Field-Induced Fermi Surface Reconstruction and Adiabatic Continuity between Antiferromagnetism and the Hidden-Order State in URu_2Si_2

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Shubnikov–de Haas oscillations reveal at high fields an abrupt reconstruction of the Fermi surface within the hidden-order (HO) phase of URu_2Si_2. Taken together with reported Hall effect results, this implies an increase in the effective carrier density and suggests that the field suppression of the HO state is ultimately related to destabilizing a gap in the spectrum of itinerant quasiparticles. While hydrostatic pressure favors antiferromagnetism in detriment to the HO state, it has a modest effect on the complex $H - T$ phase diagram. Instead of phase separation between HO and antiferromagnetism our observations indicate adiabatic continuity between both orderings with field and pressure changing their relative weight.

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The nature of the phase emerging at $T_0 \simeq 17.5$ K which coexists with antiferromagnetism (AFM) in URu_2Si_2 remains elusive and has been called the "hidden-order" (HO) state. None of the theoretical scenarios proposed [1–3] can explain satisfactorily all the available data. In this Letter we present high-field, low-temperature electrical transport measurements in URu_2Si_2 under high hydrostatic pressure showing that AFM emerges adiabatically from the HO state.

The second-order phase transition at T_0 is associated with a large reduction in carrier density, due to a gap opening on the Fermi surface. The semimetallic nature of the HO phase is evident from the thermal conductivity [4], the Hall coefficient [5,6], and the de Haas–van Alphen (dHvA) effect [7]. A small staggered moment $m \simeq 0.03\mu_B$ per U ion is observed by neutron scattering [8]. Pressure dependent neutron diffraction reveals that the staggered moment sharply increases at 7 kbar to $0.41\mu_B$/U, which is the saturation value in the AFM phase [9]. However, both muon spin relaxation (μSR) experiments [10,11] and ^{29}Si NMR under pressure [12] find evidence for spatially inhomogeneous AFM order below T_0. Under pressure, the AFM volume fraction increases at the *expense* of the HO phase. The phase separation scenario is supported by μSR experiments performed at high pressures (the AFM volume fraction increases up to 90% under 8 kbar [11]). But this scenario is not consistent with recent dHvA measurements [13], which exhibit a continuous evolution as a function of pressure. Similarly, recent thermocaloric and thermoelectric data [14] favor the intrinsic coexistence of two order parameters.

Finally, heat capacity [15], magnetization [16], and electrical transport [17] measurements at very high magnetic fields established a rather complex and rich phase diagram in the vicinity of a field-induced quantum-critical point (QCP), suggesting competing order parameters. Rh doping greatly simplifies the phase diagram around the QCP suggesting that the interplay between HO and quantum criticality is a key aspect in promoting new phases. In contrast, here we show that the pressure-induced stabilization of AFM in "detriment" of the HO state has only a modest effect on the phase diagram surrounding the QCP.

The URu_2Si_2 single crystal used in this study has dimensions of $2.2 \times 1.48 \times 0.56$ mm^3 and was grown by the Czochralski method. It displays an in-plane residual resistivity $\rho_0 \simeq 11.2$ $\mu\Omega$ cm. Resistivity measurements were performed by using standard four-point techniques with the electrical current injected along the ab plane. A piece of dimensions $0.75 \times 0.3 \times 0.025$ mm^3 cleaved from the same crystal was used for hydrostatic pressure (produced by a CuBe clamp) studies. Measurements were performed at the National High Magnetic Field Laboratory.

