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Two superconducting phases in $CeRh_{1-x}Ir_xIn_5$

M. Nicklas,* V. A. Sidorov,† H. A. Borges,‡ P. G. Pagliuso,§ J. L. Sarrao, and J. D. Thompson
Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

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Pressure studies of $CeRh_{1-x}Ir_xIn_5$ indicate two superconducting phases as a function of x, one with $T_c \geqslant 2$ K for $x<0.9$ and the other with $T_c<1.2$ K for $x>0.9$. The higher T_c phase, phase 1, emerges in proximity to an antiferromagnetic quantum-critical point; whereas, Cooper pairing in the lower T_c phase 2 is inferred to arise from fluctuations of a yet to be found magnetic state. The *T-x-P* phase diagram of $CeRh_{1-x}Ir_xIn_5$, though qualitatively similar, is distinctly different from that of $CeCu_2(Si_{1-x}Ge_x)_2$.

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As a conventional superconductor is cooled below T_c, a finite-energy gap in the electronic density of states $N(E_F)$ opens over the entire Fermi surface. This gap to quasiparticle excitations produces an exponential temperature dependence of physical properties that depend on $N(E_F)$, e.g., specific heat, thermal conductivity, and spin-lattice relaxation rates. In contrast, power-law dependences of these properties are found in superconducting heavy-fermion systems[1] as well as in cuprates,[2] ruthenates,[3] and low-dimensional organics.[4] The existence of these power laws can be understood if the superconducting energy gap, instead of being everywhere finite, is zero on parts of the Fermi surface so that the excitation spectrum starts from zero energy. These qualitative departures from conventional behavior suggest that Cooper pairing may be mediated by excitations other than phonons. In each class of materials mentioned above, a "dome" of superconductivity emerges in proximity to a magnetic transition that is tuned toward zero temperature by applied pressure or changes in chemical composition. The close proximity to magnetism and evidence for power-law behaviors below T_c argue for magnetically mediated superconductivity in which the orbital component of the superconducting order parameter is greater than zero and power laws reflect the nodal structure of the order parameter.[5]

With two notable counter examples, a single dome of superconductivity tends to appear only in a relatively narrow range of tuning parameter values. One of these counter examples is $U_{1-y}Th_yBe_{13}$. In this case, substitutions of nonmagnetic Th for U cause a nonmonotonic variation of $T_c(y)$ with a sharp, nonzero minimum in T_c near $y=0.019$ that is not due simply to pair-breaking effects, since superconductivity persists to at least $y=0.06$.[6] Pressure studies[6] of the $T_c(y)$ phase diagram reveal that the minimum in T_c near $y=0.019$ evolves into a range of y where there is no superconductivity and provide convincing evidence that the T_c minimum at atmospheric pressure delineates two distinct superconducting phases. Though weak magnetism coexists with unconventional superconductivity for $0.019<y<0.042$ at atmospheric pressure, the origin of two distinct transitions remains unclear.

The other counter example is the prototypical heavy-fermion compound $CeCu_2Si_2$.[7] Until recently, its inexplicably robust superconductivity with respect to pressure and the complex variation of $T_c(P)$ has appeared incompatible with magnetically mediated superconductivity. Detailed pressure studies of $CeCu_2Si_2$ and its slightly larger volume relatives $CeCu_2(Si_{1-x}Ge_x)_2$ reveal the existence of two distinct domes of different superconducting phases, one at low pressures controlled by proximity to an antiferromagnetic quantum-critical point and a second at higher pressures that coincides with a weakly first-order phase boundary delineating an isostructural volume collapse.[8] The former is consistent with a magnetic pairing mechanism, whereas the latter suggests that density fluctuations and associated Ce-valence fluctuations are involved in Cooper pairing.

