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High Magnetic Field Studies of the Hidden Order Transition in URu_2Si_2

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We studied in detail the low temperature/high magnetic field phases of URu_2Si_2 single crystals with specific heat, magnetocaloric effect, and magnetoresistance in magnetic fields up to 45 T. Data obtained down to 0.5 K, and extrapolated to $T = 0$, show a suppression of the hidden-order phase at $H_0(0) = 35.9 \pm 0.35$ T and the appearance of a new phase for magnetic fields in excess of $H_1(0) = 36.1 \pm 0.35$ T observed *only* at temperatures lower than 6 K. In turn, complete suppression of this high field state is attained at a critical magnetic field $H_2(0) = 39.7 \pm 0.35$ T. No phase transitions are observed above 40 T. We discuss our results in the context of itinerant versus localized f electrons.

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During the past few years there has been a true renaissance of interest in the unusual phase transition [1] that occurs in the superconducting heavy fermion system URu_2Si_2 at $T_0 \approx 17$ K, where all of the thermodynamic and transport properties exhibit a mean-field-like anomaly [2–4]. These early experimental results led to the conclusion that a magnetic phase transition, possibly of a spin density wave type, took place. However, when probed with microscopic measurements, e.g., neutron diffraction and muon spin rotation (μSR), only a very tiny magnetic moment of $\approx 0.02\mu_B/\mathrm{U}$ was found. Such a small moment could *not* account for the large changes in behavior at the phase transition, and it gradually became apparent that an unconventional type of magnetic order is at play. Indeed, recent neutron diffraction [5], nuclear magnetic resonance [6], and μSR [7] under pressure show the apparent tiny homogeneous moment to be due to a metallurgical minority phase of large moments ($\approx 0.3\mu_B$), which coexists with a majority (bulk) phase that has no magnetic moments. After more than 15 years of investigation, the nature of the bulk phase transition remains unidentified, and the term *hidden order* (HO) has recently been coined to describe this phenomenon [8]. Besides pressure, yet another external parameter is known to affect the ordered state, i.e., external magnetic fields. Pulsed-field measurements of magnetization [9–11], resistivity [12,13], Hall effect [13,14], and ultrasonic velocity changes (elastic moduli, c_{ij}) [15] exhibit a three-step transition between 35 and 40 T for which a satisfactory explanation is still pending.

In this Letter we present the first measurements of the temperature-field dependences of the specific heat, magnetocaloric effect, and resistivity from 0.5 to 20 K with fields up to 45 T to address the questions discussed above. The measurements indicate four regimes of anomalous behavior as URu_2Si_2 emerges from its hidden ordered state: (I) The continuous phase transition becomes sharper and symmetric in temperature as magnetic field is increased above 32 T. (II) There is no phase transition to be seen down to 0.5 K at ~36 T. (III) Between 36 and 39 T a first-order-like transition reappears. (IV) Above 40 T a Schottky-like maximum develops without any sign of a phase transition. These characteristics, never observed before, can then establish the basic ingredients of the HO state and form a critical test for the correct theoretical description. We consider two different scenarios to explain these behaviors: (A) Zeeman splitting of itinerant f-electron bands suppress the HO phase in region (I), which reappears as a single-spin ordered phase or an *orbital-flop* phase (the orbital current equivalent to an antiferromagnetic spin-flop phase) in region (III). This scenario is related to the exotic density wave mechanism recently proposed by Chandra *et al.* [16] although these authors have not yet considered possible high magnetic field transitions. (B) Crossing of f-electron crystal electric field (CEF) levels induces a quadrupolar ordered phase at high fields, region (III). Here we have the localized model of Santini [17] which ignores correlation effects between f electrons. Our data also suggest that quantum bicriticality may lie in the middle of the field region (II), i.e., where $T_0(H \simeq 36\ \mathrm{T}) = 0$.