Figure 1 shows the resistivity ρ of a URu_2Si_2 single crystal as a function of the external magnetic field H at $T = 50$ mK for several angles θ between $\mathbf{H}$ and the tetragonal c axis of the crystal. The increments in angle $\Delta\theta$ between traces is $\sim 8°$. The suppression of the HO state is observed at the critical field H_A, while H_B indicates the transition into a new field-induced phase (phase III following the notation of Ref. [17]). Phase III is suppressed by the field at H_C. All critical fields follow a $1/\cos(\theta)$ dependence, indicating that these transitions are induced by the component of H along the c axis. The oscillatory component of the resistivity is due to the Shubnikov–de Haas (SdH) effect. Red lines and horizontal arrows at both sides of the resistivity maximum for the $\rho(H, \theta = 0°)$ trace

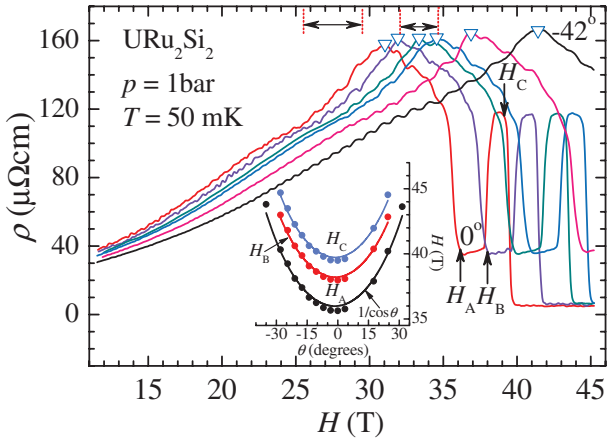

FIG. 1 (color). Resistivity ρ as a function of the field H of a URu_2Si_2 single crystal at $T \simeq 50$ mK and for several angles θ between H and the interplane c axis. Red lines and horizontal arrows indicate the two field ranges, at each side of the resistivity maximum (indicated by blue triangles) in the $\theta = 0°$ trace, where the subsequent Fourier analysis is performed. Three sharp phase transitions at magnetic fields H_A, H_B, and H_C are indicated by arrows. Inset: angular dependence of H_A, H_B, and H_C, where solid lines are fits to $1/\cos(\theta)$.

indicate the two ranges in H where we performed the subsequent Fourier analysis of the oscillatory component.

Figures 2(a) and 2(b) display the fast Fourier transform (FFT) of the SdH signal for several values of T. The range of fields to each side of the maximum in $\rho(H, \theta = 0°)$ at

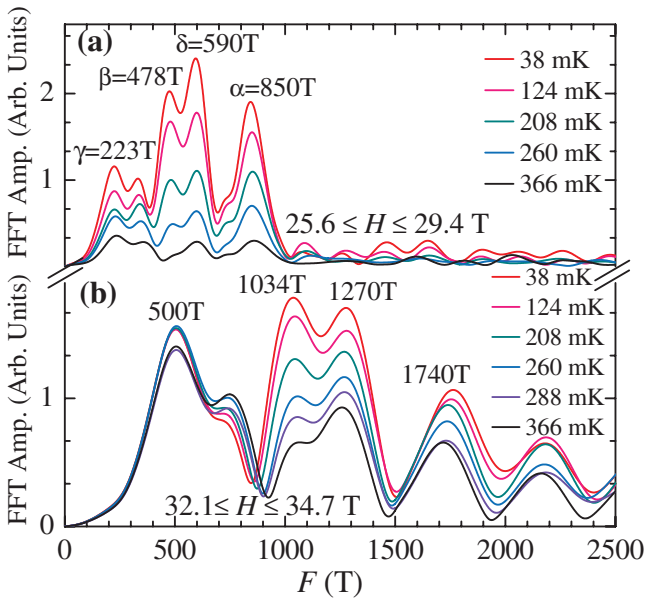

FIG. 2 (color). (a) Fast Fourier transform of the oscillatory part of the resistivity, i.e., $\tilde{\sigma} = (\rho^{-1} - \rho_b^{-1})\rho_b$, where ρ_b is the background resistivity obtained from a polynomial fit for $25.6 \leq H \leq 29.4$ T, $\theta = 0°$, and for several temperatures. (b) The same as in (a) but for $32.1 \leq H \leq 34.7$ T. Notice the frequency shift in the spectral weight.

