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SMR 1232 - 11

XII WORKSHOP ON
STRONGLY CORRELATED ELECTRON SYSTEMS

17 - 28 July 2000

Background Material for talk on Unconventional Magnetism and Superconductivity in Heavy Fermions CeRhIn5, CeIrIn5, CecoIn5 and related compounds

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Abstract Designation: 1E-01
Date: April 14, 2000

Superconductivity and Magnetism in a New Class of Heavy-Fermion Materials

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Abstract:
We report a new family of Ce-based heavy-fermion compounds whose electronic specific heat coefficients range from about 400 to over 700 mJ/mole Ce-K^2. Crystals in this family form as $Ce_nT_mIn_{3n+2m}$, where T= Rh or Ir, n=1 or 2, and m=1, with a tetragonal structure that can be viewed as n-layers of $CeIn_3$ units stacked sequentially along the c-axis with intervening m-layers of TIn_2. Ambient and high pressure studies show that the quasi-2D layers of $CeIn_3$ produce unconventional superconducting and magnetic ground states. This family should enable new understanding of the relationship between magnetism and superconductivity in heavy-fermion materials and more generally of why heavy-fermion superconductivity prefers to develop in one structure type and not another.

Keywords:
Heavy-fermion compounds,superconductivity, antiferromagnetism, phase transitions-pressure effects

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There is ample evidence that the superconductivity and small-moment magnetism found in cerium- and uranium-based heavy-fermion materials are unconventional and that the physics of these ground states may be related.[1] In particular, it appears that the unconventional superconducting ground state always appears in proximity to the equally unconventional magnetic state. This has led to the logical assumption that some limit of the same mechanism is responsible for both. Recent discoveries [2] of a pressure-induced transition from magnetic to superconducting states in the same material hold promise for more detailed understanding of the relationship between these two states. However, in several of these examples, the pressure required to induce the transition is rather high, making it particularly challenging to make more than just the most basic measurements, such as electrical resistivity. Ideally, what is needed to make progress on the heavy-fermion problem is a single family of materials whose ground states can be tuned easily with modest pressure and/or chemical substitutions. The $ThCr_2Si_2$ structure type is one such class of materials in which heavy-fermion magnetism and superconductivity seem to be favored, but we do not know why this is the case. The first known heavy-fermion superconductor $CeCu_2Si_2$ is a member of this class, and, by changing the precise Ce/Cu ratio in the compound, it can be tuned at ambient pressure between superconducting and magnetic states and for a fixed ratio from non-superconducting to superconducting states with pressure.[3] However, its delicate crystal chemistry has made it a very challenging system to study. The same is true of its relative $CeNi_2Ge_2$. Even if there were no chemistry difficulties, it would be valuable to have another Ce-based materials type that would allow us to understand more broadly the relationship between magnetism and superconductivity and even more generally why heavy-fermion superconductivity prefers to develop in one structure type and not another. Here, we report on a new family of materials that holds promise for making progress in these regards.

These new materials form with chemical compositions $Ce_nT_mIn_{3n+2m}$, where T= Rh or Ir. All members with m=1, n=1or 2 grow readily out of an In-rich flux as single crystals with characteristic size 1 cm x 1 cm x several mm. Powder x-ray patterns obtained on crushed single crystals show [4] that the compounds grow with a tetragonal unit cell that can be viewed as n-layers of $CeIn_3$ units stacked sequentially along the c-axis with intervening m-layers of TIn_2. Except for Eu, single-layer (n=1) and bi-layer (n=2) variants also grow with the light rare earths La through Gd.[5] The La-based materials are, as expected, Pauli paramagnets to 50 mK. Lattice parameters for the Ce-based compounds are given in Table 1. The in-plane lattice parameter a_0 is a measure of the Ce-Ce spacing within the cubic $CeIn_3$ units, and the c-axis parameter reflects the Ce-Ce spacing perpendicular to the planes for n=1. For n=2, the c-axis parameter is the repeat distance of $CeIn_3$ bilayers and a_0 is essentially the nearest neighbor distance between Ce atoms in adjacent layers. As might be expected from the quasi-2D structure, the magnetic susceptibility of these materials also is anisotropic and depends on the value of n as well as the transition metal T. In all cases, the effective magnetic moment, obtained from plots of the inverse susceptibility versus temperature above 200 K, is reduced somewhat from the Hund's rule value of 2.54 μ_B for Ce^{3+}, indicating the presence of crystal-field effects, and the low temperature susceptibility is always larger when a magnetic field is applied parallel to the c-axis. The ratio of χ_c/χ_a at low temperatures is larger for n=1 than for n=2 and for T=Rh than T=Ir, ranging from greater than 2 for n=1,T=Rh to 1.2 for n=2,T=Ir. The quality and degree of chemical order in these materials are reflected in part by values of the resistivity ratio $\rho(300\ K)/\rho(4\ K)$ that are on the order of 50-100 and by the appearance of very narrow lines in NQR spectra. Additional details of the structural and physical properties will be given elsewhere; in the following, we briefly discuss some of the low temperature properties of the n=1 compounds and mention those of the less-studied n=2 family.

In Fig.1 we plot the temperature dependence of the resistivity ρ, static susceptibility χ, and the specific heat divided by temperature C/T for $CeRhIn_5$. All exhibit a feature at 3.8 K that is associated with magnetic order. Approximately 30% of Rln2 entropy is found below 3.8 K, suggesting substantially reduced moment ordering in a crystal-field doublet ground state. The remaining 70% Rln2 magnetic entropy is recovered on warming to 18-20 K. It is somewhat difficult to define a Sommerfeld coefficient from the C/T data just above T_N, but a simple entropy-balance construction, $S(T_N-\varepsilon)=S(T_N+\varepsilon)$, gives $\gamma \approx 400$ mJ/mole-K^2. The susceptibility reaches a maximum near $T_{\chi m}=7.5$ K before dropping more steeply at T_N. Cerium-based correlated electron materials, in which J=5/2, commonly exhibit a low temperature maximum in their susceptibility that is expected from the theory of orbitally degenerate Kondo impurities.[6] The specific heat and crystal structure of $CeRhIn_5$, however, suggest that its ground state is doubly degenerate, in which case the Kondo effect produces the maximum susceptibility at T=0. An alternative interpretation for the maximum comes from the quasi-2D structure. It is known [7] that a 2D, spin-1/2 Heisenberg system exhibits a maximum susceptibility at $T_{\chi m}\approx 0.93\,|J|$, which for $T_{\chi m}=7.5$ K, gives $|J|=8$ K. Any deviation from purely 2D exchange produces long range order at $T_N \approx S(S+1)\,|J|/2=$ 3K, which is close to the experimentally observed value of 3.8 K. This simple picture, which neglects Kondo effects that are undoubtedly present, may at least qualitatively account for the susceptibility maximum in $CeRhIn_5$.

The qualitative view of the magnetic state inferred from specific heat and susceptibility measurements is consistent with more microscopic information obtained from ^{115}In NQR data. In the $CeRhIn_5$ structure, there are two inequivalent In sites: a single In(1) site, analogous to the single In site in $CeIn_3$, and four In(2) sites, two on each of the lateral faces of the unit cell that are equidistant above and below the Rh layer. Results of an analysis of NQR measurements on the In(1) site are shown in Fig.2. The internal magnetic field produced by magnetic order grows rapidly below T_N and saturates to a value of 0.17 T. This internal field compares to 0.5 T found at the equivalent In site in bulk $CeIn_3$, for which neutron diffraction studies show an ordered moment of 0.4 μ_B.[8] Assuming a similar hyperfine coupling in both materials, the ordered moment in $CeRhIn_5$ should be 0.1-0.2 μ_B and, by symmetry considerations, lies in the tetragonal plane. A fit of H_{int} versus $(1-T/T_N)^\beta$ near T_N gives a critical exponent $\beta=0.25\pm0.03$, which is about one half the mean field value of $\beta=0.5$. Similarly rapid growth of the sublattice magnetization below T_N also has been found in La_2CuO_4 in which a reduced critical exponent is related to its 2D magnetism.[9] Preliminary analysis of NQR spectra from the In(2) sites suggest that the ordered moments maintain a constant magnitude within each plane, but their ordered direction is modulated by a spiral rotation along the c-axis that is incommensurate with the lattice.

Application of pressure to the heavy-fermion antiferromagnet $CeIn_3$ drives its Néel temperature smoothly from 10 K at atmospheric pressure toward zero at a critical pressure of 25 kbar, where superconductivity develops with $T_c \approx 0.25$K.[10] The pressure response of $CeRhIn_5$ is qualitatively different. From resistivity measurements, we derive [11] the T-P phase diagram shown in Fig. 3b. The resistivity feature at 3.8 K shown in Fig.1 moves slowly with applied pressure to higher temperatures at a rate of about 9mK/kbar. This feature is present at 14.5 kbar but not at higher pressures. At 16.3 kbar, there is a broad transition beginning near 2 K to a zero resistance state; the transition width sharpens with increasing pressure to less than 0.05 K at 21 kbar, where the onset of superconductivity is at 2.17 K. The abrupt loss of a signature for T_N, the sudden appearance of superconductivity, and rapid sharpening of the transition width suggest a first-order like transition at a critical pressure between 14.5 and 16.3 kbar. AC susceptibility measurements on a second crystal reproduce the appearance of superconductivity and show a perfect diamagnetic response below T_c, which is almost an order of magnitude higher than in bulk $CeIn_3$.

Results of specific heat measurements on $CeRhIn_5$ at 19 kbar are shown in the upper panel of Fig. 3. There are several points to note about these data. C/T begins to increase more rapidly below about 5 K, where C/T = 390 mJ/mole-K^2, reaches a plateau near 2.5 K, and then shows a well-defined feature at T_c. Below T_c, C/T is linear in temperature, i.e., $C \propto T^2$ and not exponential as expected for BCS superconductivity. This power-law dependence of C is expected for a superconductor in which there are line-nodes in the gap function.[12] The state reflected in the plateau of C/T near 2.5 K also is found as a weak feature in resistivity measurements, and its pressure dependence is shown as the transition labeled $T_?$ in Fig. 3b. This phase transition first appears in resistivity measurements at pressures near 10 kbar, well below the pressure necessary to induce superconductivity, and persists in the pressure regime of superconductivity. We do not know its origin but believe it involves an instability of the Fermi surface. In this regard, we observe that the jump in specific heat ΔC, measured from the value of $C/T \equiv \gamma$ = 390 mJ/mole-K^2 at 5 K to the maximum at T_c, gives $\Delta C(T_c)/\gamma T_c = 1.5 \pm 0.1$, which is close to the weak coupling value of 1.43. That is, the transition $T_?$ plus the superconducting transition appear to gap the entire Fermi surface. Further, the $T_?$ transition is unchanged in a field of 9 T; whereas, in this field, superconductivity is suppressed below 0.3 K. We are led to the conclusion that the $T_?$ transition is one to a charge- or spin-density wave state that gaps part of the Fermi surface and that this density wave state coexists with superconductivity. What relationship there might be between the density wave and Néel states remains to be explored.

