Competing Ordered Phases in URu_2Si_2: Hydrostatic Pressure and Rhenium Substitution

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A persistent kink in the pressure dependence of the "hidden order" (HO) transition temperature of $URu_{2-x}Re_xSi_2$ is observed at a critical pressure $P_c = 15$ kbar for $0 \le x \le 0.08$. In URu_2Si_2, the kink at P_c is accompanied by the destruction of superconductivity, a change in the magnitude of a spin excitation gap, determined from electrical resistivity measurements; and a complete gapping of a portion of the Fermi surface (FS), inferred from a change in scattering and the competition between the HO state and superconductivity for FS fraction.

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Since its discovery over 20 years ago [1,2], the moderately heavy fermion compound URu_2Si_2 has been the focus of many theoretical and experimental efforts designed to determine the elusive, hidden order parameter associated with the phase transition occurring at $T_0 \approx 17.5$ K. The transition into this "hidden order" (HO) state is characterized by large anomalies (typical of magnetic ordering) in specific heat, electrical resistivity, thermal conductivity, and magnetization measurements [1–6]; however, only a small antiferromagnetic moment, insufficient to adequately explain the entropy released during the transition, was detected in low-temperature neutron diffraction experiments [7]. In addition to the puzzling order parameter of the HO state, URu_2Si_2 undergoes a transition into an unconventional superconducting (SC) state, which coexists with weak antiferromagnetism (AFM), at $T_c \approx 1.5$ K. The potential interplay between the two ordered phases of URu_2Si_2 as well as the nature of the HO state are underlying problems to our fundamental understanding of the properties of this compound.

In an effort to explain the observed properties of URu_2Si_2, several microscopic models have been proposed [8–14]. In addition to the theoretical pursuits, many varied experimental techniques have been employed to confirm and/or constrain the proposed models; however, the experimental results fail to converge upon an encompassing microscopic description of the ordered states of URu_2Si_2, but do provide valuable insight when contextually analyzed. Low-temperature neutron diffraction measurements provide evidence that the order parameter of the HO state must break time-reversal symmetry [15], and recent inelastic neutron scattering measurements reveal gapped, incommensurate spin excitations, possibly accounting for the large specific heat [16]. Thermal transport measurements are consistent with the opening of a gap at the Fermi surface (FS), as previously suggested by optical conductivity and specific heat studies [2,17], depleting carriers and reducing electron-phonon scattering [4,5]. These measurements tend to converge upon a description of the HO state invoking the presence of a FS instability such as a spin density wave (SDW), further suggested by high-field measurements intimating the itinerant nature of the HO state [18].

The application of pressure to URu_2Si_2 further convolutes the discussion of the nature of the hidden order parameter as well as the persistence of the SC state. While the HO transition temperature T_0 was seen to increase with applied pressure [3], the SC state was found to be suppressed; however, the reported critical pressures for superconductivity vary greatly from 4–15 kbar, possibly due to disparities in sample quality or high-pressure conditions [3,19–21]. Pressure-dependent neutron scattering, NMR, and μSR measurements all indicate an abrupt increase in the size of the ordered moment, with the magnitude of the moment saturating to a value consistent with bulk AFM above a currently disputed critical pressure [22–25]. The interpretation of this increase in moment is currently unresolved, but can be divided into two prevailing descriptions: an inhomogeneous, phase separated scenario where the increase in the moment is due to an increase in volume fraction of the observed, high-pressure moment [24,25]; and a scenario involving two order parameters describing the staggered magnetization and the HO state, with the details governed by the coupling between order parameters [14,23].

With the progression of experimental and theoretical investigations into the ordered states of URu_2Si_2, now is a critical time to clarify and categorize experimental observations in order to enhance the understanding of the underlying phenomena. The $URu_{2-x}Re_xSi_2$ system provides a unique opportunity to study the effects of pressure on a HO state whose ambient pressure transition temperature is reduced with rhenium concentration x [26]. In this letter we report new, comprehensive high-pressure electrical resistivity measurements on single crystal samples of the $URu_{2-x}Re_xSi_2$ system, emphasizing with greater clarity than previous studies the pressure dependence of the HO state and its relation to the field and pressure dependence of the SC state. A change is observed in the gap derived from electrical resistivity along with a systematic evolution of the scattering processes in URu_2Si_2. In addition, using a framework developed by Bilbro and

McMillan [27], the competition for FS fraction between the HO and SC states is quantified, engendering a consistent depiction of the pressure dependence of the ordered states of URu_2Si_2.