$CeRh_{1-x}Ir_xIn_5$ is a candidate for demonstrating two superconducting phases. $CeRhIn_5$ (Ref. 9) and $CeIrIn_5$ (Ref. 10) are isostructural, isovalent heavy-fermion compounds that form solid solutions in which the ratio of tetragonal lattice parameters, c/a, varies linearly across the series.[11] With progressive substitutions of Rh by Ir in $CeRh_{1-x}Ir_xIn_5$, the ground state at atmospheric pressure evolves continuously, just as it does in $CeRhIn_5$ with applied pressure,[12] from antiferromagnetic $(x<0.3)$ to antiferromagnetic with coexisting superconductivity $(0.3<x<0.6)$ and finally to superconducting without apparent evidence for long-range magnetic order $(x>0.6)$.[11] As the end composition $CeIrIn_5$ is approached, there is a cusp-like minimum in $T_c(x)$ near $x=0.9$ where bulk superconductivity is suppressed. The specific heat anomaly at T_c for this composition is small, $\Delta C/\gamma T_c \approx 0.14$, which is only about 10% of the weak-coupling BCS value,[13] and may be nonzero because of slight variations in Rh/Ir concentrations throughout the sample. Though suppression of bulk superconductivity with small additions of Rh in $CeIrIn_5$ might arise from Cooper-pair breaking by nonmagnetic Rh "impurities," for $x>0.9$ or $x<0.9$, the specific-heat jump at T_c is comparable to the BCS value,[11,13] and below T_c, the relaxation rate $1/T_1 \propto T^3$ and specific heat divided by temperature $C/T \propto T$, indicative of unconventional superconductivity.[14] As we will show, the cusp in T_c near $x=0.9$ in $CeRh_{1-x}Ir_xIn_5$ evolves with applied pressure to become a range of compositions that separates two superconducting phases.

Simultaneous electrical resistivity and ac susceptibility measurements were used to study the response to pressure of high quality single crystals of $CeRh_{1-x}Ir_xIn_5$ for $x=0$, 0.1, 0.25, 0.5, 0.75, 0.85, and 1. The crystals, grown from excess in flux, were carefully screened at atmospheric pressure by

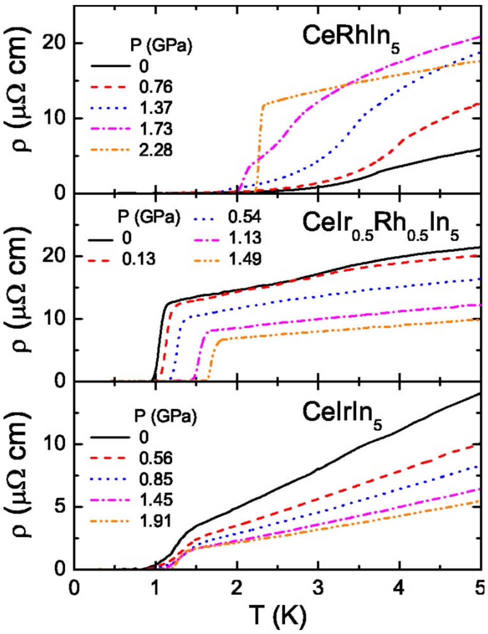

FIG. 1. (Color online) Resistivity vs temperature for three compositions of $CeRh_{1-x}Ir_xIn_5$ at representative pressures. Responses at other values of x are intermediate to those shown here.

superconducting quantum interference device magnetometry to ensure the absence of free In. Pressures to 2.3 GPa were generated in a Be-Cu clamp-type cell with Flourinert as the pressure-transmitting medium, and at least seven, approximately equally spaced, pressure measurements were made on each composition. The inductively measured shift in the superconducting transition of high purity Sn or Pb determined the clamped pressure at low temperatures.