We turn now to the other n=1 member of this family, $CeIrIn_5$. Its thermodynamic and transport properties at low temperatures are summarized in Fig. 4. At 0.38 K, there is a diamagnetic transition in ac susceptibility that is coincident with a jump in heat capacity, providing clear evidence of bulk superconductivity. By comparing the magnitude of the ac susceptibility response of a sample of tin with similar size and shape at its T_c to that of $CeIrIn_5$ at its superconducting transition temperature, we estimate that the response below 0.38 K shown in Fig. 4 corresponds to perfect diamagnetism. Just above T_c, C/T is essentially constant and gives a Sommerfeld coefficient γ= 720 mJ/mole-K^2. From the average of measurements on three different crystals, the specific heat jump ΔC at T_c is equal to $(0.76 \pm 0.05)\gamma T_c$. This ratio $\Delta C(T_c)/\gamma T_c$ is comparable to that found in other heavy-fermion materials, eg. UPt_3 [13], and is smaller than the value for $CeRhIn_5$ at 19 kbar, but in that case, we also included the effect of the $T_?$ transition. The specific heat data of $CeIrIn_5$ below T_c are well-fit to the sums of nuclear-Schottky, T^2

and T-linear contributions. (A nuclear Schottky term is expected due to the large nuclear quadrupole moment of In and presumably is present in the superconducting state of $CeRhIn_5$, but at temperatures lower than shown in Fig. 3.) A significant $C \propto T^2$ contribution suggests that, as in $CeRhIn_5$, the gap function has line-nodes. The large T-linear term of 200±50 mJ/mole-K^2 indicates the presence of ungapped quasiparticle states below T_c and provides further support for the existence of zeros in the gap. The temperature dependence of the thermal conductivity, which is insensitive to the nuclear Schottky, also is described well [14] from T_c to 50 mK by the sum of linear and quadratic terms that are consistent with corresponding terms in the specific heat.

A very peculiar aspect of the data shown in Fig. 4 is that the resistivity drops to zero at T_0=1.2 K, or at least to less than 1% of the resistivity above 1.2 K, without an obvious thermodynamic signature. Our original belief was that this resistive transition was extrinsic. However, additional measurements suggested that this is intrinsic. Measurements [15] of the specific heat, ac susceptibility and electrical resistivity, with all three measurements made on each of three separately grown crystals, in magnetic fields to 9 T applied parallel and perpendicular to each sample's c-axis show that the anisotropic response of T_c determined by specific heat and ac susceptibility is identical to that determined resistively. That is, anisotropy in the field dependences of T_c and T_0 is identical. This would seem to imply that both transitions arise from a common underlying electronic structure. Preliminary studies of the response of $CeIrIn_5$ to pressure and to Rh-doping also provide evidence that the thermodynamic and resistive transitions are intrinsic and intimately linked: in both cases the bulk T_c increases and approaches T_0, which is relatively insensitive to perturbation. Presently, we have no definitive explanation for the origin of the resistive transition, but the data suggest that it is intrinsic to $CeIrIn_5$, arising possibly from filaments of locally phase coherent electron pairs for $T_c<T<T_0$ that become globally coherent through out the sample at T_c..

The n=2 variants of these materials also appear to be quite interesting, and their study in parallel with the n=1 compounds should provide insight into the role of spatial dimensionality in controlling the nature of their heavy-fermion ground states. Interestingly, the Sommerfeld specific heat coefficient of the n=2 members is the same, to within 10%, of that for the corresponding n=1, T= Rh and Ir materials, which implies that the mechanism responsible for producing the heavy-mass state depends on the transition element T and not n. This further supports the generally held belief that the large γ, a hallmark of heavy-fermions, arises from a local many-body correlation of the f-electrons with the sea of ligand electrons. However, the ground state is affected by n. Ce_2RhIn_8 orders antiferromagnetically at 2.8 K, which is 1 K lower than in $CeRhIn_5$. Very nearly the same magnetic entropy is liberated below their respective magnetic transitions. On-going studies of the effect of pressure on Ce_2RhIn_8 show that the resistive signature for T_N changes only slightly with pressure and disappears abruptly, but at a lower pressure (less than 7 kbar) than in $CeRhIn_5$. At 17 kbar and higher, the positive slope of the resistivity increases below approximately 2 K and may signal the onset of another transition. While the ground state of Ce_2RhIn_8 appears similar to that of $CeRhIn_5$, this is not the case for the Ir-containing compounds. Ce_2IrIn_8 remains a heavy-fermion paramagnet to 50 mK, with no evidence for a phase transition. As shown in Fig. 5, there is an interesting progression in the temperature at which a broken symmetry state develops in this family of materials. When that temperature is plotted as a function of the

Sommerfeld coefficient γ_V, normalized by the unit cell volume per Ce atom, magnetism appears at small γ_V and superconductivity and paramagnetism at larger values. A similar correlation has been noted [16] previously among U-based heavy-fermion materials.

In summary, we have found a new family of heavy-fermion compounds whose ground state can be tuned easily by pressure and chemical substitution. All members of this family form as high quality single crystals with consistently reproducible properties, making them particularly amenable to careful, quantitative study. Unlike previous examples, these materials also allow investigation of the role of dimensionality in controlling the heavy-fermion state.

Acknowledgments: Work at Los Alamos was performed under the auspices of the U. S. Department of Energy. The work at Berkeley was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U. S. Department of Energy under contract DE-AC03-76SF00098. Z. F. acknowledges support through NSF grants DMR-9870034 and DMR-9971348.

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Table 1. Lattice parameters, Néel or superconducting transition temperatures, and specific heat Sommerfeld coefficients for the $Ce_nT_mIn_{3n+2m}$ family of materials.

Compound	a_0 (Å)	c (Å)	T_N or T_c (K)	γ (mJ/mole Ce-K^2)
$CeRhIn_5$	4.652(1)	7.542(1)	3.8	≈400
$CeIrIn_5$	4.668(1)	7.515(2)	0.4	720
Ce_2RhIn_8	4.665(1)	12.244(5)	2.8	≈400
Ce_2IrIn_8	4.671(2)	12.214(6)	--	700

Figure captions

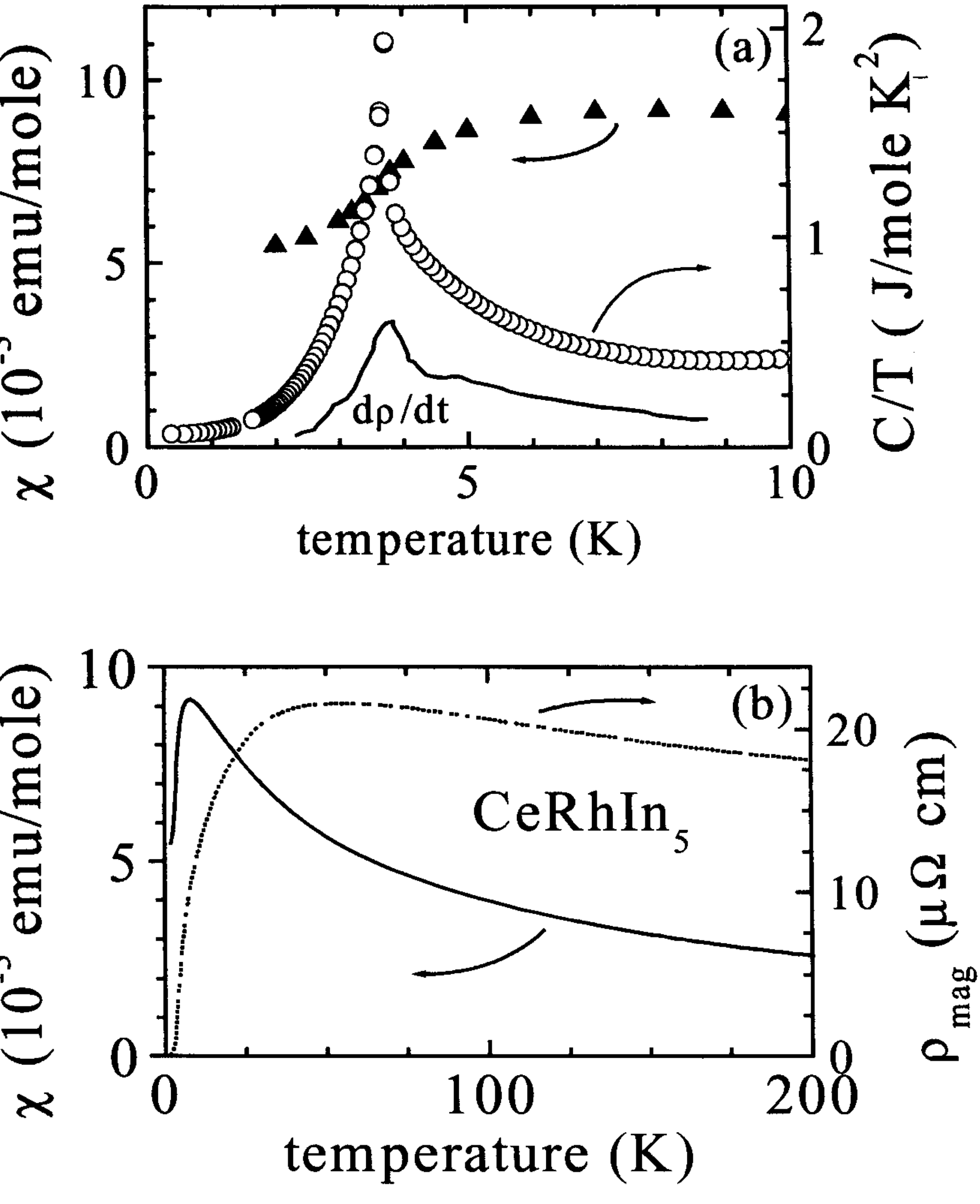
Fig. 1. (a) Magnetic specific heat ($C = C_{total}(CeRhIn_5) - C(LaRhIn_5)$) divided by temperature (circles), in-plane magnetic susceptibility χ (triangles) and temperature derivative of the electrical resistivity $d\rho/dT$ (solid line) as a function of temperature for $CeRhIn_5$ at atmospheric pressure. (b) In-plane magnetic susceptibility (solid curve) and magnetic contribution to the electrical resistivity (dotted curve) of $CeRhIn_5$ over a broader temperature range. The resistivity of $LaRhIn_5$ was subtracted from the total resistivity of $CeRhIn_5$ to obtained the magnetic contribution.

Fig. 2. Temperature dependence of the internal field at the In(1) site that develops in $CeRhIn_5$ below T_N. The solid curve is a fit to $(1-T/T_N)^{\beta}$. See text for details.

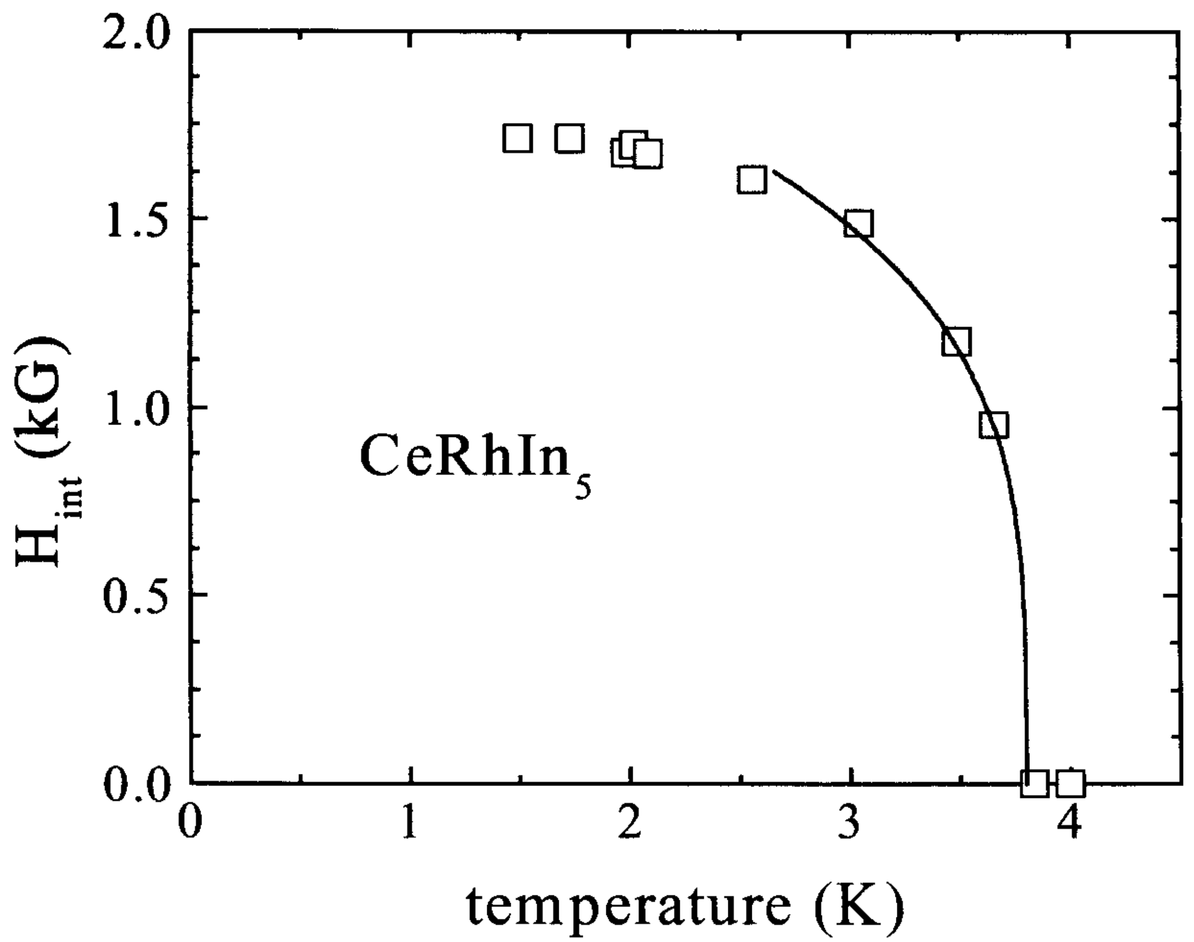
Fig. 3. (a) Magnetic contribution to the specific heat C divided by temperature (open squares) and electrical resistivity (solid circles) for $CeRhIn_5$ at 19 kbar. The magnetic contribution to C was estimated by subtracting the specific heat of $LaRhIn_5$ at atmospheric pressure from the total specific heat of $CeRhIn_5$ at pressure. (b) The temperature-pressure phase diagram determined from resistivity measurements. T_N, T_c and $T_?$ correspond to the Néel temperature, onset temperature for superconductivity, and the transition to an unknown state, respectively.