Single crystals of $URu_{2-x}Re_xSi_2$ with $x = 0$, 0.01, 0.02, 0.04, 0.06, and 0.08 were grown using the Czochralski method and then annealed at 900 °C for 7 days. The Laue method was used to orient the single crystals, which were subsequently spark cut and polished into electrical resistivity specimens. Electrical resistivity measurements under pressure were performed with a beryllium-copper, piston-cylinder cell using a Teflon capsule filled with a 1:1 mixture of n-pentane:isoamyl alcohol as the pressure-transmitting medium to ensure hydrostatic conditions during pressurization at room temperature. The pressure in the sample chamber was calibrated from the inductively determined, pressure-dependent superconducting critical temperature of a lead manometer.

Displayed in Fig. 1(a) are representative electrical resistivity $\rho(T)$ data illustrating the evolution of T_0, defined as the local minima occurring between approximately 16.5 and 20 K, with applied pressure. The data show no qualitative change in appearance as the characteristic trough and peak structure of the HO transition is preserved to high pressures. The inset of Fig. 1(a) shows the electrical resistivity normalized at 2 K, $\rho(T)/\rho(2\ \mathrm{K})$, emphasizing the pressure dependence of the SC transition. The horizontal dashed lines represent—from top to bottom—90%, 50%, and 10% of the normal state value, with the SC critical temperature T_c defined as the temperature at which $\rho(T)/\rho(2\ \mathrm{K}) = 0.5$. As a consequence of the inherently broad SC transition in URu_2Si_2, incomplete transitions were observed to the highest measured pressures.

Figures 1(b) and 1(c) display $\rho(T)$ data near T_0 for specimens of $URu_{1.96}Re_{0.04}Si_2$ and $URu_{1.92}Re_{0.08}Si_2$, respectively. While the absolute value of T_0 and the relative value of the height of the transition in ρ are altered with x, no dramatic changes in the qualitative shape of the feature at T_0 were observed.

In addition to the pressure dependence of the SC state seen in the inset of Fig. 1(a), field-dependent measurements of the SC state were performed for $P < 9.2$ kbar with $H \parallel c$ (inset of Fig. 2). The determined critical field curves were fit by a semiempirical, parabolic expression to extract $H_{c2}(0)$, the zero-temperature upper critical field. Using these values of $H_{c2}(0)$, the scaled critical field curves $H_{c2}(T)/H_{c2}(0)$ were plotted as a function of reduced temperature T/T_c (Fig. 2). The data scale very close to 1 another using these simple criteria, indicating that the SC state evolves continuously up to the critical pressure, in contrast to other measurements displaying an abrupt destruction of superconductivity at lower pressures [19,20].

For all x measured, T_0 and T_c are plotted as a function of P in Fig. 3(a), where T_0 evinces a distinct kink in its pressure dependence at 15 kbar, regardless of the ambient pressure value of T_0. The presence of this kink, when taken

