Magnetic-Field-Induced Quantum Critical Point and Competing Order Parameters in URu_2Si_2

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A comprehensive transport study, as a function of temperature and continuous magnetic fields of up to 45 T, reveals that URu_2Si_2 possesses all the essential hallmarks of quantum criticality at fields around 37 ± 1 T. The formation of multiple phases at low temperatures at and around the quantum critical point suggests the existence of competing order parameters.

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A common picture emerging in strongly correlated metals is that exotic superconductivity may have as much to do with quantum criticality [1–3] as with unconventional pairing mechanisms [4,5]. *Quantum criticality* refers to our emerging understanding of phase transitions that occur at zero temperature (quantum critical points) in which quantum fluctuations play the key role. The physical properties of such systems can then be dominated by the quantum fluctuations even at very high temperatures up to and above room temperature [6]. Furthermore, the abundance of low energy excitations that persist near quantum critical points provides a perfect recipe for instability; thereby providing opportunities to form new phases that would otherwise not exist. The proximity to a quantum critical point (QCP) is known to give rise to novel ground states, such as magnetism [7] and unconventional forms of superconductivity [4,5]. It might also explain high-temperature superconductivity and non-Fermi-liquid behavior observed in the cuprates [8].

A QCP can be explicitly produced by depressing a second order phase transition toward absolute zero, by tuning an external control parameter, such as hydrostatic pressure [4], chemical composition [9], or magnetic field [10,11]. More recently, a new type of field-induced quantum criticality has been observed in systems, such as $Sr_3Ru_2O_7$ [12], that does not involve broken symmetry phases. Rather than being associated with an ordered phase, the QCP in $Sr_3Ru_2O_7$ is proposed to originate from itinerant electron metamagnetism at a critical field H_M. A first order quantum critical end point is produced by depressing the terminating point of a line of first order transitions to zero [13], giving rise to a broad crossover at finite temperatures. A qualitatively similar type of quantum criticality is proposed to occur in itinerant heavy fermion systems $CeRu_2Si_2$ [14] and UPt_3 [15] in which a distinct change in Fermi surface topology is known to occur at H_M. While the precise nature of the metamagnetic QCP has yet to be elucidated by theory, the experimental protocols for itinerant electron metamagnetic quantum phase transitions appear to be well established. The magnetoresistance undergoes a pronounced maximum in the vicinity of H_M, and fits of the function $\rho = \rho_0 + AT^2$ to the lowest temperature resistivity data yield a Fermi-liquid parameter A that diverges near H_M [12,14], corresponding to a divergence in the quasiparticle effective mass $m^* \propto A^{1/2}$ and vanishing of the effective Fermi energy $\varepsilon_F \propto A^{-1/2}$.

Since quantum criticality is becoming increasingly recognized as a universal phenomenon in condensed matter physics [16], it is of paramount importance to categorize and understand how field-tuned QCPs might stabilize new phases. While new phases in the vicinity of a field-tuned itinerant metamagnetic QCP were not found in $Sr_3Ru_2O_7$, $CeRu_2Si_2$, or UPt_3, URu_2Si_2 has been recently suggested as a possible candidate [17]. Here, we show that the electrical resistivity of URu_2Si_2 does indeed exhibit strong indications of quantum criticality, at fields and temperatures around the recently discovered ordered phase. This establishes the existence of quantum fluctuations involving the itinerant electrons at the Fermi surface. The low temperature phase diagram also reveals more intriguing phases than previously reported. Extensive hysteresis between multiple first order phase transitions at low temperatures could be a consequence of competing order parameters.

The resistivity $[\rho(H, T)]$ measurements of URu_2Si_2 as a function of both T and H, that form the basis of this work, were performed in the 45 T hybrid magnet at the National High Magnetic Field Laboratory in Tallahassee. H was swept mostly at a rate of 2 T/min, during the measurements, and was reduced to 0.1 T/min between $\mu_0 H = 33$ and 40 T to confirm the integrity of the H-dependent hysteresis. The standard four-probe method was employed with the current and H applied along the c axis of an oriented barlike URu_2Si_2 single crystal, with T between 0.5 and 20 K measured using a precalibrated capacitance. Identical resistivity behavior was observed in a second sample.