Two different samples were measured: sample 1 was used for specific heat versus T and magnetoresistance measurements, and sample 2 for magnetocaloric effects. Single crystals of sample 1 were fabricated by triarc melting (Czochralski method) stoichiometric amounts of U, Ru, and Si. After the growth process the compound was annealed at 950 °C for one week. The crystal was characterized by Debye-Scherrer and Laue x-ray diffraction, and electron probe microanalysis. These results showed the crystal to be of excellent quality: on stoichiometry and no second structural phases. Measurements of the specific heat and magnetoresistance, which show HO transition at $T_0 = 17.1$ K, were carried out on oriented platelike and barlike samples, formed by spark erosion so that the external field is always parallel to the tetragonal c axis. Single crystals of sample 2, $T_0 = 17.4$ K, were grown by arc melting followed by vertical float-zone refining as described elsewhere [18].

The specific heat of sample 1 (see Fig. 1) was measured on a bar-shaped 9 mg piece with the c axis along the bar principal axis. We used a standard thermal relaxation method, with both *small* and *large* ΔT [19], to determine the specific heat as a function of the temperature at constant magnetic fields up to 45 T. The temperature was measured with a Cernox bare chip resistance thermometer (Lakeshore, Inc.) calibrated as described before [20]. Measurements were performed at the National High Magnetic Field Laboratory (NHMFL), Tallahassee, in both a water cooled resistive magnet operating to 32 T, and a hybrid magnet operating to a total field of 45 T. From the total specific heat measured (C_{tot}) we subtracted the contribution from phonons (C_{ph}) measured in a sample of $\mathrm{ThRu_2Si_2}$ [21]. Figure 1(a) displays $C_m/T = (C_{\mathrm{tot}} - C_{\mathrm{ph}})/T$ vs temperature for magnetic fields up to 33.5 T. Our data at low fields are in excellent agreement with previous measurements [22]. We observe that the anomaly associated with the HO phase in $\mathrm{URu_2Si_2}$ is shifted to lower temperatures by the external magnetic field, becoming sharper and more symmetric, without changing the amount of entropy recovered at the transition, which remains close to $0.15R$ (where R is the Rydberg gas constant). The sharpening of the anomaly indicates a gradual switch from continuous (second order) to discontinuous (first order) in temperature; however, we do not observe the hysteresis expected for such a transition. Figure 1(b) displays C_m/T measured at 36 and 38 T. We see here the complete suppression of the peak in C_m/T heat associated with the HO phase. Indeed, the data at $H = 36$ T show only a small step feature resembling that of $\mathrm{CeRu_2Si_2}$ near the metamagnetic transition at $H_m = 7.7$ T [23]. At $H = 38$ T, yet another large anomaly develops in C_m/T. This anomaly is suppressed with a magnetic field of 40 T, and its origin is unknown at the present time. The inset of Fig. 1(b) shows the C_m/T measured at $H = 40$, 42, and 45 T. In this regime all that is left in C_m is a Schottky-like anomaly that shifts from $T_{\max} \sim 5.6$ to 7.4 K ($\Delta T_{\max} \simeq 2$ K, $\sim$35%) when the magnetic field is increased by only 2 T (5%). We fitted our data with an expression for a Schottky anomaly using the following parameters: $\Delta_{40\ \mathrm{T}} = 1.55$ meV, $\Delta_{42\ \mathrm{T}} = 2.03$ meV, $\Delta_{45\ \mathrm{T}} = 2.48$ meV, and degeneracy equal to 0.6, giving an associated entropy $\sim(0.3 \pm 0.02) \times R$. Both $T_{\max}$ and Δ point to possible singlet f-electron CEF levels that cross at $H \simeq 36$–38 T. Such a level crossing was proposed as a semiquantitative explanation for the observed phenomenology of $\mathrm{URu_2Si_2}$ at high fields [17]. The upturn in C_m/T vs T at $H \geq 42$ T could be due to a phonon component that differs from that of $\mathrm{ThRu_2Si_2}$.