$H_{\max} \simeq 31$ T is shown by the red lines and horizontal arrows in Fig. 1 and the definition of the SdH signal is given in the caption of Fig. 2. Previous dHvA effect studies in URu_2Si_2 in fields up to 15 T [7] resolved three main frequencies: $F_\alpha = 1.05$, $F_\beta = 0.42$, and $F_\gamma = 0.19$ kT, respectively. In this study, we found field-dependent SdH frequencies: while F_α decreases, $F_{\beta,\gamma}$ increase slightly with H. For the limited field range $25.6 < H < 29.4$ T, we obtain $F_\alpha = 0.85$, $F_\beta = 0.48$, and $F_\gamma = 0.22$ kT, with effective masses $\mu_\alpha = 17 \pm 1$, $\mu_\beta = 19 \pm 1$, and $\mu_\gamma = 14.2 \pm 0.4$, respectively, with all masses expressed in units of the free electron mass m_0. The quasiparticle effective masses are slightly field dependent. We also find an extra peak at $F_\delta = 0.59$ kT, not previously reported. Following Ref. [7], which finds that F_α and F_β occupy, respectively, $\simeq 0.7\%$ and 0.12% of the volume of the first Brillouin zone (V_{FBZ}) and assuming that F_γ, F_δ as well as the peak at $\simeq 0.33$ kT occupy a total volume roughly similar to that of F_α, one obtains a total Fermi surface (FS) volume of $\sim 1.52\%$ of V_{FBZ} equivalent to ~ 0.03 carriers per unit cell volume. This value is remarkably close to those extracted from Hall effect measurements, i.e., between 0.03 and 0.05 holes per U [6,18], suggesting that we detected most of the FS sheets of URu_2Si_2. Although as noted in [7], their combined effective masses would not account for the Sommerfeld coefficient in the heat capacity [$\gamma \sim 75$ mJ/(mol K^2) in our crystal]. However, there is the more remote possibility of large electron and hole Fermi surfaces compensating each other in the Hall effect and without closed orbits or very heavy masses so that they remain undetected in the SdH signal. This is difficult to reconcile with a factor of 5 increase in the Hall coefficient, the sharp decrease in the electronic scattering rate, and the increase in the entropy per carrier combined with the drastic reduction in entropy per volume at the HO transition [6].

As seen in Fig. 2(b), all frequencies increase by a factor >2 in the field range $32.1 < H < 34.7$ T, i.e., above the maximum in $\rho(H)$, indicating a remarkable increase in the FS cross-sectional areas. The effective masses also decrease considerably above the maximum. Peaks in the FFT are much broader due to (i) the limited range in H^{-1} and (ii) the fact that the geometry of the FS is changing as the suppression of the HO state at ~ 36 T is approached. The sudden increase in the FS volume is correlated with the sudden decrease in the Hall constant at $H_{\max}$ [5,6]. Both effects, taken together, are the signature of an abrupt increase in the number of itinerant carriers. Since F_α increases approximately by a factor 2.1, so that, assuming parabolic bands, the increase in carriers is about $2.1^{3/2} \sim 3$, in agreement with Hall effect data [5,6], showing a decrease of R_H by a factor of 3. This indicates the reconstruction of the FS, e.g., the field-induced partial suppression of a gap within the spectrum of the itinerant quasiparticles. However, neither a step in the magnetiza-

tion [16] nor an anomaly in the magnetocaloric effect [15] has been observed at H_{max} at higher temperatures. The abrupt increase in volume of *all* FS sheets *within* the HO state implies that its field-induced suppression cannot result from the spin polarization of the β and γ FS sheets since the actual Pauli critical fields significantly exceed the values claimed in Ref. [18].

All features seen in our isothermal field scans of the resistivity at ambient pressure (not shown here), albeit sharper, are in excellent agreement with those reported in Ref. [17]. Although the application of hydrostatic pressures in the order of $p \geq 10$ kbar has been claimed to stabilize AFM as the majority phase [9,11] in URu_2Si_2, the overall behavior of ρ as a function of field and temperature remains very similar to the one observed at $p =$ 1 bar. The main differences are: (i) p induces the suppression of the highly hysteretic region associated with phase V [17] and (ii) p moves the suppression of the HO state and the subsequent high-field phases to even higher fields but lower temperatures.