Figure 1 shows the electrical resistivity at various pressures for $x=0$, 0.5, and 1 in $CeRh_{1-x}Ir_xIn_5$. These responses are representative of the series. For $x<0.5$, the low-temperature resistivity increases initially with applied pressure and the temperature T_{max} at which the resistivity is a maximum (not shown) decreases with P. Near and above $x=0.5$, opposite trends appear—the low-temperature resistivity decreases and the resistivity maximum moves to higher temperatures with applied pressure. As seen in Fig. 1, Rh/Ir substitutions have a small effect on potential scattering since the limiting resistivity just above either an antiferromagnetic or superconducting phase transition at atmospheric or high pressure varies from about 2 $\mu\Omega$ cm for $x=0$ and 1.0 to about 7 $\mu\Omega$ cm for $x=0.5$. Qualitatively, this reflects Nordheim's rule for isovalent substitutions[15] and is a further indication of sample homogeneity. Superimposed on this frozen disorder scattering are comparable or larger pressure-dependent changes in the inelastic scattering rate. For $x<0.5$, pressure enhances the scattering rate as magnetic order is replaced by superconductivity; whereas, for $x=0.5$, the scattering rate at atmospheric pressure is already relatively large and decreases with applied pressure, and this trend continues with increasing x. The variation in the low-temperature resistivity of this $CeRh_{1-x}Ir_xIn_5$ series at atmospheric pressure is analogous to responses found in

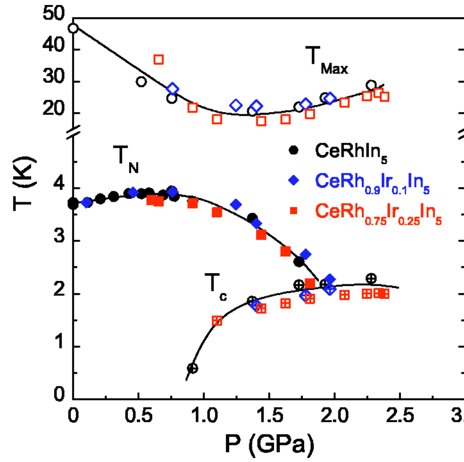

FIG. 2. (Color online) Pressure dependence of the temperature T_{max} where the resistivity is a maximum, the Néel temperature T_N, and superconducting transition temperature T_c for $x=0$ (circles), 0.1 (diamonds), and 0.25 (squares) in $CeRh_{1-x}Ir_xIn_5$. Data, shifted by constant pressures of 0, 0.1, and 0.6 GPa for $x=0$, 0.1 and 0.25, respectively, scale onto common curves as shown.

several antiferromagnets as they are tuned by applied pressure toward a quantum critical point.[16] This analogy argues that Ir substitution for Rh acts principally as an effective applied pressure and that there is a quantum-critical point at atmospheric pressure in the series near $x\geqslant 0.5$. Indeed, the ambient-pressure Néel temperature drops to $T=0$ at $x_c\approx 0.65$ where the specific heat begins to diverge logarithmically,[17] and, as shown in Fig. 2, Ir substitution and applied pressure are demonstrably equivalent for $x\leqslant 0.25$. The rigid shift by a constant pressure of the superconducting transition $T_c(P)$, the Néel temperature $T_N(P)$, and the temperature $T_{max}(P)$, where the resistivity is a maximum, scales each onto a common curve. For these three compositions, the rigid pressure shift P_r(GPa) $\approx 10x^2$, which, extrapolating to $x=1$, implies that $CeIrIn_5$ is under an effective chemical pressure of about 10 GPa relative to $CeRhIn_5$. This straightforward scaling breaks down for $x>0.3$, indicating additional effects of Ir substitution.

Linear interpolations of $T_c(P)$, defined by the onset of a diamagnetic response in ac susceptibility, and $T_N(P)$, determined from a change in slope of $\rho(T)$, for each value of x allow the construction of isobaric $T-x$ phase diagrams. Data in the upper panel of Fig. 3 are results from ambient-pressure measurements,[11] and those in the middle and lower panels are representative $T-x$ diagrams at pressures of 1.0 and 1.75 GPa. Similar isobaric diagrams at intermediate pressures confirm the smooth evolution seen in Fig. 3, and, in particular, the $T-x$ phase diagram at 2 GPa shows no evidence for antiferromagnetism. As seen in Fig. 3, the cusplike suppression of T_c near $x=0.9$ at $P=0$ evolves with applied pressure to become a range of compositions $0.75\leqslant x\leqslant 0.85$, where no bulk superconductivity is detected above 0.3 K by ac susceptibility. Therefore, in $CeRh_{1-x}Ir_xIn_5$ under pressure, there is a range of compositions separating two superconducting phases, phase 1 with $T_c\geqslant 2$ K for $x<0.75$ and phase 2 with $T_c<1.2$ K for $x>0.85$.[18]