All of the evidence for quantum criticality and multiple phase transitions in URu_2Si_2 is presented in Fig. 1. Region I identifies the upper field limit of the enigmatic hidden order (HO) phase [18–23]. Specific heat studies [24,25] recently revealed the transition temperature $T_0 \approx 17$ K into this phase to be suppressed with increasing field, terminating at a critical field $\mu_0 H_I = 35.0 \pm 0.3$ T. This was then followed by a new ordered phase, region III, at slightly higher magnetic fields, which was subsequently shown to lie below a metamagnetic crossover field at higher temperatures [17], indicated by squares in Fig. 1(a). Region IV was proposed to be a field-induced recovery of the normal metallic phase, with some or all of the f electrons aligned by H. Each of the previously known [24,25] phase boundaries into phases I and III are reproduced in the current study by plotting extremities in the derivatives of $\partial\rho/\partial H$ and $\partial\rho/\partial T$ [solid colored circles in Fig. 1(a)]. Examples of raw $\rho(H)$ and $\rho(T)$ data, from which these phase boundaries are extracted, are shown in Figs. 2 and 3, respectively. The high sensitivity of ρ to changes in the ground state of URu_2Si_2 enables us to identify another phase II, recently detected also in ultrasound velocity measurements [26]. In specific heat measurements [24,25], the high-temperature limit of this phase matches with a kink in C/T, from which begins a plateau in C/T at lower temperatures. Most strikingly, however, our resistivity measurements find a complex series of sharp phase transitions (orange lines) and a new ordered phase, V. All phase transitions are found to become hysteretic as a function of H for $T <$ 3 K, unambiguously establishing them to be first order. Open and solid circles in the inset to Fig. 2 depict increasing and decreasing H, respectively.

It was conjectured on the basis of a magnetization study that phase III could be created in the vicinity of a quantum critical end point [17]. While a direct causality link between quantum criticality and phase III remains unproven, the present measurements do reveal a QCP involving the itinerant electrons, similar to that observed in other itinerant electron metamagnets.