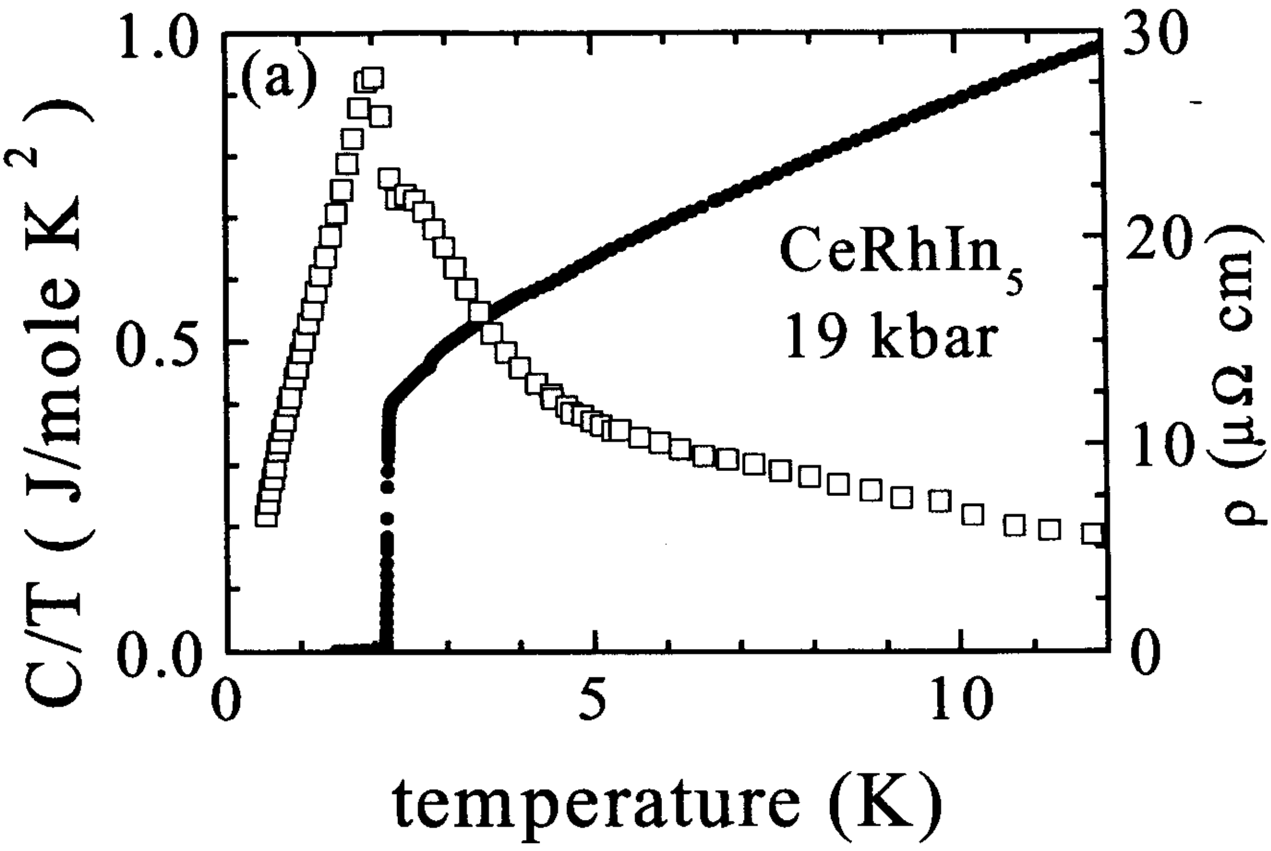
Fig. 4. Magnetic specific heat ($C = C_{total}(CeIrIn_5) - C(LaIrIn_5)$) divided by temperature (circles), ac magnetic susceptibility χ (triangles) in arbitrary units and electrical resistivity ρ (squares) as a function of temperature for $CeIrIn_5$. The solid line through C/T data is a fit as described in the text.

Fig. 5. Temperature at which a phase transition develops in the $Ce_nT_mIn_{3n+2m}$ family of materials. The horizontal axis is the Sommerfeld coefficient of specific heat normalized by the average volume of a Ce atom in a unit cell. Solid symbols denote magnetism, open symbols superconductivity or paramagnetism.

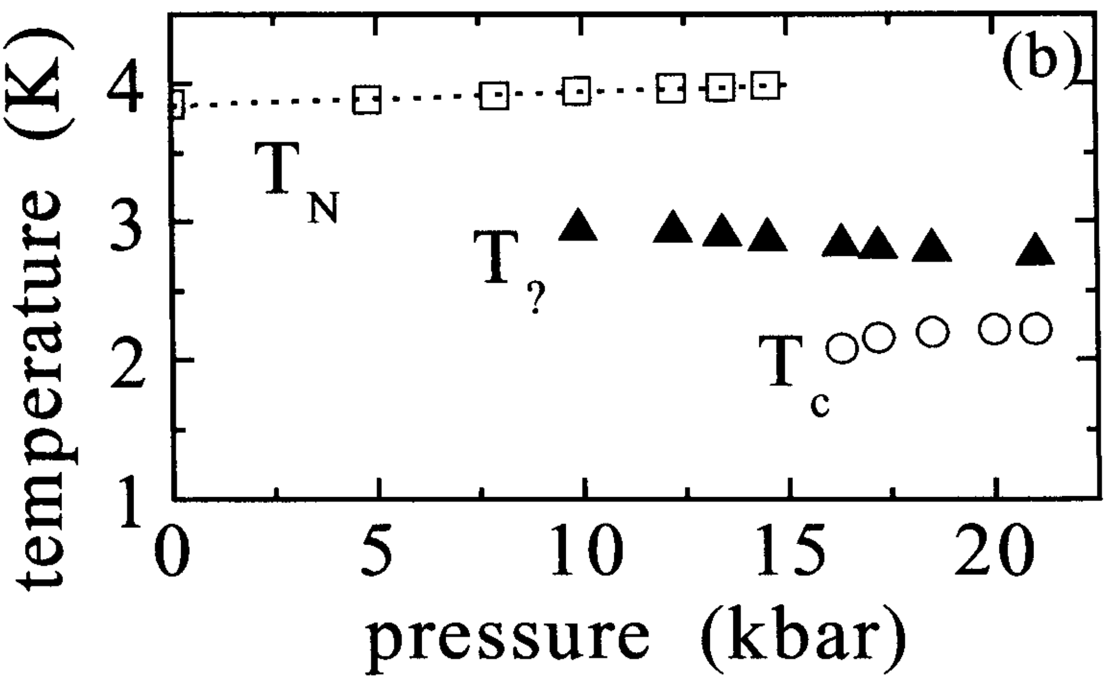

(a)
dρ/dt
χ (10^-3 emu/mole)
C/T (J/mole K^2)
temperature (K)
(b)
CeRhIn_5
χ (10^-3 emu/mole)
ρ_mag (μΩ cm)
temperature (K)