Strong similarities between features observed for the different data sets, i.e., data within the HO state and within the mainly AFM state, are also observed in the T dependence of the resistivity under different external fields as shown in Figs. 3(a) and 3(b) for $p = 1$ bar and 11 ± 1 kbar, respectively. The size and shape of the resistive feature at the HO transition (black markers) is very similar between both data sets, although naively one would expect it to become less pronounced at 11 kbar when most carriers compose the AFM phase. Under pressure the features associated with the phases close to the quantum-critical point appear at higher magnetic fields.

Using the isothermal field scans as well as the temperature dependency of the resistivity under different values of the external field, we constructed in Fig. 4 the $T - H$ phase diagram of URu_2Si_2 for a limited field range but for three values of hydrostatic pressure: $p = 1$ bar [Fig. 4(a)], $p =$ (8 ± 1) kbar [Fig. 4(b)], and $p = (11 \pm 1)$ kbar [Fig. 4(c)].

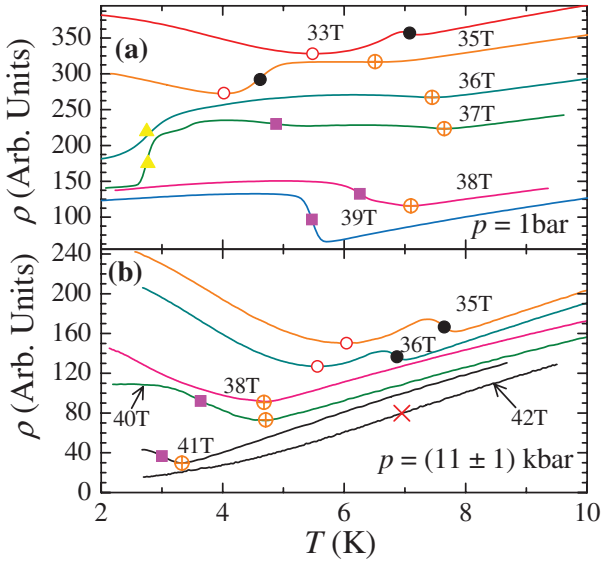

FIG. 3 (color). (a) ρ as a function of T for several values of the external field. Black solid dots indicate the phase boundary between the metallic and the HO state, while yellow solid triangles and magenta solid squares indicate, respectively, the transition towards and the field-induced suppression of phase III (following the nomenclature of Ref. [17]). Red dots indicate the position in T of a minimum observed in ρ within the HO state, while orange dots indicate the phase transition towards the field-induced phase II. (b) Same as in (a) but for a pressure $p =$ 11 ± 1 kbar. For clarity all curves have been vertically displaced.