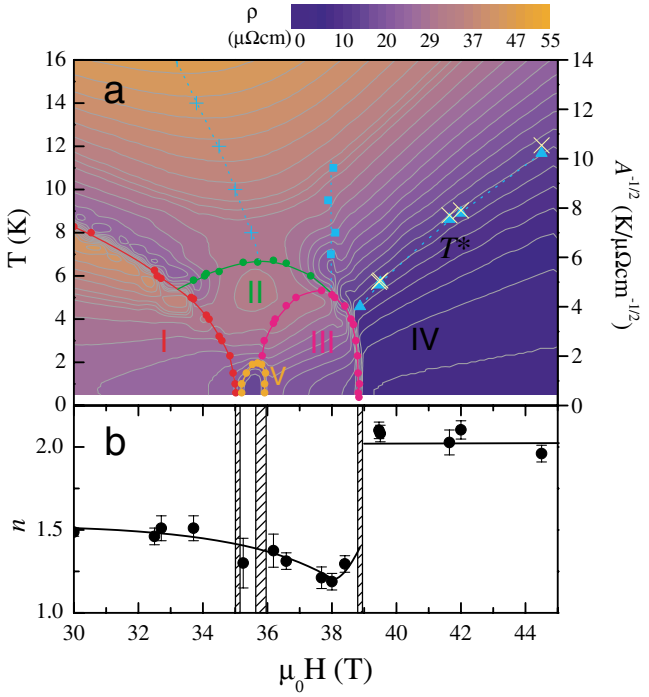

FIG. 1 (color). (a) High field-phase diagram of URu_2Si_2 obtained from our ρ vs H and ρ vs T data combined with a contour plot of the resistivity in which contour values are indicated. Solid colored circles (connected by colored lines) denote phase transitions extracted from extremities in $\partial\rho/\partial H$ (from H-decreasing sweeps) and $\partial\rho/\partial T$ (T sweeps); examples of raw data are shown in Figs. 2 and 3. + symbols indicate the broad maximum in $\rho(H)$ observed at higher temperatures, solid squares denote the high-temperature metamagnetic transition field from magnetization data [17], while solid triangles denote the crossover temperature, T^*, to T^2 behavior in the Fermi-liquid region IV. Dashed lines show the results of fits as described in the text. Region I refers to the hidden order phase, while II, III, and V constitute newly discovered phases. n vs H and $A^{-1/2}$ vs H ($\times$ symbols) are plotted in (b) and in the right axis of (a), respectively, following fits of $\rho(T)$ for ~ 0.6 K $\leq T \leq 3$ K in Fig. 3 to the formula $\rho = \rho_0 + AT^n$. It is noted that $A^{-1/2}$ is proportional to T^*. The hatched regions refer to fields where the fitting could not be performed due to hysteresis.

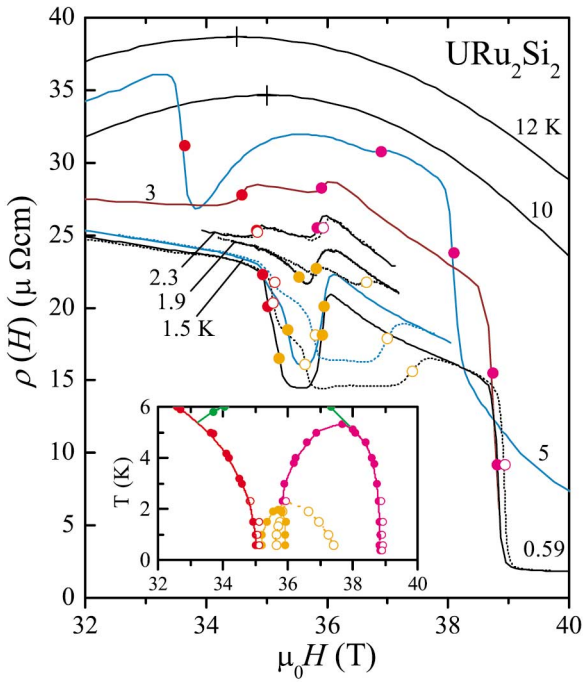

FIG. 2 (color). Examples of ρ vs H close to the QCP in URu_2Si_2 at selected temperatures and intervals in H. Solid and open circles indicate extremities in $\partial\rho/\partial H$ observed on falling and rising field sweeps, respectively. The phase boundary lines extracted on rising and falling field sweeps (and on sweeping T) are shown in the inset, evidencing significant hysteresis in phase V.

We begin by considering the broad magnetoresistivity maximum centered on ≈ 34 T at 12 K in Figs. 1(a) and 2. If we discount the elliptical region occupied by the HO phase (below ≈ 17 K and ≈ 35 T), this maximum (indicated by + symbols) narrows and systematically shifts to higher fields as the temperature is reduced down to ~ 6 K. Its location extrapolates to $\mu_0 H_{T=0} = 36.3 \pm 0.3$ T in Fig. 1(a) upon fitting its T versus H locus to a simple power law of the form $T \propto |H - H_{T=0}|^{\alpha_m}$, where $\alpha_m = 0.51 \pm 0.03$: power laws of this form are of common usage in the scaling theory of quantum phase transitions [6]. Similar resistivity maxima occur in $Sr_3Ru_2O_7$ [12], $CeRu_2Si_2$ [14], and UPt_3 [15] near their itinerant metamagnetic transitions; however, they persist down to the lowest temperature of ~ 0.1 K without any signs of order, in contrast to the present URu_2Si_2 results.