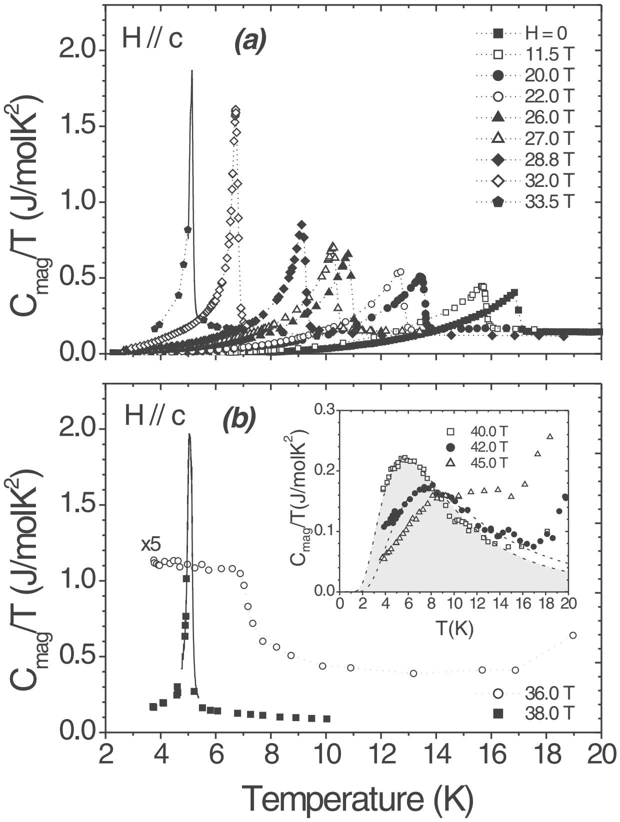

FIG. 1. (a) C_m/T vs T for magnetic fields up to 33.5 T in sample 1. The solid line indicates large delta T method. Dotted lines are guides to the eye. (b) C_m/T vs T for $H = 36$ T, with no sharp anomaly present, and $H = 38$ T where a new anomaly is evident. Inset: C_m/T vs T for $H = 40$, 42, 45 T. Dashed lines indicate fits with the Schotkky expression.

In order to follow the anomalies observed in these experiments down to mK temperatures, we polished a bar of sample 1 to dimensions $0.15 \times 0.4 \times 3\ \mathrm{mm}^3$ with its longer side along the crystallographic c axis and attached four gold leads using a spot welder, for magnetoresistance in pulsed fields. The electrical contact resistance when prepared in this way resulted in $\simeq 0.1\ \Omega$ each. We then mounted the sample on our Si/sapphire sample holder parallel to the direction of the applied magnetic field, such as to have $H \parallel c \parallel i$, where i is the applied electrical current. The small mass and large area of the sample helps keep the temperature constant during pulses. For these measurements we used a capacitor driven pulsed magnet able to produce a 400 ms pulse, and magnetic fields up to 50 T, at the NHMFL, Los Alamos. The sample resistivity (ρ) was measured using a standard lock-in amplifier detection technique operating at 173.2 kHz, and an excitation current of not more than 4 mA. Figure 2 displays ρ vs H at constant temperature for temperatures ranging from 0.5 to 20 K (bottom to top). Curves were displaced for clarity. Only a broad maximum around 40 T is observed above the HO phase transition $T_0 \simeq 17$ K, possibly related to the onset of metamagnetism [10], but below T_0 a clear minimum appears in ρ vs H. The minimum shifts monotonically

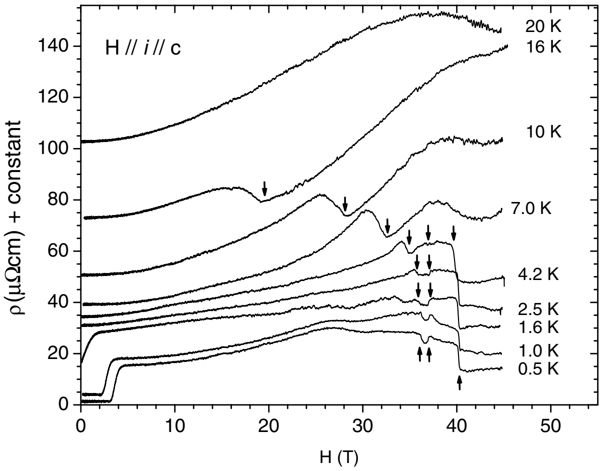

FIG. 2. Magnetoresistivity of sample 1 at constant temperature. All curves, except for $T = 0.5$ K, were displaced for clarity purposes. Anomalies associated with phase boundaries are indicated by arrows.

to higher fields as the temperature is reduced. We also observe a broad bump which narrows at higher fields, until at 4.2 K the resistance abruptly changes shape. Here and below we start seeing three anomalies, first a drop, then an increase, and finally a large drop in the sample resistance. Our higher temperature data agree partially with previous results [12]. Note that while the magnitude of the resistivity obtained with the ac technique used in this study may be slightly affected by capacitive/inductive effects, the magnetic fields at which anomalies are observed are not.

