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The suppression of hidden order and the onset of ferromagnetism in URu₂Si₂ via Re substitution

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Abstract

Substitution of Re for Ru in the heavy fermion compound URu₂Si₂ suppresses the hidden order transition and gives rise to ferromagnetism at higher concentrations. The hidden order transition of URu_{2-x}Re_xSi₂, tracked via specific heat and electrical resistivity measurements, decreases in temperature and broadens, and is no longer observed for $x > 0.1$. A critical scaling analysis of the bulk magnetization indicates that the ferromagnetic ordering temperature and ordered moment are suppressed continuously towards zero at a critical concentration of $x \approx 0.15$, and this is accompanied by the additional suppression of the critical exponents γ and $\delta - 1$ towards zero. This unusual trend appears to reflect the underlying interplay between Kondo and ferromagnetic interactions, and perhaps the proximity of the hidden order phase.

(Some figures in this article are in colour only in the electronic version)

For well over two decades, the identity of the ordered phase found in URu₂Si₂ at temperatures below 17 K has eluded researchers. This hidden order (HO) phase coexists with a heavy fermion state, but yields to a superconducting ground state below 1.5 K. In order to develop a better understanding of the HO phase, URu₂Si₂ has been extensively studied by various techniques, and these efforts have uncovered fascinating behavior under magnetic field, applied pressure, and chemical substitution. We describe the novel phase changes induced by Re substitution for Ru, including the suppression of the HO transition, the nearby emergence of ferromagnetic (FM) order, and the unique critical behavior of this phase.

Chemical substitution studies have been used primarily to investigate the robustness of the HO phase, and substitutions have been made for all three elements in URu₂Si₂. Replacing U with Th or rare earths quickly suppresses the HO transition temperature [1, 2], emphasizing the critical role of the U f-electrons to the HO phase. In contrast, substitution for Si has no dramatic effects [3]. Most of the neighboring elements of Ru: Mn, Tc, Th, Re, Os, Rh, and Ir have been substituted onto the Ru site, and in all cases except Os, the HO transition is suppressed by about 5% substituent concentration [4–6].

For a few elements, the phase diagrams at higher concentrations have been studied. In the Rh case, in

addition to the HO phase, there are two other nominally antiferromagnetic (AFM) phases, which are separated by nonmagnetic regions [7]. In contrast to the HO, these AFM phases are characterized by a larger moment [8, 9], and a complex magnetic structure exists at intermediate Rh concentrations [10]. A closer examination of the HO phase found that for almost half of the range of Rh concentration over which the HO phase exists, the order is actually AFM [11].

Substituting into URu₂Si₂ with Re, Tc, or Mn leads instead to ferromagnetic (FM) order [6], whose discovery constituted the first experimental realization of a heavy fermion FM instability. In URu_{2-x}Re_xSi₂, a maximum $T_C \approx 40$ K is found at $x = 0.8$, near the concentration limit below which the material remains single phase. The itinerant FM order is characterized by a small moment, with no anomalies in electrical resistivity or specific heat at T_C . Across the Re substitution series, the heavy fermion state is reflected in an enhanced electronic contribution to the specific heat [4] and a narrow Drude peak in optical conductivity at low temperatures [12]. The long-range nature of the FM phase has been confirmed by neutron scattering [13] and ²⁹Si NMR [14]. Within the FM phase, non-Fermi liquid (NFL) behavior is observed in the low-temperature bulk properties [15] and in energy-temperature scaling of the dynamic magnetic susceptibility [16]. The origin of the NFL

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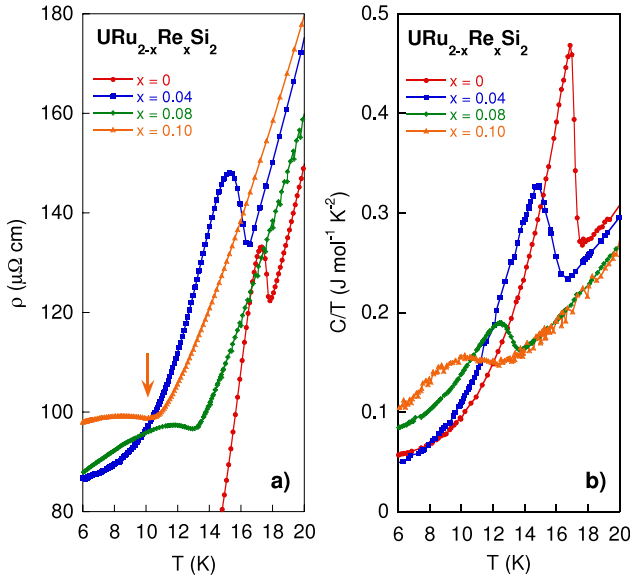


Figure 1. Electrical resistivity and specific heat anomalies associated with the hidden order transition in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$. (a) The transition temperature T_0 decreases with increasing x , as the height of the anomaly in $\rho(T)$ shrinks and the width broadens. The arrow points to the minimum in $\rho(T)$ for $x = 0.10$. (b) The jump in C/T at T_0 decreases in magnitude as T_0 decreases with increasing x .

effects remains an open question, partly because it has been difficult to conclusively define the FM phase boundary. As only polycrystalline samples have been studied so far, it is also necessary to exclude orientational averaging or grain boundary effects as causes for these unusual properties.