Thompson, *et al.* Fig. 2

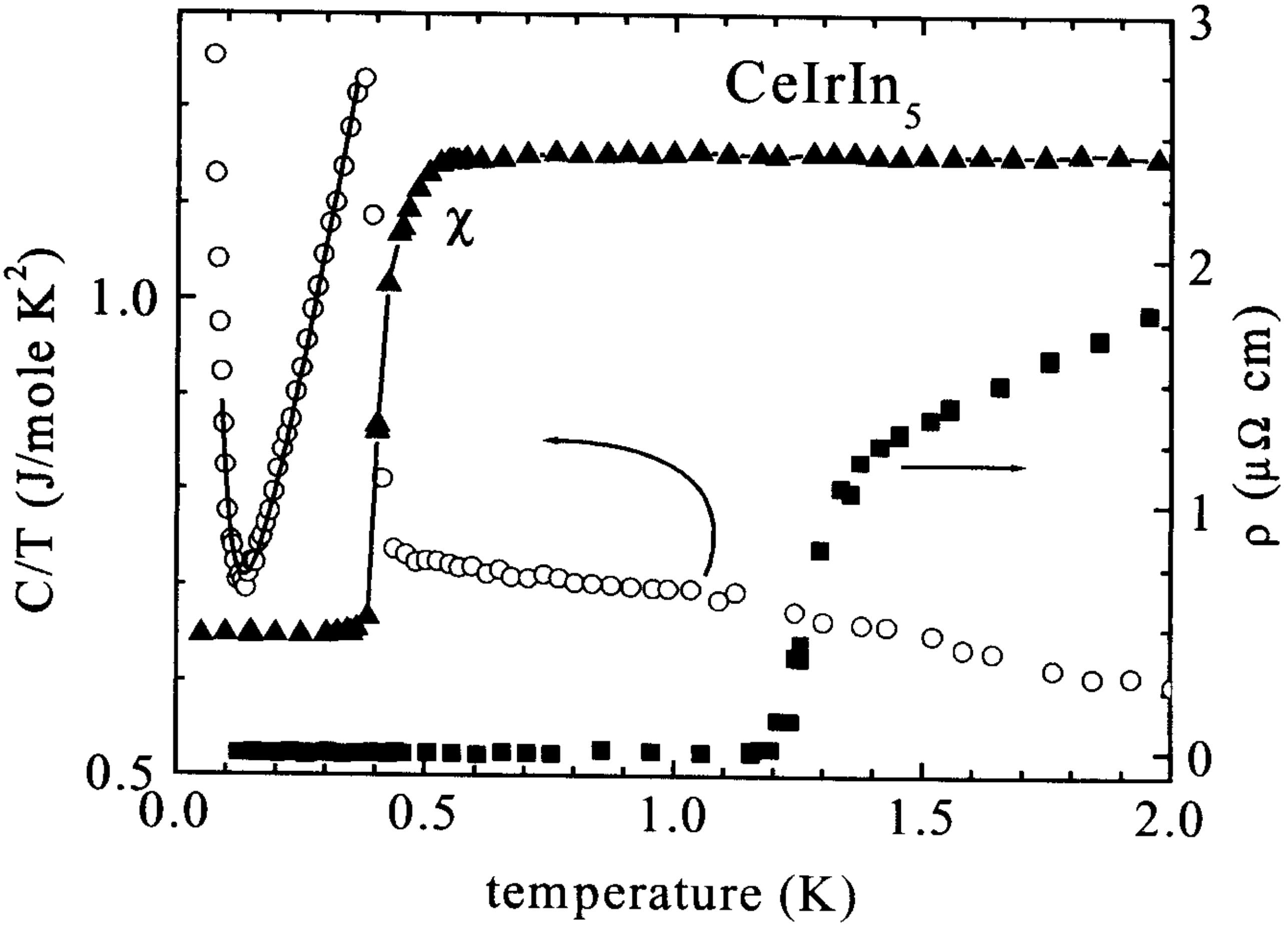




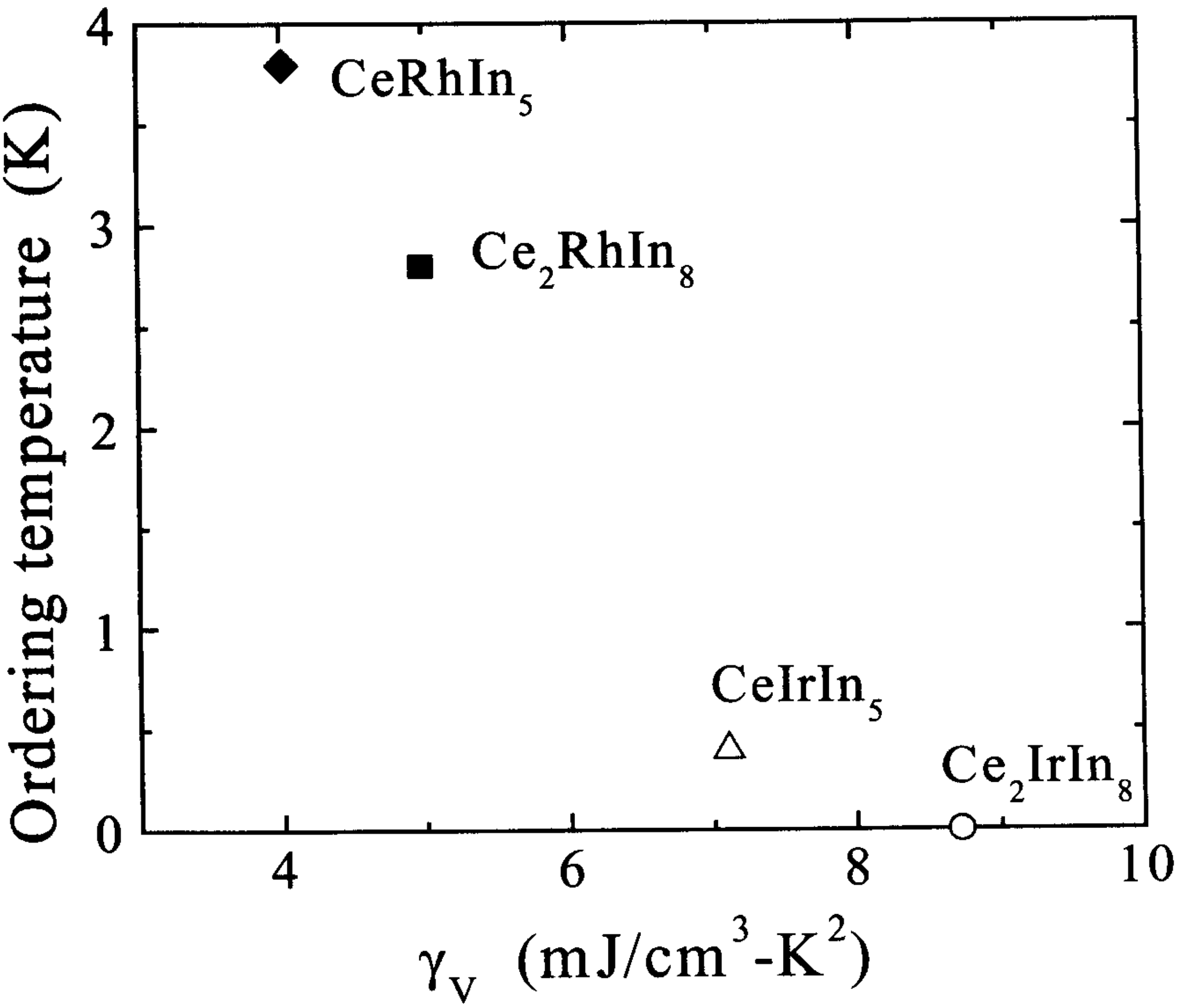
1.0
0.5
0.0
C/T (J/mole K 2)
(a)
CeRhIn5
19 kbar
30
20
10
0
ρ (μΩ cm)
0
5
10
temperature (K)



4
3
2
1
temperature (K)
(b)
T N
T ?
T c
0
5
10
15
20
pressure (kbar)


CeIrIn5
χ
C/T (J/mole K2)
ρ (μΩ cm)
temperature (K)
0.5
1.0
0.0
0.5
1.0
1.5
2.0
0
1
2
3

Thompson, *et al.* Fig. 5



Pressure-Induced Superconductivity in Quasi-2D $CeRhIn_5$

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(Received 22 November 1999)

$CeRhIn_5$ is a new heavy-electron material that crystallizes in a quasi-2D structure that can be viewed as alternating layers of $CeIn_3$ and $RhIn_2$ stacked sequentially along the tetragonal c axis. Application of hydrostatic pressure induces a first-order-like transition from an unconventional antiferromagnetic state to a superconducting state with $T_c = 2.1$ K.

PACS numbers: 74.70.Tx, 74.62.Fj, 75.30.Mb, 75.40.-s

The relationship between magnetism and superconductivity is a recurring theme of research on heavy-fermion materials. In these materials, the evolution of ground states as functions of pressure or chemical environment frequently is discussed in terms of an intuitively appealing, but qualitative, model first proposed by Doniach. This model [1] considers a one-dimensional chain of Kondo impurity atoms that experience the effect of competing long-range Ruderman-Kittel-Kasuya-Yosida (RKKY) and short-range Kondo interactions, both of which depend on a coupling constant proportional to the magnetic exchange $|J|$. Because of their different functional dependencies on $|J|$, quadratic (RKKY) and exponential (Kondo), RKKY interactions dominate for small values of $|J|$, the spin system orders magnetically, and the Néel temperature T_N increases initially with the exchange. For modest values of $|J|$, the magnetic singlet state favored by the Kondo effect competes with long-range order, producing a maximum in T_N versus $|J|$, and at sufficiently large $|J|$ drives the magnetic state toward a zero-temperature transition. Cerium is a well-known Kondo impurity ion that, when periodically placed in an appropriate crystal lattice, orders magnetically out of a strongly correlated electron state and is well suited for comparison to the Doniach model. There are several examples [2] where pressure has been used to tune the exchange in Ce-based compounds, and the observed pressure dependence of magnetic order is qualitatively that expected from this model provided $\partial|J|/\partial P > 0$. Interestingly, in several of these materials, heavy-fermion superconductivity appears as the zero-temperature magnetic-nonmagnetic boundary is approached by applied pressure [3–6]. These observations support the widely held view that heavy-fermion superconductivity is mediated by spin fluctuations that are present near this boundary [7]. Experiments also show [8] that in many of these cases superconductivity develops out of an unconventional normal state in which the electrical resistivity increases quasilinearly with temperature, in contrast to the T^2 dependence expected of a Landau Fermi liquid. Such non-Fermi-liquid behavior is expected [9] near a zero-temperature phase transition, where quantum-critical fluctuations dominate temperature dependencies of thermodynamic and transport properties.

Of the several Ce-based compounds whose ground states evolve as described above, all but one crystallize in the $ThCr_2Si_2$ body-centered tetragonal structure; cubic $CeIn_3$, which forms in the Cu_3Au structure, is the notable exception and an archetypal example. At atmospheric pressure it orders antiferromagnetically at $T_N \approx 10$ K in a Γ_7 crystal-field doublet ground state, with reduced ordered moments of about $0.5\mu_B$ that are expected as the magnetic-nonmagnetic boundary is approached. Applying pressure [5] to $CeIn_3$ monotonically reduces T_N toward zero temperature at a critical pressure $P_c \approx 25$ kbar where superconductivity sets in at temperatures $T_c \leq 0.25$ K, and for $T \gtrsim T_c$, the resistivity follows a power law $\rho - \rho_0 \propto T^n$, where $n \leq 1.5$. In the absence of contradictory observations, the substantial body of data available for these materials has provided general validation of the qualitative interpretation that follows from Doniach's model. In the following, we present results of ambient and high-pressure studies on single crystals of $CeRhIn_5$ whose response to pressure suggests that possibilities are richer than this qualitative picture envisions.

Crystals of $CeRhIn_5$ with dimensions up to 1 cm^3 were grown from an In flux. X-ray diffraction showed that the material was single phase and formed in the primitive tetragonal $HoCoGa_5$ structure type, with lattice parameters $a_0 = 4.652$ Å and $c_0 = 7.542$ Å. In this structure, $CeRhIn_5$ can be viewed as alternating layers of $CeIn_3$ and $RhIn_2$ stacked sequentially along the c axis [10]. There is a single Ce site with $4/mmm$ symmetry. The a_0 parameter corresponds to that of $CeIn_3$ units in this structure and is smaller than a_0 of bulk $CeIn_3$. Taking the bulk modulus $B = 650$ kbar [11] for $CeIn_3$, the difference in lattice parameters implies that the $CeIn_3$ units in $CeRhIn_5$ experience a chemical pressure of approximately 14 kbar relative to three-dimensional (3D) $CeIn_3$ at atmospheric pressure.

Figure 1 summarizes ambient-pressure thermodynamic and transport properties of $CeRhIn_5$. The total specific heat divided by temperature [Fig. 1(a)] is a minimum near 9 K, where its value is approximately 400 mJ/mole K^2, before reaching a maximum at 3.8 K that signals the onset of magnetic order. The inset of Fig. 1(a) plots the entropy obtained by integrating the magnetic specific

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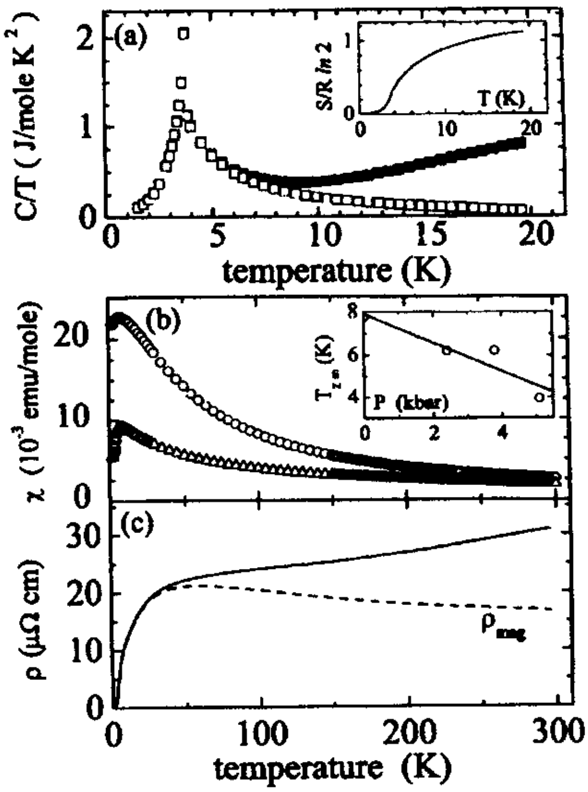