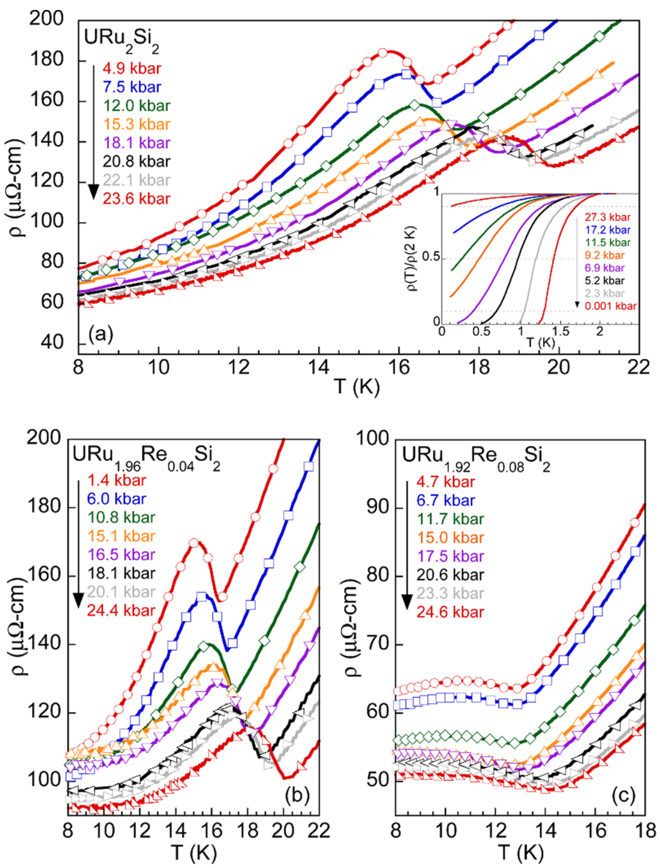

FIG. 1 (color online). (a) $\rho(T, P)$ of URu_2Si_2 as a function of T. Inset: normalized electrical resistivity, $\rho(T)/\rho(2\ \mathrm{K})$, vs T near T_c (dashed lines defined in the text). $\rho(T, P)$ data for specimens of $URu_{2-x}Re_xSi_2$ with $x = 0.04$ (b) and 0.08 (c).

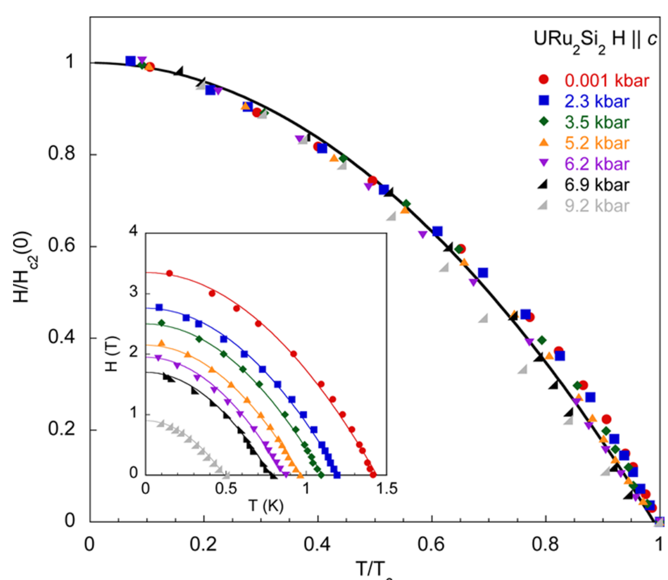

FIG. 2 (color online). Scaled critical field curves, $H_{c2}(T)/H_{c2}(0)$ vs T/T_c, of the SC state of URu_2Si_2 for $P \leq 9.2$ kbar and $H \parallel c$. The solid line is a guide to the eye. Inset: $H_{c2}(T)$ vs T of the SC state. The solid lines are fits to a parabolic expression yielding values of $H_{c2}(0)$.