Single crystals of $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ were synthesized via the Czochralski technique in a tri-arc furnace, and subsequently annealed in Ar. Phase homogeneity was verified by powder x-ray diffraction. Samples were oriented via the back-reflection Laue method and spark cut and sanded to size. Electrical resistivity measurements as a function of temperature $\rho(T)$ were performed in a liquid ^4He cryostat. Measurements of magnetization in field $M(H)$ were made using a Quantum Design Magnetic Properties Measurement System (MPMS). Measurements of the specific heat C were performed in a ^3He refrigerator utilizing a semi-adiabatic heat pulse technique.

As seen in figure 1, in the vicinity of the HO transition, the $\rho(T)$ data exhibit a small peak, qualitatively similar to that seen in elemental Cr at the ordering transition into a spin density wave. The transition into the HO phase is marked by a large specific heat anomaly, which is related to the incomplete gapping of the Fermi surface in URu_2Si_2 [17–19]. The general shape of these anomalies persists despite a suppression of the ordering temperature as x increases, suggesting that the transition into the HO state always involves a Fermi surface instability. As Re concentration increases, the anomalies shrink in height and broaden in width, becoming more poorly defined near $x = 0.10$. For $x = 0.12$, no HO anomaly can be identified in $\rho(T)$ or $C(T)$.

Isotherms of $M(H)$ are shown in figure 2 for $\text{URu}_{1.70}\text{Re}_{0.30}\text{Si}_2$, which orders ferromagnetically below approximately 5 K. As the Re concentration increases from

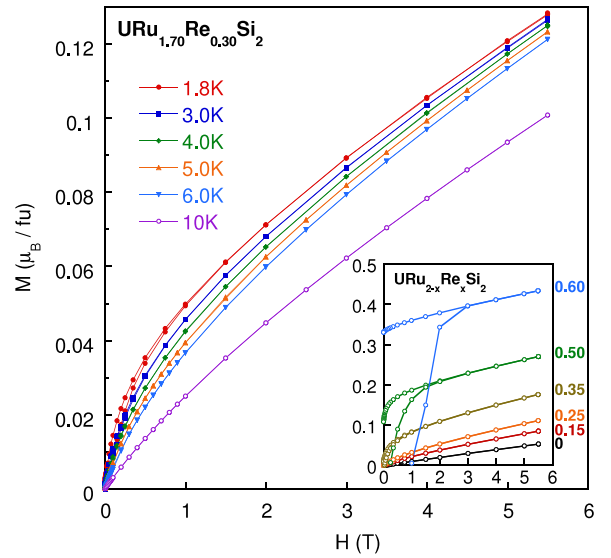


Figure 2. Magnetization isotherms for $\text{URu}_{1.70}\text{Re}_{0.30}\text{Si}_2$. Inset: a direct comparison of low-temperature data for different Re concentrations. Data for $x = 0.60$ were measured at 1.8 K, while the other data were taken at 2.0 K.

$x = 0.20$ to 0.60 , the magnitude of the magnetization increases, the isotherms become more sharply curved and hysteresis develops, which is clearly shown in the inset of figure 2. Although the parent compound URu_2Si_2 exhibits linear $M(H)$ in this field range, in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$, the isotherms exhibit gentle curvature, which does not saturate. Despite the dramatic increase of the low- T magnetization with Re concentration, even at $x = 0.60$, well within the FM state, the moment is a small fraction of the high-temperature paramagnetic effective moment of approximately $3.8 \mu_B$.