FIG. 1. (a) Specific heat divided by temperature versus temperature for $CeRhIn_5$. Solid symbols are the total specific heat; open symbols are the magnetic contribution to C/T estimated by subtracting the specific heat of $LaRhIn_5$, which has a Sommerfeld coefficient of 5.7 mJ/mole K^2 and Debye temperature of 245 K. The inset shows the magnetic entropy as a function of temperature. (b) Static magnetic susceptibility χ measured in an applied field of 1 kOe for the field along the c axis (circles) and a axis (triangles) of $CeRhIn_5$. The inset gives the pressure dependence of the temperature $T_{\chi m}$ at which χ is a maximum. The solid line is a linear fit to the data. (c) Temperature dependence of the electrical resistivity $\rho(T)$ for $CeRhIn_5$ (solid curve). At 300 K, the c-axis resistivity is approximately twice as large as that in the basal plane. The dashed curve is the magnetic contribution estimated by subtracting the resistivity of $LaRhIn_5$.

heat as a function of temperature. Only 30% of $R \ln 2$ entropy is released below T_N, suggesting a substantial Kondo-compensated ordered moment, consistent with nuclear quadrupole resonance measurements [12] that find an ordered moment of $(0.1-0.2)\mu_B$. The remaining 70% $R \ln 2$ entropy is recovered on warming to 18–20 K and is reflected in the long high-temperature tail in the magnetic contribution to C/T. From C/T data, it is difficult to define precisely the electronic specific heat above T_N, but a simple entropy-balance construction $[S(T_N - \varepsilon) = S(T_N + \varepsilon)]$ gives a Sommerfeld coefficient $\gamma \gtrsim 420$ mJ/mole K^2. The magnetic susceptibility χ [Fig. 1(b)] has no detectable in-plane anisotropy and an easy axis of magnetization along the c direction. Paramagnetic Curie-Weiss temperatures, obtained from linear fits of the inverse susceptibility for $T > 100$ K, are -79 K in the a-b plane and $+16$ K for the field along the c axis. A polycrystalline average of these data gives a high-temperature effective moment $\mu_{eff} = 2.38\mu_B$, reduced somewhat from the Hund's rule value of $2.54\mu_B$ for Ce^{+3} by crystal-field splitting of the $J = 5/2$ manifold, and paramagnetic Weiss temperature of -31 K. The susceptibility exhibits a weak maximum near $T_{\chi m} = 7.5$ K, independent of field direction, before dropping more steeply at T_N. Ce-based compounds, in which $J = 5/2$, commonly exhibit a low-temperature maximum in χ that is expected from the theory [13] of orbitally degenerate Kondo impurities. However, specific heat measurements as well as the crystal symmetry suggest that the ground state of $CeRhIn_5$ is doubly degenerate, in which case the Kondo effect produces the maximum susceptibility at $T = 0$. The electrical resistivity ρ of $CeRhIn_5$ [Fig. 1(c)] is similar in magnitude and temperature dependence to that of bulk $CeIn_3$[5] and is typical of nearly defect-free cerium heavy-fermion materials: weakly temperature dependent above 100 K and falling rapidly at lower temperatures to a value of approximately 0.4 $\mu\Omega$ cm at 0.4 K. Traditionally, this behavior is interpreted [14] as indicating a crossover from incoherent Kondo scattering at high temperatures to the formation of heavy-electron Bloch states at low temperatures. At 3.8 K, there is a weak anomaly in $\rho(T)$ that coincides with T_N found in C/T and χ. Together, the data in Fig. 1 establish $CeRhIn_5$ as a new heavy-fermion compound in which an antiferromagnetic instability develops in a crystal-field doublet ground state. The small ordered moment below T_N implies the presence of relatively strong Kondo spin compensation, and, consequently, would place $CeRhIn_5$ close to the magnetic-nonmagnetic boundary in the Doniach model.

From this perspective, applying pressure to $CeRhIn_5$ should drive T_N toward $T = 0$ and perhaps induce superconductivity. Figure 2 shows the resistivity measured at various applied pressures generated in a clamp-type cell [15] with Flourinert-75 as the pressure medium. The most prominent feature of these data is the evolution of a well-defined maximum in the resistivity at a temperature T_{max}, which, as shown in the inset, initially moves to lower temperatures with increasing pressure, reaches a minimum near 12–13 kbar, and then increases at the highest pressures. This negative $\partial T_{max}/\partial P$ is highly unusual, whereas the positive $\partial T_{max}/\partial P \approx 1$ K/kbar above 15 kbar is typical of Ce-based heavy-fermion materials [2]. Pressure studies of both Ce impurities in nonmagnetic hosts [16] and of Ce-based heavy-fermion compounds [2] point to $\partial|J|/\partial P > 0$. Because the Kondo temperature $T_K \propto \exp(-1/|J|)$, $\partial T_K/\partial P > 0$ and T_{max} should increase with pressure. The results of Fig. 2 suggest, then, either that $\partial|J|/\partial P < 0$ or that another mechanism is competing with the Kondo effect to produce the initial decrease of T_{max} with pressure.

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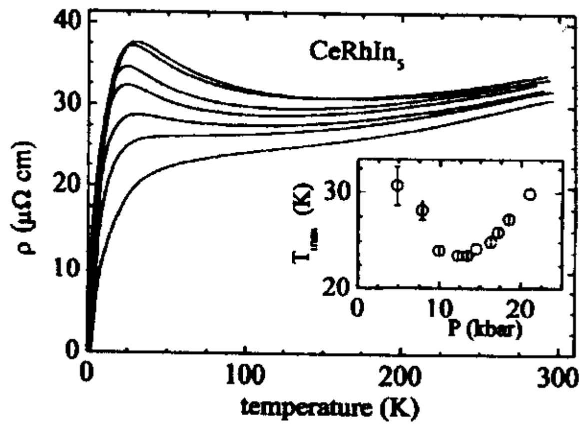

FIG. 2. Temperature dependence of the electrical resistivity of $CeRhIn_5$ at representative applied pressures. Data shown correspond to pressures of 0.001, 4.8, 7.9, 12.2, 14.5, 18.5, and 21.0 kbar and are associated, respectively, with curves of increasing resistivity at 50 K. The inset is a plot of the pressure dependence of the temperature T_{max} where the resistivity is a maximum.

In this regard, measurements of $\chi(T,P)$ in a SQUID magnetometer [17] to 5 kbar show [inset of Fig. 1(b)] that the temperature $T_{\chi m}$ at which $\chi(T)$ is a maximum decreases approximately linearly with increasing pressure and extrapolates to $T = 0$ at 13 ± 4 kbar, a value near the pressure at which $T_{max}(P)$ is a minimum.

The low-temperature response of the resistivity to pressure, plotted in Fig. 3, is remarkable. There is a large, reversible increase in the low-temperature resistivity; at 2.5 K, $\rho(T,P)$ increases from less than 1 $\mu\Omega$ cm at $P =$ 1 bar to approximately 12 $\mu\Omega$ cm at $P = 21$ kbar. Accompanying this increase in the magnitude of $\rho(T,P)$ is a systematic change in its temperature dependence. The Néel temperature increases weakly with pressure for $P \leq 14.5$ kbar, above which there is no resistive signature for T_N. At 16.3 kbar and higher, there is a transition beginning near 2 K to a zero-resistance state; the transition width is initially broad and sharpens to less than 0.05 K at 21 kbar where the onset of superconductivity is at 2.17 K. Experiments to 17 kbar on a second single crystal, grown independently, reproduce the appearance of superconductivity; simultaneous measurements of the ac susceptibility of this second crystal and a piece of Sn with the same geometry show nearly identical diamagnetic responses at their respective superconducting transition temperatures. By this measure, the pressure-induced superconductivity in $CeRhIn_5$ is a bulk effect. Upper critical field measurements in fields to 10 T on the first crystal give $-\partial H_{c2}/\partial T|_{T_c} \geq 14$ T/K at $P = 21$ kbar.

Preliminary specific heat measurements [18] at 20.8 kbar confirm bulk heavy-fermion superconductivity but also show a relatively large anomaly at 2.75 K that clearly is not due to superconductivity of free In. Whatever the origin of this anomaly, it also is reflected in $\rho(T)$ at 21 kbar and possibly at pressures as low as 9.9 kbar, where there is a very small change in slope near 3 K.

The T-P phase diagram constructed from these data is plotted in Fig. 4. The abrupt loss of a signature for T_N, the sudden appearance of superconductivity, and the rapid sharpening of the superconducting transition width suggest a first-order-like transition at a critical pressure P_c between 14.5 and 16.3 kbar that is independent of the anomaly, labeled $T_?$, found in resistivity and specific heat data above 9 kbar. This phase diagram is unlike any previously reported for Ce heavy-fermion compounds [3–6] and is apparently contrary to that expected from the Doniach model.

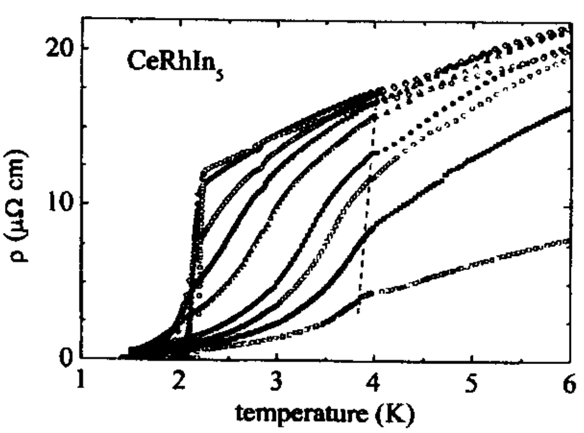

FIG. 3. Low-temperature response of $CeRhIn_5$ to applied pressures. With increasing resistivity at 2.3 K, the curves correspond to applied pressures of 0.001, 7.9, 9.9, 12.2, 14.5, 16.3, 17.2, 18.5, and 21.0 kbar. The diagonal dashed line is a guide to the eye.

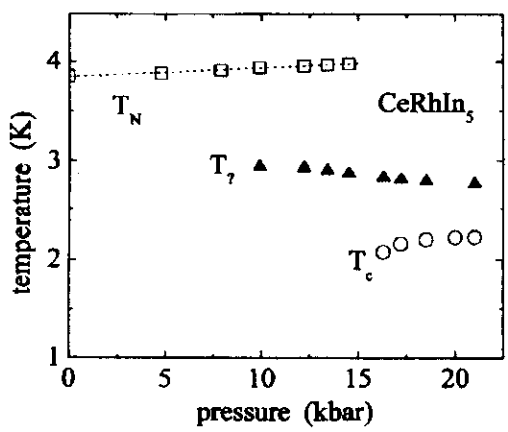

FIG. 4. Temperature-pressure phase diagram for $CeRhIn_5$ constructed from data shown in Fig. 3. Squares and circles give the pressure dependence of the Néel and onset superconducting transition temperatures, respectively. Triangles correspond to resistance features of unknown origin; see text. The dashed line has a slope of 9 mK/kbar.

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The first-order-like transition is approximately coincident with the change in sign of the pressure dependence of T_{max} and the pressure at which $T_{\chi m}$ extrapolates to $T = 0$.

A qualitative interpretation of the ambient- and high-pressure properties of $CeRhIn_5$ is suggested by its quasi-2D crystal structure. The smaller a_0 of $CeRhIn_5$ compared to that of bulk $CeIn_3$ implies that the $CeIn_3$ building blocks are under roughly 14 kbar chemical pressure. At 14 kbar, the Néel temperature of 3D $CeIn_3$ is 8 K [5]. It, therefore, is reasonable to associate the maximum at 7.5 K in the susceptibility of $CeRhIn_5$ with the development of 2D spin correlations in its $CeIn_3$ layers [19]. This association leads to the following implications. (1) $T_{\chi m}$ should decrease to $T = 0$ at a pressure of 11 kbar, which is the applied pressure required, in addition to the 14 kbar chemical pressure, to compress the $CeIn_3$ layers to an equivalent of 25 kbar (where T_N of 3D $CeIn_3$ goes to zero). This value of 11 kbar agrees with our experimental estimate of 13 ± 4 kbar. (2) It is not a coincidence that T_{max} is a minimum near the pressure where $T_{\chi m}$ extrapolates to $T = 0$. As $T_{\chi m}$ moves toward $T = 0$, 2D antiferromagnetic fluctuations are enhanced at low temperatures, leading to an increase in the low-temperature resistivity and the development of a maximum in $\rho(T, P)$ that initially follows $T_{\chi m}(P)$. There should be a crossover from antiferromagnetic- to quantum-fluctuation dominated scattering as $T_{\chi m} \to 0$. (3) Néel order arises from a weak, but strongly pressure dependent, interlayer exchange $|J|_{\perp}$. The competition between increasing $|J|_{\perp}$ and decreasing $\langle \mathbf{S} \cdot \mathbf{s} \rangle$ results in a critical point at $14.9 < P_c < 16.3$ kbar. From the preceding discussion, we would expect P_c to be somewhat lower. At present, we do not know if this difference of a few kilobars is significant or simply is within uncertainty in estimating where $T_{\chi m} \to 0$.