We have compiled all our data in Fig. 3. In Fig. 3(a) we have a phase diagram for sample 1 where we plotted the temperature and magnetic fields at which we observe anomalies in C_m/T vs T extracted from Fig. 1 (solid symbols) and anomalies in ρ vs H curves extracted from Fig. 2 (open symbols). We establish in this figure the new high field phase in URu_2Si_2 [region (III)], and note that the corresponding critical fields extrapolated to zero temperature are $H_0(0) = (35.9 \pm 0.35)$ T for the transition between regions (I) and (II), $H_1(0) = (36.1 \pm 0.35)$ T, for the transition between regions (II) and (III), and $H_2(0) = (39.7 \pm 0.35)$ T for the transition between regions (III) and (IV). Within experimental error we find $H_0(0) = H_1(0)$, a fact that could be coincidental or, more interestingly, could indicate that regions (I) and (III) are closely related. Our phase diagram resembles one proposed before [11], derived from susceptibility measurements at $1.3 \text{ K} \leq T \leq 4.2$ K.

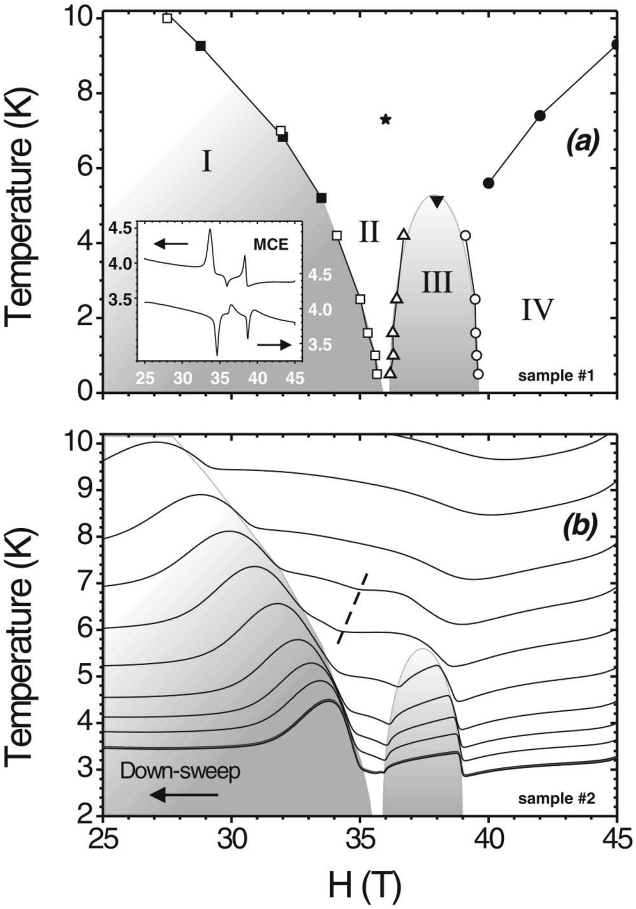

FIG. 3. (a) Phase diagram for sample 1 . (■), specific heat maximum in the low fields regime; (★), steplike transition; (▼), intermediate field peak; (•), position of Schottky anomaly at higher fields. (□, △, ○), anomalies in the resistance vs H. Inset: magnetocaloric effects sweeps. (b) Magnetocaloric effect in sample 2. Darker shade indicates where transitions are sharper.