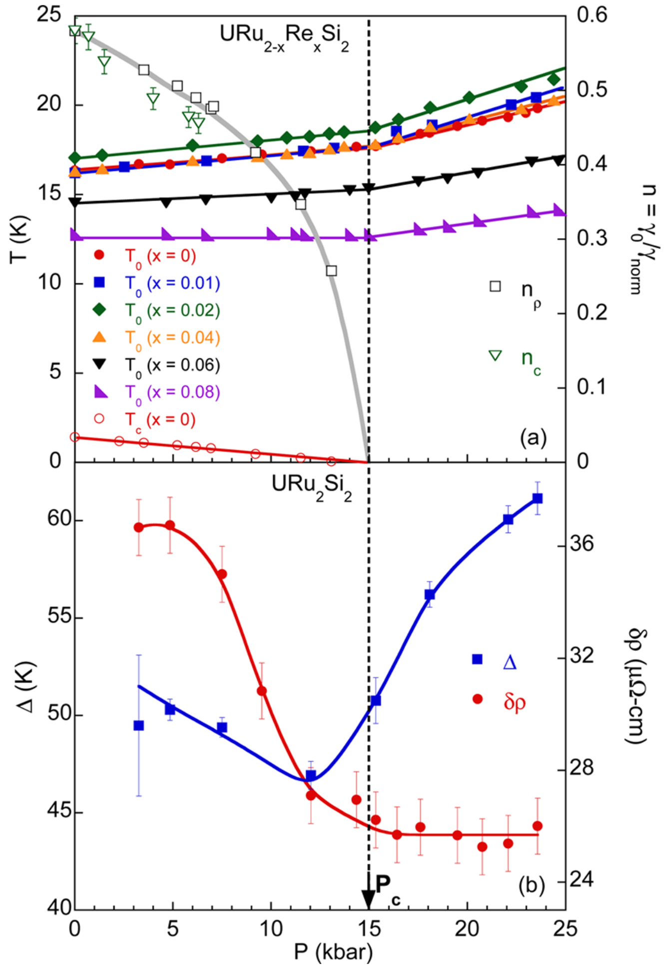

FIG. 3 (color online). (a) T-P phase diagram (left axis) of $URu_{2-x}Re_xSi_2$ showing the evolution of the HO and SC states. An x-independent kink in $T_0(P)$ is visible at $P_c = 15$ kbar. The fraction of the FS left ungapped by the HO transition, $n \equiv \gamma_0/\gamma_{\rm norm}$ (right axis), vs P as determined from T_0 and T_c, n_ρ (open black squares), and as estimated from previous $C(T, P)$ measurements [31], n_c (open, inverted green triangles). (b) Gap determined from resistivity Δ (left axis) from fits to $\rho(T)$ below T_0. $\delta\rho$ (right axis) as defined in the text. The lines are guides to the eye. The vertical dashed line marks the critical pressure $P_c = 15$ kbar.

within the context of the analysis of Mineev and Zhitomirsky [14], indicates a scenario where there exists no coupling between the order parameters of the HO state and that of the high-pressure AFM phase, and furthermore suggests that sample impurities or uncharacterized strains may be culpable for the observed moment at low pressures. The persistence of this kink and its static position upon reducing T_0 with increased x is consistent with a vertical or nearly vertical HO/AFM transition occurring at $P_c = 15$ kbar; as this transition is indirectly probed, there can be no unambiguous determination as to the degree of its order. This purported HO/AFM transition has been directly observed in other measurements [20,23,28]; however, the reported critical pressures are much lower than 15 kbar, typically near 7 kbar. This discrepancy in P_c could be due to sample dependence or potential nonhydrostatic conditions present in previous experiments utilizing a mixture of Fluorinert FC70/77, which remains liquid (hydrostatic) at room temperature only up to approximately 8 kbar [29]. In addition to the persistent kink at 15 kbar, T_c for URu_2Si_2 is suppressed very near P_c, consistent with previous results suggesting that superconductivity and AFM are mutually exclusive [21]. In fact, if nonhydrostatic conditions are responsible for the early onset of AFM at $P < 15$ kbar, then one would expect the SC state to be suppressed at these lower pressures as reported by several researchers [19,20].