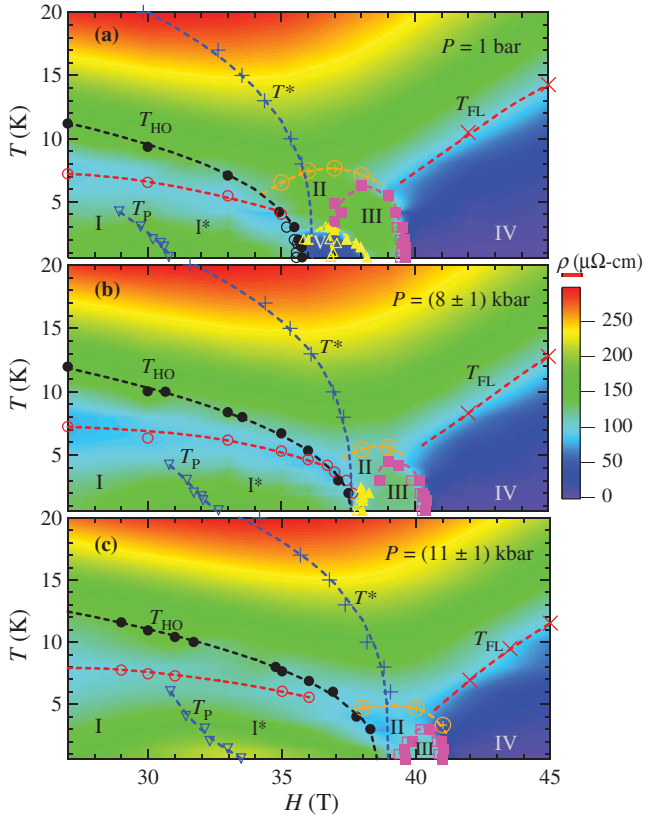

FIG. 4 (color). (a) The resulting $H - T$ phase diagram of URu_2Si_2 at $p = 1$ bar, shown for comparative purposes. The phase boundary towards the HO state (phase I, following the nomenclature of Ref. [17]) is indicated by black dots. The position of a minimum in ρ within the HO state is indicated by red circles. The position of the maximum observed in ρ within the HO state at T_p and where the geometry of the Fermi surface changes significantly is indicated by blue triangles. We nominate this new phase as phase $I^\star$. The boundary of the high-field phase III is defined by magenta squares, while the boundaries of phases II and V are defined, respectively, by orange circles and yellow triangles. The recovery of a FL state at T_{FL} is indicated by red crosses. Finally, the crossover from positive to negative magnetoresistance within the higher temperature metallic state is indicated by blue crosses. (b) Same as in (a) but under a pressure $p = 8 \pm 1$ kbar. (c) Same as in (a) but for a pressure $p = 11 \pm 1$ kbar.

Clearly, the development of AFM with pressure (AFM should occupy about 90 to 100% of the sample volume at 8 and 10 kbar [11]) does not alter significantly the thermodynamic phase boundaries between HO/AFM phases and the metallic state, nor between these and the field-induced phases. The boundary between metallic and HO/AFM states was fit (black dashed lines) to the expression $T = |H - H_c|^{\alpha}$ [17] yielding no clear dependence of α on pressure, i.e., $\alpha = 0.39$, 0.42, and 0.40, with $H_c = 35.8$, 37.7, and 38.6 T, for $p = 1$ bar, (8 ± 1), and (11 ± 1) kbar, respectively. The ''mixed'' state composed of AFM and HO is stabilized by pressure and with increasing p it occupies a larger area of the $H - T$ phase space in detriment of the field-induced phases, suggesting adiabatic continuity between AFM and HO, i.e., that they conform a single thermodynamic phase.

There are two scenarios for the semimetallic mixed HO and AFM phase of URu_2Si_2: (i) μSR [10,11] and ^{29}Si NMR under pressure [12] suggest phase separation, i.e., that HO and AFM order do not coexist in the same regions of space, while (ii) dHvA [13] and the present measurements favor coexistence in a single phase. Scenario (i) suggests that a percolation transition of AFM domains should occur as a function of pressure (in analogy to manganites, e.g., [19]) which so far has not been observed. Although ^{29}Si NMR shows the emergence of AFM sites under pressure, it reveals no evidence for domain walls between AFM and HO droplets, a necessary ingredient for phase separation. In scenario (ii) the evolution from HO to AFM under pressure is a crossover rather than a phase transition with two competing coupled order parameters in the mixed phase, as considered in Refs. [2,3]. At ambient pressure the HO order parameter dominates and gradually with increasing pressure the AFM order parameter takes over. Polarized neutron scattering under fields up to $H = 17$ T [20] also detects two gaps in the magnetic excitations, one at the AFM zone center which increases strongly with H (as it does under pressure) and a second one that is nearly field independent. Field-induced suppression of AFM would explain the FS reconstruction observed by us.

Our observations favor scenario (ii), i.e., adiabatic continuity between HO and AFM with gradual change in their weight with field and pressure. The picture of the HO state emerging from this study indicates a semimetallic state containing a dipolar component which has FS pockets enclosing a volume of $\sim$1.5% of the Brillouin zone. It becomes slightly more metallic (in agreement with the Hall effect) under field, as indicated from the increased FS cross sections in the SdH oscillations at T_p, then into a sequence of field-induced phases (which are shifted to higher fields and lower temperatures under pressure), and finally into the metallic FL phase IV.

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