To analyze $M(H)$ data from single crystals, a critical scaling approach based on the Arrott–Noakes equation of state [20] has been employed. This approach does not assume mean-field behavior, and yields values of T_C , the ordered moment M_0 and the magnetic critical exponents β , γ , and δ . The three exponents are defined in terms of M , H , and reduced temperature $t = \frac{(T-T_C)}{T_C}$ by $M \sim t^\beta$ for $t < 0$, $M \sim H^{1/\delta}$ for $t = 0$, and $\frac{\partial M}{\partial H} = \chi \sim t^{-\gamma}$ for $t > 0$. Only two exponents are independent, as $\delta - 1 = \gamma/\beta$. The self-consistent determination of their values requires agreement between three different analyses: an initial estimate of δ and T_C made by identifying the isotherm with constant power-law behavior over the widest range of H ; scaling in $|M|/|t|^\beta$ versus $H/|t|^{\delta\beta}$, i.e., the collapse of $M(H)$ data onto two diverging curves, for $t > 0$ and $t < 0$; and modified Arrott plots of $|M|^{1/\beta}$ versus $|H/M|^{1/\gamma}$. Figure 3 exhibits the results of the scaling analysis for the $M(H)$ data measured on $\text{URu}_{1.70}\text{Re}_{0.30}\text{Si}_2$. This analysis has been applied successfully to samples with $0.20 \leq x \leq 0.60$, which demonstrates the existence of FM order in this range of Re concentration.

In figure 4, it is apparent that the x -dependence of the exponents is linear for all $0.20 \leq x \leq 0.60$, but the x -dependence of T_C and M_0 can be fit to a line only for $x \leq 0.50$. In $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$, T_C , M_0 , γ and $(\delta - 1)$ all extrapolate to 0

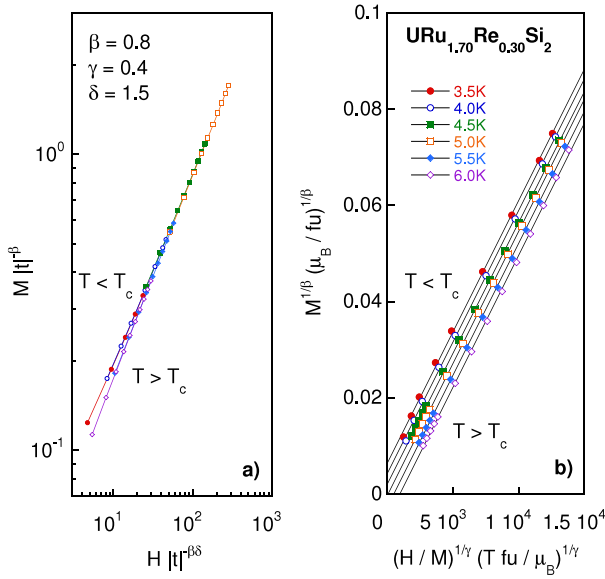


Figure 3. Arrott–Noakes scaling law and modified Arrott plots for $\text{URu}_{1.70}\text{Re}_{0.30}\text{Si}_2$. (a) In the scaling plots, $|M|/|t|^\beta$ versus $H/|t|^{\beta\delta}$, the $M(H)$ data collapse onto two curves for $T > T_c$ and $T < T_c$. (b) In the modified Arrott plots, $|M|^{1/\beta}$ versus $(H/M)^{1/\gamma}$, the $M(H)$ data are linearized and evenly spaced in temperature.

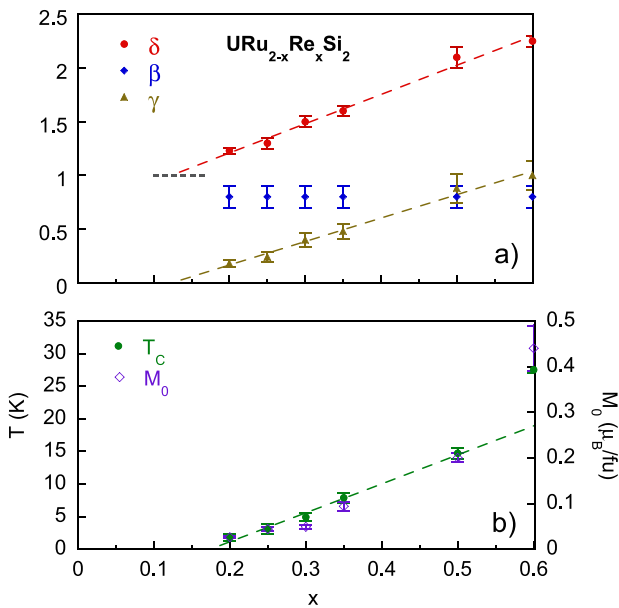


Figure 4. Re concentration dependence of magnetic critical exponents, critical temperature, and ordered moment of $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$. The x -dependence of δ and γ , and T_c and M_0 (for $x \leq 0.5$) is well described by linear fits. The values of $\delta = 1$, γ , T_c , and M_0 extrapolate to zero near $x = 0.15$. Error bars denote the range of values that satisfy the scaling analysis.

at $x = 0.15 \pm 0.03$. This is the first documented example of such a trend. The critical exponents deviate substantially from mean-field values and values describing classical FM transitions, where $\beta < 0.5$ and $\delta > 3$. Near the quantum phase transition, as x decreases, $\delta \rightarrow 1$ as the $M(H)$ isotherms (at $T = T_c$) become less curved. Because β remains constant, $\gamma \rightarrow 0$, implying a sharpening of the curvature of the