In summary, $CeRhIn_5$ is a new heavy-fermion antiferromagnet that becomes a bulk heavy-fermion superconductor at pressures above 16.3 kbar, with an initial transition temperature nearly 10 times higher than the maximum T_c of bulk $CeIn_3$ [20]. The evolution from antiferromagnetic to superconducting states is markedly different from all previous examples. $CeRhIn_5$ emphasizes the need to consider dimensionality effects in a more realistic generalization of the widely accepted views that follow from Doniach's Kondo-necklace model.

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A New Heavy-Fermion Superconducting Prototype $CeIrIn_5$: Relative of the Cuprates?

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Abstract: The new heavy-fermion superconductor $CeIrIn_5$ forms in a quasi-2-dimensional structure and exhibits physical properties analogous to those in the cuprate superconductors. It suggests a path of investigation for bridging our understanding of unconventional normal and superconducting state phenomena in these classes of materials.

Of the vast number of metallic compounds, only a small fraction enter a superconducting state at low temperatures, and of this small number, an even smaller fraction develop superconductivity out of a normal state in which electronic correlations produce orders-of-magnitude enhancement of the conduction electrons' effective mass *(1)*. This subset of materials, known as heavy-fermion superconductors, has been an influential area of research in condensed matter physics since its first member $CeCu_2Si_2$ was discovered *(2)* in 1979. Unlike all previously known superconductors, the presence of a magnetic ion (in this case Ce) was essential for superconductivity and the temperature dependence of physical properties below the superconducting transition temperature T_c was inconsistent with the well-established Bardeen-Cooper-Schrieffer theory of superconductivity. Over the past two decades, five uranium-based compounds at atmospheric pressure and four cerium-based compounds under applied pressure have been added to this class *(3)*. Interestingly, all but one of the pressure-induced heavy-fermion superconductors and one of the U-based superconductors form in the same $ThCr_2Si_2$ tetragonal structure as $CeCu_2Si_2$, suggesting that this structure type is particularly favorable for heavy-fermion superconductivity.

Experimental and theoretical study of the superconductivity in these materials has formed a substantial basis for understanding more broadly classes of unconventional superconductors, including the high-T_c cuprates, in which the electron-pairing interaction responsible for superconductivity may be mediated by spin fluctuations *(1)*. In spite of progress, the heavy-fermion problem and heavy-fermion superconductivity in particular remain challenges to experiment and theory *(4)*. Though heavy-fermion behavior has been found in several structure types, it appears that, like conventional BCS superconductivity, heavy-fermion superconductivity may be favored by particular crystallographic structures. Because of the limited number of examples, $CeCu_2Si_2$ being still the only Ce-based one at atmospheric pressure, we know ver little about relationships that should exist between the structure and properties of these materials. Any predictive understanding of how superconductivity can emerge in the highly correlated ground state has to be able to explain why it appears in one crystal structure and not another. This makes the discovery of a new prototype structure for heavy-fermion superconductivity *(5)* of special interest. We report such a structure here in which a new Ce-based heavy-fermion superconductor is discovered at ambient pressure. Unlike $CeCu_2Si_2$, this new compound grows easily and reproducibly as large, very pure single crystals, opening the possibility for unprecedented study.

$CeIrIn_5$ is a member of this new family that forms as $R_nT_mIn_{3n+2m}$, where n=1 or 2, m=1, R= La through Gd (except Eu), and T is a transition metal. All members grow readily as cm-sized, plate-like single crystals out of an In-rich flux. Crystals were obtained by combining stoichiometric amounts of Ce and T with excess In in an alumina crucible, encapsulating the crucible in an evacuated quartz ampoule, heating to 1100 C, and slowing cooling to 600 C. At this temperature, the ampoule was removed from the oven and the excess In removed by centrifugation. Powder x-ray patterns obtained on crushed single crystals show that $CeIrIn_5$ crystallizes in the tetragonal $HoCoGa_5$ structure type, with a=4.668(1) Å and c=7.515(2) Å *(6)*. Compounds for which n=1 can be viewed as alternating layers of $CeIn_3$ and TIn_2 stacked sequentially along the tetragonal c-axis and for n=2 form as bilayers of $CeIn_3$ separated by a single layer of TIn_2. A large resistivity ratio $\rho(300\ K)/\rho(2\ K)$= 50-80 for the Ce-materials attests, in part, to the high quality of the crystals as does the observation of resolution-limited Laue diffraction and NQR spectra. The hallmark of a heavy-fermion system is the magnitude of its electronic coefficient of specific heat γ, which is a measure of the effective mass enhancement of conduction electrons produced by electronic correlations *(1)*. All of the Ce-based members of this new family exhibit heavy-fermion behavior as indicated by their large

Sommerfeld specific heat coefficients γ, which range from ≈400 mJ/mole-Ce K^2 for antiferromagnetic $CeRhIn_5$ and Ce_2RhIn_8 to ≈700 mJ/mole-Ce K^2 for $CeIrIn_5$ and Ce_2IrIn_8. In contrast, the La-analogues, which do not contain an f-electron, are Pauli paramagnets with coefficients γ of about 5 mJ/mole-K^2 that are typical of simple metals. Additional details of the preparation and characterization of this family will be given elsewhere.

The overall temperature dependence of the resistivity ρ and magnetic susceptibility χ of $CeIrIn_5$ is shown in Fig. 1. The magnetic susceptibility is anisotropic, with χ larger by nearly a factor of two at low temperatures for a magnetic field applied along the tetragonal c-axis. Plots of 1/χ are linear in temperature for T≥ 200K. From the linear regime, we find a paramagnetic Curie temperature Θ_P, which is +12.5 K (–67.4 K) for a magnetic field of 1 kOe applied parallel (perpendicular) to the c-axis. A polycrystalline average of the high temperature data gives an effective moment μ_{eff} =2.28 μ_B that is reduced somewhat from the free-ion moment of Ce^{3+}, 2.54 μ_B, due to the presence of crystalline electric fields that lift the degeneracy of the J=5/2 Hund's-rules multiplet. Characteristic of Ce-based heavy-fermion compounds, the resistivity passes through a maximum at low temperatures that typically is attributed to the cross-over from strong, incoherent scattering of electrons at high temperatures to the development of strongly correlated bands at low temperatures. The magnitude and temperature dependence of ρ are similar to those of the heavy-fermion antiferromagnet $CeIn_3$ *(7)*.

Thermodynamic and transport properties of $CeIrIn_5$ at low temperatures are summarized in Fig. 2. Above 0.4 K, the specific heat divided by temperature C/T≡ γ=720 mJ/mole-K^2 and is nearly temperature independent. At T_c=0.40 K, there is a jump in C/T and a prominent signature in ac susceptibility χ_{ac}. Comparing the magnitude of this χ_{ac} response to that of a piece of superconducting tin having a similar size and shape as the $CeIrIn_5$ sample, we estimate that the χ_{ac} signature corresponds to a change in susceptibility of $-(1\pm 0.1)/4\pi$, as expected for a bulk superconductor. From the average of measurements on three different crystals, the specific heat jump ΔC at T_c is equal to $(0.76 \pm 0.05)\gamma T_c$. This ratio $\Delta C(T_c)/\gamma T_c$ is comparable to that found in other heavy-fermion superconductors, such as $CeCu_2Si_2$ and UPt_3 *(4)*, and provides compelling evidence that superconductivity in $CeIrIn_5$ develops among the heavy quasiparticles. The specific heat data below T_c fit well to the sums of nuclear-Schottky, T^2 and T-linear contributions. (A nuclear Schottky term is expected due to the large nuclear quadrupole moments of Ir and In. *(8)*) The $C \propto T^2$ contribution suggests that the superconducting gap function goes to zero along certain portions of the Fermi surface *(9)*. The large T-linear term of 200±50 mJ/mole-K^2 indicates the presence of ungapped heavy quasiparticle states below T_c and provides further support for the existence of zeros in the gap. The temperature dependence of the thermal conductivity, which is insensitive to the nuclear Schottky, also is described well from T_c to 50 mK by the sum of linear and quadratic terms that are consistent with corresponding terms in the specific heat.

A peculiar aspect of the data in Fig. 2 is that the resistivity drops to zero, or at least to less than our instrumental resolution of 0.01μΩ-cm, at T_0=1.2 K without an obvious thermodynamic or magnetic signature. As shown in Fig. 3, measurements of the specific heat, ac susceptibility and electrical resistivity in magnetic fields applied parallel and perpendicular to the c-axis of $CeIrIn_5$ find that the anisotropic responses of T_c, determined by specific heat and ac susceptibility, and T_0, determined resistively, are identical. Within the scatter of data in Fig. 3, these results are reproduced in three independently-grown crystals. It, therefore, seems reasonable to conclude that both transitions are intrinsic and arise from a common underlying electronic structure that band structure calculations *(10)* and preliminary de Haas-van Alphen measurements *(11)* show to be quasi-2D. Though coming from a common electronic background, T_c and T_0 develop out of apparently dissimilar manifestations of the highly correlated normal state. Just above T_c, the large, nearly constant C/T is typical of a strongly correlated Landau Fermi liquid. However, in all crystals we have studied, the electrical resistivity varies as $\rho(T)-\rho(T_0) \propto T^n$, with $n=1.3\pm 0.05$, for $T_0 \leq T \leq 5$ K. This is not the quadratic temperature dependence expected of a Landau Fermi liquid, and it persists from ~5 K to 60 mK when a magnetic field is applied to suppress T_0. Similar power-law variations in the electrical resistivity are found in the cuprates *(12)* and in heavy-fermion systems tuned by pressure to a magnetic/superconducting boundary *(13)*. One suggestion for its origin is the scattering of conduction electrons by antiferromagnetic spin fluctuations whose characteristic wave-vector connects portions of the Fermi surface *(14)*.

Tuning the hybridization between the 4f and ligand electrons by substituting Rh for Ir induces small-moment, incommensurate antiferromagnetism *(15)* in the end member $CeRhIn_5$, which has a Néel temperature of 3.8 K. Magnetization and nuclear quadrupole-resonance studies indicate substantial similarities *(15,16)* of the magnetism in $CeRhIn_5$ with that found in La_2CuO_4 from which high-T_c superconductivity develops with hole doping. When Rh is added substitutionally into $CeIrIn_5$, T_0 decreases and T_c increases, which it also does when $CeIrIn_5$ is subjected to hydrostatic pressure. For x=0.25 and 0.5 in $CeIr_{1-x}Rh_xIn_5$, a small, less than $0.01(-1/4\pi)$, diamagnetic response appears in χ_{ac} at T_0 that is followed by a much larger diamagnetic signal at T_c. Midway between the end-points, $CeIr_{0.5}Rh_{0.5}In_5$, T_c more than doubles to 0.86K, $\Delta C(T_c)/\gamma T_c$, γ, and C(T) below T_c are virtually unchanged relative to $CeIrIn_5$, and T_0 decreases to 1.0 K. These trends with isoelectronic substitution, which tunes f-d hybridization, are striking similar to those observed in the cuprates *(17)* with hole doping, which tunes band filling.

