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 URu₂₋ₓReₓSi₂, 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 ≈ 0.15, and this is accompanied by the additional suppression of the critical exponents γ and δ − 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.

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.

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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 URu$_{2-x}$Re$_x$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 $^3$He 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 $^3$He 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$_2$Si$_2$ [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 $C(T)$.

Figure 1. Electrical resistivity and specific heat anomalies associated with the hidden order transition in URu$_{2-x}$Re$_x$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 $x$.

Figure 2. Magnetization isotherms for URu$_{1.70}$Re$_{0.30}$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.

Isotherms of $M(H)$ are shown in figure 2 for URu$_{1.70}$Re$_{0.30}$Si$_2$, which orders ferromagnetically below approximately 5 K. As the Re concentration increases from $x = 0.20$ 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$_2$Si$_2$ exhibits linear $M(H)$ in this field range, in URu$_{2-x}$Re$_x$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 $\beta$, $\gamma$, and $\delta$. The three exponents are defined in terms of $M$, $H$, and reduced temperature $t = \frac{M}{M_0}$ by $M \sim t^\beta$ for $t < 0$, $M \sim H^{1/\gamma}$ for $t > 0$, and $\frac{dM}{dH} \sim x^{1/\delta} t^{-\gamma}$ 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 exponents: an initial estimate of $\delta$ 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 $H/|t|^{1/\gamma}$, 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 URu$_{1.70}$Re$_{0.30}$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 URu$_{2-x}$Re$_x$Si$_2$, $T_C$, $M_0$, $\gamma$ and $(\delta - 1)$ all extrapolate to 0.
In the modified Arrott plots, the $M(H)$ data collapse onto two curves for $T > T_c$ and $T < T_c$. The critical exponents deviate substantially from mean-field values and values describing classical FM behavior: power-law exponents describing unconventional FM Kondo lattice models for both light and heavy magnetic bands are consistent with the measured enhanced specific heat [21, 22]. Although the HO phase boundary does extrapolate to 0 K near $x \approx 0.15$, where 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 URu$_2$-$x$Re$_x$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 $x = 0.10$. 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].
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 $\sim180$ K, whereas in the Re phase diagram, $T_C$ reaches only $\sim40$ 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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