35 T [4] obtained from the current

measurements (see Fig. 1) as well as specific heat [4]. Given that the product $\sigma g \mu_B$ in f-electron systems can depart from the free electron value [11], this agreement may be merely circumstantial. However, a further prediction of mean field theory is that both the transition temperature $T_0(B)$ 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 a series of even powers of B and T, respectively. A plot of T_0^2 versus B^2 should therefore yield a line that intercepts both axes in an approximately linear fashion. This is confirmed in Fig. 3(f) upon making such a plot with actual URu_2Si_2 data. An interesting situation then develops in the vicinity of B_M, enabling the realization of a reentrant hidden order (RHO) phase with a modified translational vector $\mathbf{Q}_\downarrow$. Strong magnetic fluctuations at B_M can be associated with the vanishingly small energy that separates spin-up electrons in localized and itinerant states at ε_F [2]. 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 realize if the periodic potential $V(\mathbf{r} \cdot \mathbf{Q}_\downarrow)$ 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 $\mathbf{Q}_\downarrow$ 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 ε_F, and this would then account for the observed narrow gap in the spin-up f-electron band [see Fig. 3(d)]. The value of $\mathbf{Q}_\downarrow$ need only be optimized 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 behavior is expected to be restored, but with the spin-up $f$electrons being fully polarized as depicted in Fig. 3(e). 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 [4].

In summary, we present M data which show that the HO parameter is first destroyed by Zeeman splitting in a magnetic field but then restored in a reentrant phase [4]. The T and B dependence of χ reveals that IEM plays a role in its reentrance, 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 characterized by a common spin-singlet translational symmetry-breaking order parameter with slightly different translational vectors $\mathbf{Q}_0$ and $\mathbf{Q}_\downarrow$.

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