In addition to the specific heat data, we measured the temperature changes in URu_2Si_2 due to the magnetocaloric effect (MCE) during magnetic field sweeps across the metamagnetic transition. The inset of Fig. 3(a) shows MCE data taken at 3.5 K sweeping the magnetic field from 25 to 45 T, and then back to 25 T at a constant rate of $\simeq 12$ T/min. Here we observe three *reversible* features; i.e., they change sign with the field ramp. When the magnetic field is increasing we see a temperature drop at $H_0 = 34.5$ T, then a peak at $H_1 = 36.5$ T, and another drop at $H_2 = 39$ T. We observe peaks, instead of steps, because of the calorimeter's finite relaxation time constant τ_{cal}. Since the total entropy of sample and stage should be conserved within times $t < \tau_{\text{cal}}$, a temperature drop at H_0 implies an increase of magnetic entropy (S_m) in the sample. The peak in the temperature vs field at H_1 indicates a drop in S_m, and the drop at H_2 indicates an increase in S_m. Our results suggest that the suppression of the hidden-order phase in URu_2Si_2 is accompanied by dramatic electronic band structure effects, in which the density of available quantum states increases causing the greater entropy of the system. Furthermore, the two features observed at higher fields strongly indicate that we cross through region (III), described above. Figure 3(b) exhibits the temperature changes observed in sample 2, when during the down-sweep various initial temperatures were used. For this sample we see the same features observed in sample 1 at slightly different fields, confirming that all results previously discussed are sample independent. We notice kinks in the temperature vs field that indicate the high field and low field phases converge to the same critical field (~ 36 T) at zero temperature. We also see a small anomaly (connected by dashed line) where we see the steplike feature in the C_m/T vs T of sample 1 displayed in Fig. 1(b). We note that while the jump in temperature at $H_2 \sim 39$ T is sharp, suggesting a first-order-like transition in field, the anomalies at $H_0 \sim 35.5$ T and $H_1 \sim 36$ T are more rounded, i.e., second-order-like transition in field. A plot of temperature traces during the field up-sweep has similar characteristics.

To explain the observed properties we analyzed two different cases: (A) The normal state of URu_2Si_2 is the coherent heavy fermion state in which f electrons acquire itinerant character upon hybridization with conduction electrons. In this itinerant band scenario the HO phase in region (I) is destroyed by a magnetic field due to Zeeman splitting of spin-down and spin-up bands, and a new single-spin phase is stabilized in region (III). Ordered states that may be affected in this way are those that involve singlet pairing at a characteristic translational (*nesting*) wave vector **Q**, such as a charge density wave, or the recently proposed incommensurate orbital antiferromagnetic phase [16]. Region (III) may then be the reentrance of the HO phase with a different nesting wave vector, or an orbital flopped phase. The Fermi surface instabilities produced by the Zeeman effect may also explain the steps observed in the magnetization vs field [9] as new phases are stabilized. (B) Localized f electrons dominate the low temperature behavior of URu_2Si_2 and the phase transitions near 40 T are a consequence of crossing singlet f-electron CEF levels, as proposed by Santini. Using a reduced quadrupolar coupling parameter λ [17], it can be shown [24] that region (III) may be a magnetic field induced antiferromagnetic quadrupolar phase. We observe two problems with this scenario. First, region (I) remains unexplained. Second, a Schottky contribution to the specific heat should be observed on both sides of the level crossing field, which is not supported by our experiments. A bicritical point in URu_2Si_2, a temperature below which a coexistence line separates regions (I) and (III) in the phase diagram, may exist below 0.5 K, and thus quantum fluctuations (a quantum critical point) may control the macroscopic thermodynamic and transport properties. Further experiments near $H_0 = 36$ T are under way.

In summary, we determined systematically the low temperature/high magnetic field phase diagram of URu_2Si_2 with measurements of specific heat versus temperature in continuous magnetic fields up to 45 T, magnetocaloric effect measurements, and magnetoresistance measurements in pulsed magnetic fields at temperatures from 0.5 to 20 K for the first time. The specific heat anomaly observed at the onset of the HO phase $T_0 \simeq 17$ K is completely suppressed in a magnetic field of 36 T, and a new phase is revealed between 36 and 40 T in which C_m/T shows a sharp first-order-like anomaly at $T = 5.2$ K. At 40–45 T no magnetic phase transition is observed, and C_m/T is dominated by a Schottky-like contribution. We also show that the magnetocaloric effect can be used to study the high field phases in detail.

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Note added.—After completion of this work we became aware of a related measurement that corroborates our phase diagram [25].

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