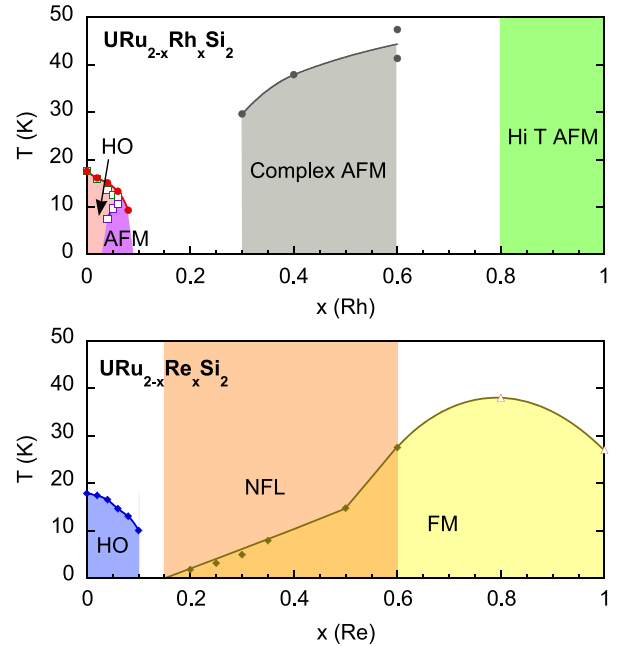


Figure 5. Comparison of the phase diagrams of $\text{URu}_{2-x}\text{Rh}_x\text{Si}_2$ and $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ up to 50% substitution. In $\text{URu}_{2-x}\text{Rh}_x\text{Si}_2$, the hidden order appears to be suppressed by $x = 0.10$, although for $0.04 \leq x \leq 0.06$, HO gives way to larger-moment AFM order. The complex AFM and high- T AFM phases are characterized by local-moment order. Closed circles follow [10] and open squares follow [11]. In $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$, the hidden order persists to $x = 0.10$. Ferromagnetic order appears to emerge near $x \approx 0.15$, and T_c increases with x until $x = 0.8$. Structural heterogeneity sets in near $x = 1$. Open triangles follow [4]. The range of NFL behavior was established in [15, 16].

divergent susceptibility at T_c . While this behavior looks like a trend towards a first-order transition, note the simultaneous suppression of the order parameter M_0 .

The Re concentration dependence of the HO and FM phases in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ is shown in figure 5. Although the HO transition can only be determined for $x \leq 0.10$, the HO phase boundary does extrapolate to 0 K near $x \approx 0.15$, where it might meet the FM phase at a multicritical point. There is no direct evidence yet that the HO and FM phases meet, but it has been observed that AFM correlations and magnetic excitations associated with the hidden order phase persist to $x = 0.35$ [16]. Such correlations provide a plausible explanation for the unusual critical behavior of the FM ordered phase. Another possible cause is a competition between magnetic order and the heavy fermion state in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$. However, the details of this interplay are not certain, nor is it known whether itinerant FM arises from a light band or a heavy band in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$. FM Kondo lattice models for both light and heavy magnetic bands are consistent with the measured enhanced specific heat [21, 22]. Also shown in figure 5 is the range of x over which NFL behavior has been observed [15, 16], which appears to coincide with the FM ordered phase. There is some correlation between the NFL effects and the novel critical behavior: power-law exponents describing unconventional T -dependence of the magnetic susceptibility [15] are in good agreement with values of γ determined by the scaling analysis, implying that they actually describe the same phenomenon.

The overall phase diagram of $\text{URu}_{2-x}\text{Rh}_x\text{Si}_2$ [10, 11] differs significantly from that of $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ (figure 5), despite similar suppression of the HO phase at low x . Whereas $\text{URu}_{2-x}\text{Rh}_x\text{Si}_2$ appears to only support AFM order, the long-range magnetic order in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ is FM. The temperature scales differ rather dramatically between the two phase diagrams, as well. In particular, at high Rh concentrations, AFM order sets in at ~ 180 K, whereas in the Re phase diagram, T_C reaches only ~ 40 K. It is interesting that the intermediate AFM phase of $\text{URu}_{2-x}\text{Rh}_x\text{Si}_2$, characterized by a multi- q domain structure [10], persists over roughly the same range as the NFL behavior does in the Re case. The complex magnetic structure in $\text{URu}_{2-x}\text{Rh}_x\text{Si}_2$ and the AFM-like correlations in the FM phase of $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ [16] suggest that in both cases, competing magnetic correlations in this range of x play an important role in determining the magnetic structure. An open question is whether the HO phase in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ gives way to an AFM phase, as it does in the Rh case [11].

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