More definitive evidence for quantum criticality is found at fields greater than 39 T within region IV. While at low temperatures, region IV can be characterized as a good metal (see Fig. 3) for which $\rho = \rho_0 + AT^n$ with $n \approx 2$, typical of a normal Fermi liquid [27] (see Fig. 1 for low temperature values of A and n), above 6 K the exponent switches to $n \lesssim 1$, which is atypical of a Fermi liquid at these low temperatures. This crossover gives rise to a broad maximum in $\partial\rho/\partial T$ which we denote as a characteristic temperature T^*, delineated by triangles in Fig. 1(a). A power law fit of the form $T^* \propto |H - H_{T=0}|^{\alpha^*}$ (where $\alpha^* = 0.61 \pm 0.03$) extrapolates to the field axis at $\mu_0 H_{T=0} = 37.3 \pm 0.3$ T inside phase III. The field dependence of the Fermi-liquid parameter A below T^* also exhibits divergent behavior on approaching $H_{T=0}$. A plot of $A^{-1/2}$ ($\propto \varepsilon_F$ for a Fermi liquid) versus H in Fig. 1(a) yields a line that extrapolates to zero within phase III. More specifically, a fit of $A^{-1/2} \propto |H - H_{T=0}|^{\alpha}$ to the data yields $\mu_0 H_{T=0} = 36.8 \pm 0.6$ T, where $\alpha = 0.71 \pm 0.09$. Given that $A^{-1/2} \propto \varepsilon_F$, the apparent proportionality between $A^{-1/2}$ and T^*, shown in Fig. 1(a), implies that T^* must scale with the effective Fermi energy ε_F which then extrapolates to 0 at ~ 37 T. This corresponds to a divergence in $m^* \propto \varepsilon_F^{-1}$ and may explain the divergent behavior seen in the susceptibility χ [17].

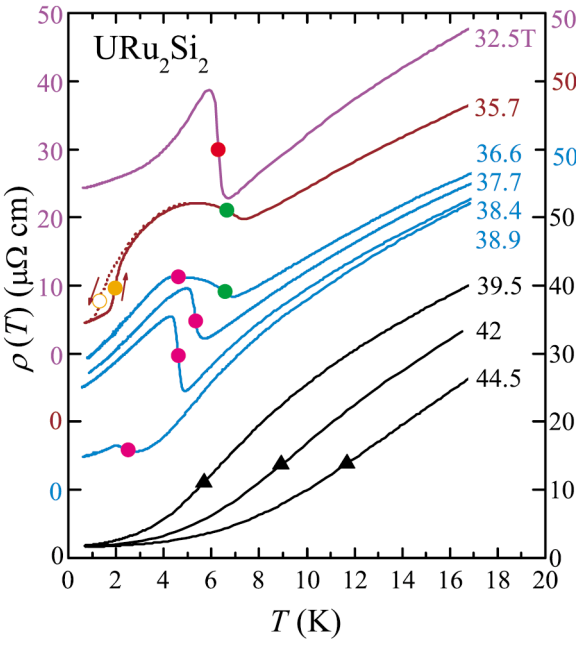

FIG. 3 (color). ρ vs T data of URu_2Si_2 at various constant magnetic fields, where shifts of 10 $\mu\Omega$ cm are made as indicated for clarity. All data are for increasing temperature except where the hysteresis is identified at $\mu_0 H = 35.7$ T at ~ 1.7 K, indicating that hysteresis also occurs as a function of T on entering or exiting phase V.

A universal property of quantum fluctuations is the existence of non-Fermi-liquid behavior in the temperature dependence of the resistivity within a sector of phase diagram emerging roughly in the form of a $\vee$ shape above the critical point [1]. The present resistivity data yield sublinear (i.e., $n \leq 1$) behavior on fitting $\rho = \rho_0 + AT^n$ to its temperature dependence above ~ 6 K, which is consistent with such behavior. The persistent metallic behavior within the various ordered phases enables the effect of critical fluctuations on the quasiparticles to be studied in the presence of ordering. Fits of $\rho = \rho_0 + AT^n$ over the temperature interval between $\sim 0.6 \leq T \leq 3$ K at many different fields in URu_2Si_2, reveal a continuous drop in n from ~ 1.5 at 30 T to ~ 1.1 at ≈ 38 T. The normal Fermi-liquid value of $n = 2$ is recovered in a discontinuous fashion only when $\mu_0 H > 39$ T. While it is conceivable that Fermi-liquid behavior could also be recovered below 0.6 K for $\mu_0 H < 39$ T, exponents that depart significantly from $n = 2$ at low temperatures could also be interpreted as evidence for non-Fermi-liquid behavior [28]. The continuous trend in n for $\mu_0 H < 39$ T suggests that the nature of the order parameter in all of the ordered phases is related. The presence of the anomalous resistivity exponent $n \sim 1$ centered on 38 T further suggests that, rather than being completely quelled by ordering, quantum critical fluctuations could continue to play a role in URu_2Si_2 at low temperatures. Such behavior is reminiscent of the situation in the high-temperature superconductors, where spin-fluctuation effects persist inside the superconducting phase [29].