An analogy with the cuprates suggests a possible interpretation for the origin of T_0. As a function of temperature and doping in the cuprates, a decrease in, for example, spin susceptibility and electrical resistivity defines a boundary in the T-x phase diagram that marks a cross-over from a paramagnetic to a pseudo-gap state out of which superconductivity develops *(18)*. Many experiments are consistent with the formation of local (static or dynamic) spin/charge pair correlations without global phase coherence, concepts for which there is growing theoretical support *(17)*. In the cuprates, this boundary is smeared by inhomogeneity introduced by hole doping. Without such inhomogeneity, one might expect this boundary to become a sharply defined phase transition with signatures similar to those found in our case at T_0. We, however, would expect a specific heat feature at T_0, which is not seen. Quite plausibly this feature could be small compared to the large, heavy-electron specific heat out of which it develops and not detected within our experimental resolution. Again drawing on the cuprates, the bulk transition at T_c might be interpreted as the (Bose) condensation of electron pairs 'preformed' at T_0 or as the temperature at which Josephson coupling among pairs produces global phase coherence throughout the sample.

In summary, the new superconductor $CeIrIn_5$ suggests that the physics of heavy-fermion materials is much richer than previously imagined and that, when crystallizing in a quasi-2D structure, shows features analogous to those in the cuprate superconductors *(19)*. The search for yet other examples of 2-D structure types that form with Ce appears to be a fruitful path of investigation as does additional study of $CeIrIn_5$, which may bridge our understanding of more nearly 3-D heavy-fermion metals and the copper oxides.

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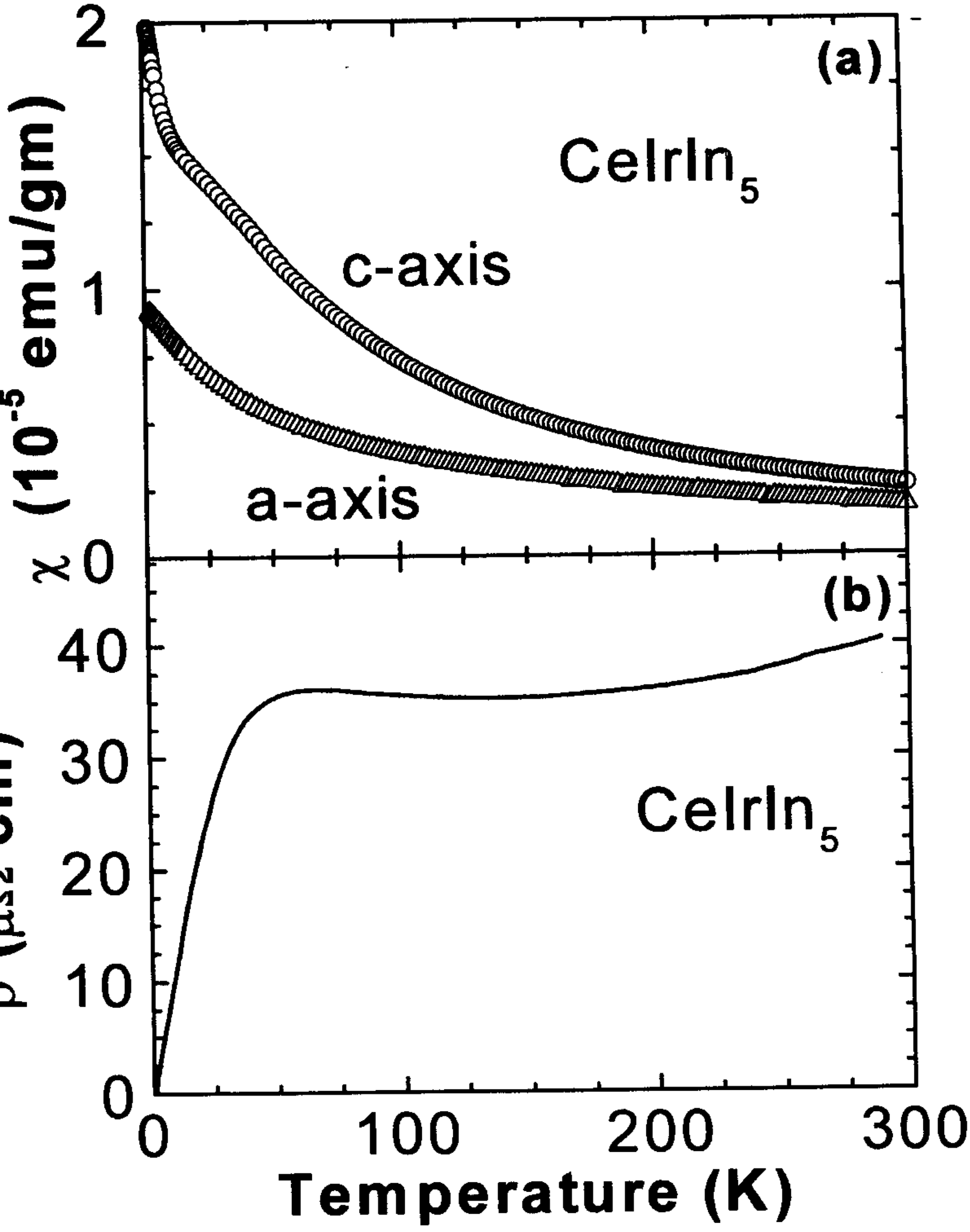
Figure Legends:

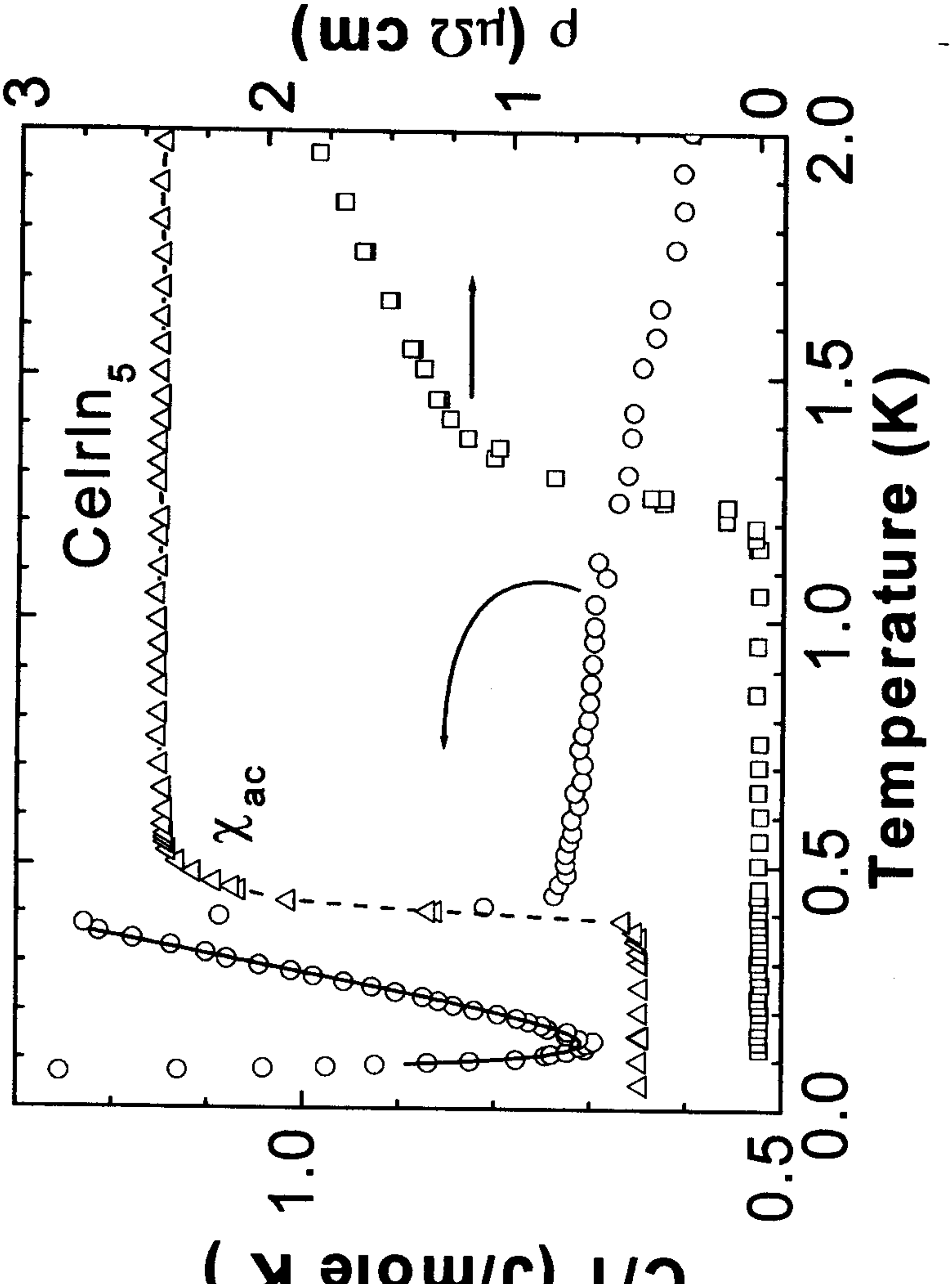
Fig. 1. (a). Magnetic susceptibility χ as a function of temperature for a 1-kOe field applied parallel to the c- (circles) and a-axis (triangles) of $CeIrIn_5$. Measurements were made in a Quantum Design Superconducting Quantum Interference Device magnetometer. (b). Electrical resistivity ρ versus temperature measured with a 4-lead ac resistance bridge.

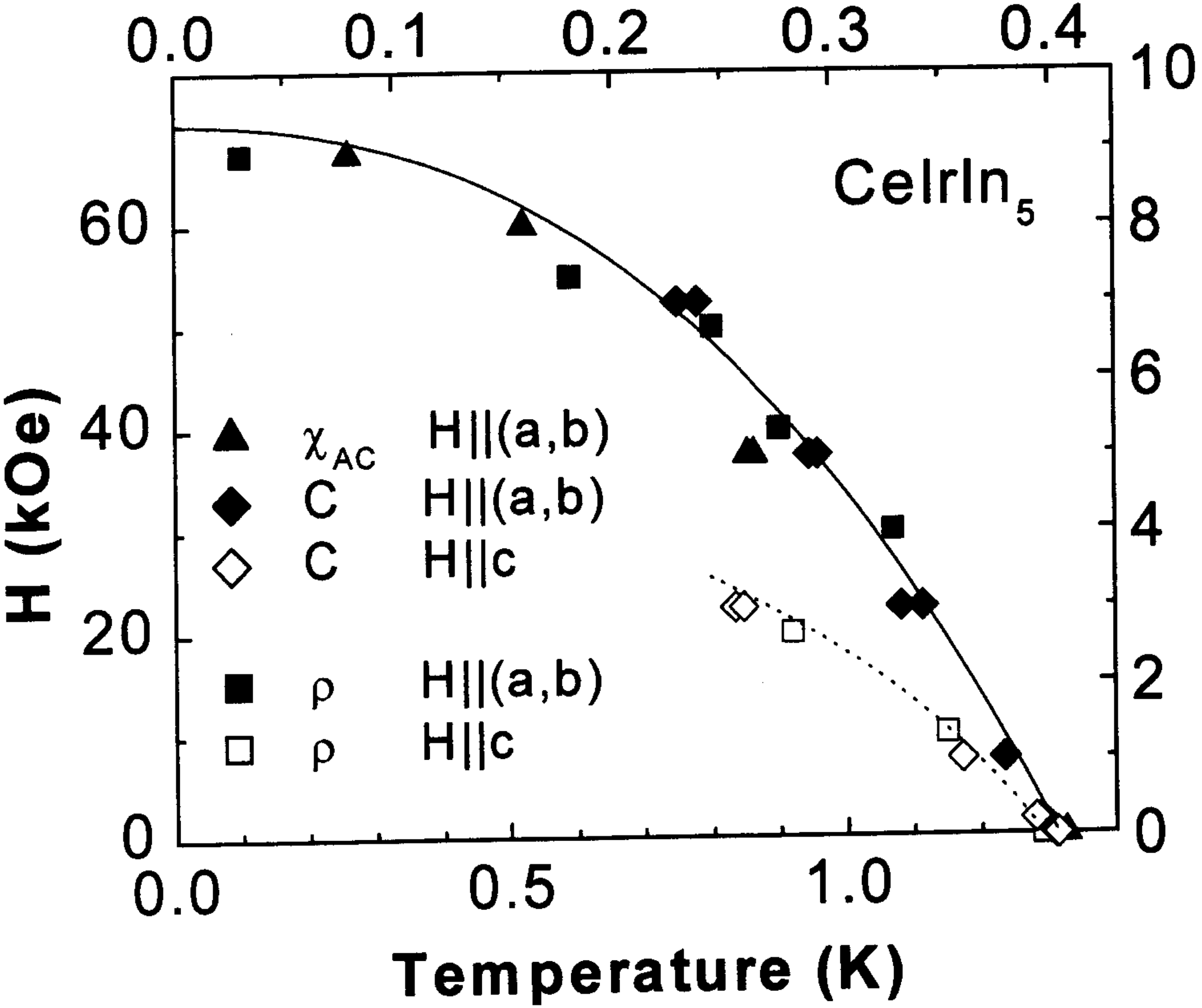
Fig. 2. Specific heat divided by temperature C/T (circles, left ordinate), ac magnetic susceptibility χ_{ac} (triangles, arbitrary units) and electrical resistivity ρ (squares, right ordinate) of $CeIrIn_5$ as functions of temperature. The solid line is a fit to C/T data below T_c and is given by $C/T = 0.00029T^3 + 2.54T + 0.234$. The dashed line through χ_{ac} (T) is a guide to the eye.