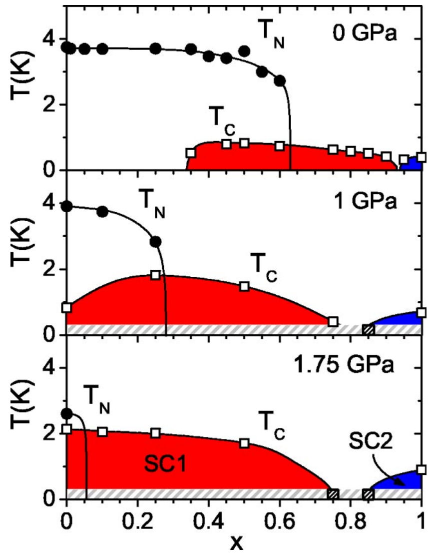

FIG. 3. (Color online) Representative $T-x$ phase diagrams for $CeRh_{1-x}Ir_xIn_5$ at $P=0$, and 1, 1.75 GPa. The cusplike suppression of T_c near $x=0.9$ at $P=0$ evolves into a range of compositions where $T_c<0.3$ K at higher pressures. Plots of $T_c(P)$ for $x=0.75$ and 0.85 (not shown explicitly) strongly suggest that $T_c=0$ for these compositions at 1.75 GPa and higher pressures. SC1: phase-1 superconductivity; SC2: phase-2 superconductivity.

The results of Fig. 3 appear analogous to the evolution of $T_c(y,P)$ in $U_{1-y}Th_yBe_{13}$, particularly, if we consider $CeRh_{1-x}Ir_xIn_5$ as Rh-doped $CeIrIn_5$, as well as to the observation of two superconducting phases in $CeCu_2(Si,Ge)_2$. In the latter, each dome of superconductivity is controlled by proximity to a distinctly different transition that is tuned to $T\to 0$ by pressure. This conclusion was possible by realizing that Ge substitution for Si expands the unit-cell volume and that this expansion can be compensated by an externally applied pressure to produce nearly identical superconducting phase diagrams as a function of cell volume for both $CeCu_2Si_2$ and $CeCu_2Ge_2$.[8] A similar argument is inferred from the pressure scaling shown in Fig. 2. If the primary role of Ir substitutions for Rh is to decrease the cell volume, then the observation of two superconducting phases in $CeRh_{1-x}Ir_xIn_5$ suggests that a second superconducting phase also might emerge in $CeRhIn_5$ at much higher pressures than investigated here. Besides a dome of superconductivity centered near the antiferromagnetic critical point at $P_c \approx 2.5$ GPa, where T_c exceeds 2 K, Muramatsu *et al.*[19] have reported a second dome of superconductivity in $CeRhIn_5$ with a maximum $T_c\approx 1$ K near 6.5 GPa. Considering that details of Ir/Rh substitution were ignored in estimating the effective pressure in $CeIrIn_5$, this estimate and the observed pressure of 6.5 GPa are in good agreement and further suggest that the second, high-pressure dome of superconductivity in $CeRhIn_5$ is analogous to phase-2 superconductivity in $CeRh_{1-x}Ir_xIn_5$.

This simple volume-based extrapolation was implied from the empirical observation that $P_r\propto x^2$ for $x\leqslant 0.25$. Studies at atmospheric pressure show, however, that T_c's of $CeRh_{1-x}Ir_xIn_5$ are a linear function of the ratio of tetragonal lattice parameters c/a and not cell volume (a^2c).[20] This apparent dichotomy suggests that c/a is not a monotonic function of pressure even though the cell volume is. Pressure-dependent structural studies of $CeRhIn_5$ confirm this suggestion:[21] c/a exhibits two maxima as a function of pressure, one near 2.5 GPa and a second near 6 GPa. The correspondence between these maxima and those in $T_c(P)$ reinforces the relationship between T_c and c/a found in the Rh/Ir solid solutions at atmospheric pressure. The pronounced nonmonotonic variation of $c/a(P)$ in $CeRhIn_5$, though not directly established in other members of $CeRh_{1-x}Ir_xIn_5$ or in isostructural $CeCoIn_5$, also may account for the different responses of T_c to uniaxial pressure observed in $CeIrIn_5$ and $CeCoIn_5$.[22]