It was suggested previously that the HO transition partially gaps a portion of the FS with the remaining ungapped portion undergoing superconductivity at low temperatures [2]. In this scenario, the existence of superconductivity is predicated upon the incomplete FS gap induced by the onset of HO, and, as such, the two ordered states compete for FS fraction. Bilbro and McMillan proposed a model to quantitatively analyze the effects of ordered phases competing for FS fraction [27]:

$$T_{c0} = T_c(P)^{n(P)} T_0(P)^{1-n(P)}, \qquad (1)$$

where T_{c0} is the value of the SC critical temperature in the absence of a high-temperature, FS-gapping transition and $n(P)$ is a measure of the residual ungapped FS—which can be determined from specific heat $C(T)$ measurements as $n \equiv \gamma_0/\gamma_{\rm norm}$, the ratio of the electronic specific heat coefficient above T_c to that above T_0. Using the previously determined value of $n(0) = 0.58$ [2], and evaluating Eq. (1) at ambient pressure results in a value of $T_{c0} = 3.9$ K, which can then be used along with the values of $T_0(P)$ and $T_c(P)$ to quantify the fraction of the FS that is gapped by the HO transition. The results of this analysis (labeled n_ρ) are contained in Fig. 3(a)–where the HO transition would appear, invoking a reasonable extrapolation, to completely gap its portion of the FS near P_c [30], thus suppressing superconductivity in the vicinity of P_c as seen. Also plotted in Fig. 3(a) are extracted results for $n(P)$ from $C(T)$ data under pressure (labeled n_c) [31], which are in excellent agreement with the analysis of the $\rho(T)$ data presented herein, although $C(T)$ data to higher pressures would be desirable to confirm this postulate.

The electrical resistivity of Fig. 1(a) was fit from approximately 2 K up to 90% of T_0 with the previously employed expression [3,32,33]:

$$\rho(T) = \rho_0 + AT^2 + B\frac{T}{\Delta}\left(1 + \frac{T}{\Delta}\right)e^{(-\Delta/T)}, \qquad (2)$$

which accounts for the residual resistivity ρ_0, a heavy Fermi liquid term, and scattering from gapped spin excitations. The magnitude of the gap Δ, which has no simple relation to the magnon gaps extracted from neutron diffraction experiments [15], was found to undergo a change in its pressure dependence near P_c, as shown in Fig. 3(b), where the error bars result from the fitting algorithm used. Within a SDW formalism, this gap Δ would be associated with the magnitude of the FS gap [34], and, although other conceivable pressure-dependent parameters are involved in the relationship, $\Delta(P)$ is an indirect probe of the pressure-dependent evolution of the FS. Also shown in Fig. 3(b) is the magnitude of the HO anomaly $\delta\rho$, defined by extrapolating the temperature dependences above and below the HO transition to T_0 and evaluating the difference in the resultant electrical resistivity; the ascribed error bars result from small differences in permissible extrapolations. The quantity $\delta\rho$ decreases with pressure up to P_c, after which it remains constant. This behavior can be understood within the context of the fraction of FS gapped by the HO transition: with applied pressure, the fraction of FS gapped by the HO transition grows larger and reduces the number of available states into which quasiparticles can scatter, resulting in a reduction in the scattering and a consequent reduction in $\delta\rho$; at and above P_c, the portion of the FS associated with HO is completely gapped and the scattering processes along with $\delta\rho$ become constant.

The salient features of the results presented herein can be summarized as follows: (i) a distinct kink in $T_0(P)$ is seen at $P_c = 15$ kbar and the kink is independent of the value of $T_0(0)$ as modified by increasing x; (ii) the HO state coincides with superconductivity, and the mechanism for superconductivity appears unaltered according to the variations of T_c and $H_{c2}(T)$ with P; (iii) the HO and SC states compete for FS fraction, with the former fully gapping a portion of the FS near 15 kbar; (iv) the gap Δ associated with magnetic excitations inferred from $\rho(T, P)$ measurements evinces a change in behavior near 15 kbar. It would appear likely that at the critical pressure of $P_c = 15$ kbar, URu_2Si_2 undergoes a distinct HO/AFM transition, although the degree of order and nature of the transition remain uncertain. Furthermore, this HO/AFM transition seemingly occurs along a vertical or near vertical phase boundary near 15 kbar. The coincidence of the HO/AFM boundary, the fully gapped portion of the FS where HO resides, and the change in Δ is not currently understood. No particular microscopic model has been invoked to discuss the results, although the simultaneous presence of a spin and FS gap strongly favor the formation of a SDW-like FS instability at T_0.

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