Collectively, all of the above findings (as summarized in Fig. 1) establish the existence of a field-induced quantum critical point in URu_2Si_2 at $\mu_0 H = 37 \pm 1$ T. These findings can be listed as follows: (i) a single metamagnetic transition observed at temperatures above ~ 6 K at ~ 37.9 T; (ii) a single broad maximum in $\rho(H)$ above ~ 6 K (+) that converges with the metamagnetic transition upon extrapolation to $T = 0$; (iii) a crossover temperature T^* (from $n \lesssim 1$ to 2) at fields above the metamagnetic transition that converges with both the metamagnetic transition and the magnetoresistance maximum upon extrapolation to $T = 0$; (iv) an apparent divergence in the Fermi-liquid parameter A within phase

III occurring at the same field where the above features converge; and (v) a gradual reduction of the resistivity exponent n from ~ 1.5 to ~ 1 as H is swept through successive phases, reaching its lowest value at ~ 38 T in Fig. 1(b).

The close similarities in the underlying quantum critical behavior in URu_2Si_2 to those in $CeRu_2Si_2$ [14] and UPt_3 [15] implies that the mechanisms for quantum criticality are probably related. In both these systems, metamagnetism is connected with a change in Fermi surface topology that occurs when the magnetic field causes the f electrons to revert from itinerant to localized behavior. However, *the formation of multiple phases near the metamagnetic QCP is presently unique to* URu_2Si_2. The location of the QCP inside phase III (and its close proximity to phases II and V), suggests the possibility of a causality relationship between the itinerant electron metamagnetic QCP and phase formation, like that proposed to occur for superconducting phase formation in the heavy fermion antiferromagnets under pressure [4]. Two effects are thought to participate in the formation of new phases at QCPs. The first is the divergency in m^*, or, equivalently, the divergency in the density of electronic states. Such a divergency is energetically unfavorable, enabling the system to easily lower its energy by the opening of an energy gap associated with an ordered phase. The above resistivity measurements reveal such a divergency to be present in URu_2Si_2 prior to the formation of the ordered phase(s). The second effect is that pertaining to the existence of fluctuations at $T = 0$ capable of mediating novel pairing interactions. Theoretical models have yet to ponder the questions as to whether this can be true for a metamagnetic system in which the primary fluctuations involve the magnetization [3]. However, the existence of multiple interactions is something that is already known to be true in URu_2Si_2 even in the absence of a magnetic field. In addition to being a superconductor at ~ 1.8 K [18], URu_2Si_2 also develops the HO (phase I) parameter below 17.5 K [23] that is further known also to compete with antiferromagnetism [30].

Finally, the extensive hysteresis between phases II, V, and III at low temperatures implies that these ordered phases all have similar energies over an extended interval in magnetic field. Their corresponding order parameters Δ_{II}, Δ_V, and Δ_{III} must each be capable of lowering the energy of the system over a region in field at least as wide as that over which the hysteresis occurs. Given their potential to overlap with the QCP at 37 ± 1 T, it is conceivable that these order parameters compete to lower the total free energy of the system at finite temperatures. The absence of a single dominant order parameter in URu_2Si_2 is something that may be equally true in the cuprates as well as the heavy fermion superconductors [1]. Our ability to resolve competing mechanisms in the present study might result from the quantized manner in which H couples to the various orbital and spin degrees of freedom in URu_2Si_2.

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