Though the emergence of two superconducting phases in $CeRh_{1-x}Ir_xIn_5$ and $CeRhIn_5$ under pressure appears similar to the nonmonotonic variation of $T_c(P)$ in $CeCu_2(Si,Ge)_2$, there is an important distinction. In the latter, there are well-defined regimes of pressure where $T_c<1$ K and $T_c>2$ K, but, the high-pressure, high-T_c regime is accompanied by topological changes in the Fermi surface[23] and/or an increase in ground-state degeneracy[24,25] so that superconductivity with different T_c's develops out of qualitatively different electronic states. This is not true in $CeRh_{1-x}Ir_xIn_5$ and $CeRhIn_5$ under pressure. de Haas–van Alphen studies find that, except for expected quantitative changes due to their slightly different ratio of tetragonal lattice parameters, $CeIrIn_5$ at $P=0$ and superconducting $CeRhIn_5(P>0)$ have the same Fermi-surface topology and comparably large quasiparticle masses.[26,27] Furthermore, at atmospheric pressure, the electronic entropy to 5 K is $(0.5\pm 0.05)R\ln 2$ for all x,[11] indicating the same ground-state degeneracy.

On the basis of scaling shown in Fig. 2, we assume reasonably that superconductivity in phase 1 has the same origin as in $CeRhIn_5$ near and below 2.5 GPa, namely that superconductivity is mediated by excitations associated with proximity to an antiferromagnetic quantum-critical point. The pairing mechanism for phase-2 superconductivity is not so obvious since antiferromagnetic order appears to be well removed from this part of the phase diagram and there is no evidence for a line of valence transitions as a function of x or P. Like superconductivity in phase 1 where $C/T\propto T$ and $1/T_1\propto T^3$ below T_c, the same power laws are found[28] in $CeIrIn_5$, which is representative of phase-2 superconductivity, and indicate an unconventional mechanism for superconductivity in phase 2. The pairing mechanism for phase 2 is suggested from thermal expansion measurements on $CeIrIn_5$ in a field sufficient to destroy bulk superconductivity. In these experiments, the coefficient of c-axis thermal expansion $\alpha_c=aT^{0.5}+bT$, a temperature dependence expected for thermal expansion dominated by three-dimensional critical fluctuations at an antiferromagnetic quantum-critical point.[22] These observations, together with a non-Fermi-liquid like $1/T_1$ above T_c in $CeIrIn_5$,[14,29] imply that phase-2 superconductivity in $CeRh_{1-x}Ir_xIn_5$ for $x>0.85$ and, by inference, in $CeRhIn_5$ at $P>5$ GPa is mediated by fluctuations arising from some form of hidden magnetic order. One possibility is that this hidden order manifests itself in field-induced magnetic transitions observed in $CeIrIn_5$, near 40 T (Ref. 30) and in $CeRhIn_5$ near 50 T.[31] Whatever the precise nature of this hidden magnetic order, the lower T_c of phase-2 superconductivity suggests that pair-mediating fluctuation spectrum is more nearly three-dimensional, coupling electronic states less efficiently than magnetic excitations associated with T_N and phase-1 superconductivity.

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*Present address: Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Strasse 40, 01187 Dresden, Germany.

†Permanent address: Institute for High Pressure Physics, Russian Academy of Sciences, Troitsk, Russia.

‡Permanent address: Pontifícia Universidade Católica Rio de Janeiro, Dept. Física, BR-22452-970 Rio De Janeiro, Brazil.

§Permanent address: Cidade Universitaria, BR-13083-970 Campinas-SP, Brazil.

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