Fig. 3. Magnetic field H versus temperature phase diagram constructed from specific heat C, ac magnetic susceptibility χ_{ac} and electrical resistivity ρ measurements on a $CeIrIn_5$ crystal with a magnetic field applied parallel and perpendicular to the c-axis. Transition midpoints are used to define the diagram. The left ordinate and bottom abscissa correspond to resistivity data. The right ordinate and top abscissa correspond to specific heat and χ_{ac} data. Open symbols are for H parallel to the c-axis. Solid symbols are for H perpendicular to the c-axis. Note the difference in field and temperature scales and that the anisotropy in these data are identical irrespective of the measurement technique. Solid and dashed lines are guides to the eye.



(a)
CeIrIn5
c-axis
a-axis
χ (10-5 emu/gm)
2
1
0
(b)
CeIrIn5
ρ (μΩ cm)
40
30
20
10
0
0
100
200
300
Temperature (K)


CeIrIn5
χac
Temperature (K)
C/T (J/mole K²)
ρ (μΩ cm)
0.0
0.5
1.0
1.5
2.0
0
1
2
3



0.0
0.1
0.2
0.3
0.4
10
8
6
4
2
0
CeIrIn5
60
40
20
0
H (kOe)
χAC H||(a,b)
C H||(a,b)
C H||c
ρ H||(a,b)
ρ H||c
0.0
0.5
1.0
Temperature (K)

Raising T_c in magnetically mediated superconductors.

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Superconductivity is a field unusual in the correlation often evident between structure and properties: certain crystal structures or substructures favor superconductivity.[1] This is evident both in conventional and high-T_c materials, yet no good theoretical understanding of this exists. In particular, what underlies this relationship in the high T_c cuprates and heavy Fermion materials which border so closely on magnetically ordered phases is of essential interest both fundamentally and in the search for new materials.[2,3] We report here the discovery of a new quasi-2D heavy-Fermion superconductor $CeCoIn_5$ with the highest known ambient pressure T_c (2.3 K) and rationalize this finding with respect to the pressure induced low T_c of related 3D $CeIn_3$ and perhaps the high T_c of the layered cuprates.[3]

Superconductivity in correlated-electron materials presents a paradox. On one hand, it is a rather rare ground state; on the other, once found it is quite robust and often appears in structurally related materials. This paradox recently has been termed a 'quantum protectorate:' emergent low-energy physical phenomena are regulated by higher organizing principles without regard for microscopic details.[4] In practice what this means for the materials physicist is that the unexpected observation of superconductivity in a new structure type is highly prized because once found, other examples are probably close at hand. For example, fully half of the known heavy Fermion superconductors crystallize in the tetragonal $ThCr_2Si_2$ structure, which is also the structure type of the La_2CuO_4 family of high T_c copper-oxide superconductors.[5] Here, we report the discovery of another such family: superconductivity in $CeCoIn_5$ as the highest-T_c representative of a family of new Ce-based heavy Fermion superconductors.

The heavy Fermion superconductors are a class of materials in which superconductivity emerges out of a normal state in which electronic correlations, due to the presence of an appropriate magnetic ion – in this case Ce, enhance the effective mass m* of conduction electrons by several orders of magnitude.[6] In the more than twenty years since the first member of this class of materials was discovered ($CeCu_2Si_2$),[7] only two other Ce-based materials have been found which unambiguously show superconductivity at ambient pressure: $CeIrIn_5$,[8] and now, $CeCoIn_5$. These earlier examples exhibit rather complex phenomena and/or metallurgy, making their study challenging. The ground state of $CeCu_2Si_2$ can be either antiferromagnetic or superconducting depending on very small changes in unit-cell volume or composition;[9] $CeIrIn_5$ shows zero or very small resistivity near 1 K but does not produce a thermodynamic signature of superconductivity until 0.4 K.[8] Although the complexity of these materials is a direct manifestation of the richness of their physics, our fundamental understanding of the nature of heavy-Fermion superconductivity would profit substantially from the existence of a much simpler example. $CeCoIn_5$ promises to be such a material.

$CeCoIn_5$ forms in the tetragonal $HoCoGa_5$ crystal structure with lattice constants a=4.62 Å and c=7.56 Å.[10] The structure is built up of alternating layers of $CeIn_3$ (a known heavy Fermion antiferromagnet in which superconductivity can be induced with applied pressure[3]) and '$CoIn_2$.'[11] As will be discussed below, the creation of a lower dimensional form of $CeIn_3$ is central to the large number of relatively high-transition-temperature superconductors that are observed in the $CeMIn_5$ family (M=Co, Rh,[12] Ir[8]). Single crystals of $CeCoIn_5$ were synthesized from an In flux by combining stoichiometric amounts of Ce and Co with excess In in an alumina crucible and encapsulating the crucible in an evacuated quartz ampoule. Because of the deep eutectic formed between Ce and Co and the strong phase stability of $CeIn_3$, growth of $CeCoIn_5$ appears to be optimized in dilute (3 at% Ce) melts with a two-stage cooling process – an initial rapid cooling from 1150 C, where the molten material is homogenized, to 750 C and then a slower cool to 450 C, at which point the excess flux is removed with a centrifuge. The resultant crystals are well-separated, faceted platelets with characteristic dimensions 3 mm x 3 mm x 0.1 mm.

Figure 1 shows the magnetic susceptibility χ and electrical resistivity ρ of $CeCoIn_5$. The magnetic susceptibility is anisotropic, with χ larger for a magnetic field applied along the tetragonal c axis. The rapid upturn at low temperature for $\chi \parallel c$ is intrinsic and is a common feature of the $CeMIn_5$ materials.[8,12] At high temperatures (T>200), χ^{-1} is linear in temperature and a paramagnetic Weiss temperature of –54 K (-83 K) is found when the magnetic field is applied parallel (perpendicular) to c. The effective moment obtained from a polycrystalline average of these data is 2.59 μ_B, consistent with the free-ion expectation for Ce^{+3} (2.54 μ_B). The resistivity of $CeCoIn_5$ is typical of heavy-Fermion materials: weak temperature dependence above a characteristic temperature, here ~ 30 K, followed by a rapid decrease at lower temperature. This behavior is generally attributed to a crossover from strong, incoherent scattering of electrons at high temperature followed by the development of strongly correlated Bloch states at low temperature. Below ~ 20 K the resistivity of $CeCoIn_5$ is nearly linear in temperature, a functionality found commonly in magnetically mediated superconductors and frequently associated with proximity to a quantum critical point.[3] The low value of ρ at 2.5 K (3 $\mu\Omega$cm) indicates minimal defect scattering. The inset of

Figure 1 reveals clear evidence for superconductivity: zero resistance and full-shielding diamagnetism is achieved at 2.3 K.

The specific heat divided by temperature for $CeCoIn_5$ is shown in Figure 2. For T>2.5 K, the large value of C/T=290 mJ/molK^2 indicates substantial mass renormalization ($C/T = \gamma \propto m^*$). At T_c a surprisingly large jump in heat capacity is observed. The inferred value of $\Delta C/\gamma T_c$=4.5 which suggests unprecedented strong coupling. (The expectation for a weak-coupling superconductor is $\Delta C/\gamma T_c$=1.43.[13]) However, application of a magnetic field of 50 kOe suppresses superconductivity and reveals that entropy is conserved between the normal and superconducting states by 2.3 K because of the continued increase of C/T with decreasing temperature. Because there is presently no theory of superconductivity that directly accounts for a temperature dependent γ below T_c, this effect calls into question simple estimates of whether $CeCoIn_5$ is a weak- or strong-coupling superconductor. The enhanced normal-state γ (~ 1J/molK^2) in 50 kOe is also clear evidence that $CeCoIn_5$ is indeed a heavy Fermion material. The temperature dependence of C below T_c is clearly non-exponential, suggesting the presence of nodes in the superconducting gap. (Power law fits of the form $C/T = \gamma_0 + aT^x$ yield x~2, with the precise value of x depending somewhat on the range of temperatures fitted.) That the gap has nodes is also indicated by the finite value of C/T that is observed in the superconducting state as T approaches zero. Both the size of the heat capacity jump at T_c and the temperature dependence of C below T_c are reminiscent of UBe_{13}, another heavy Fermion superconductor.[14]

Figure 3 presents an H-T phase diagram for $CeCoIn_5$. For both heat capacity and resistivity measurements, the magnetic field is applied parallel to the c axis. Because of the large ΔC at the superconducting transition, one might suspect that the observed superconductivity is parasitic to a magnetic phase. However, the onset of zero resistance tracks the heat capacity feature identically as a function of field, and the observed transitions remain quite sharp at the highest fields, arguing against a co-existing magnetic phase. Additional resistivity measurements with magnetic field applied perpendicular to the c axis reveal an upper-critical-field anisotropy of at least a factor of two.

$CeCoIn_5$ appears to be a rather simple realization of heavy Fermion superconductivity with T_c=2.3 K. But, why should T_c in this material be over a factor of two higher than that of $CeCu_2Si_2$ or $CeIrIn_5$? Furthermore, how does one rationalize the existence of three heavy-Fermion superconductors ($CeRhIn_5$, $CeIrIn_5$, and $CeCoIn_5$) in the same structure type?

Recent theoretical arguments suggest three important ingredients in achieving high transition temperatures in magnetically mediated superconductors:[3,15] 1) T_{sf}, the characteristic temperature for spin fluctuations, is high; 2) the carrier density must be tunable in the vicinity of a magnetic critical point; and 3) the electronic structure is quasi-2D rather than 3D. $CeCoIn_5$, at least in a relative sense, seems to satisfy all three requirements. In a highly correlated electron system, a quasi-2D pairing interaction favors d-wave pairing which can minimize electron-electron Coulomb repulsion.[15] For an isotropic pairing interaction the Coulomb repulsion cannot be minimized in this way, suggesting why the pressure-induced T_c of $CeIn_3$ (~100 mK)[3] is so much lower than the 2.3-K T_c of layered $CeCoIn_5$. Although the intrinsic electronic anisotropy in the $CeMIn_5$ materials remains to be determined definitively (i.e., via inelastic neutron scattering), it seems intuitive that structural layering produces a material that is less 3D than cubic $CeIn_3$. Preliminary de Haas van Alphen and nuclear quadrupole resonance measurements are beginning to confirm this intuition.[16,17] The anisotropy of H_{c2} in $CeCoIn_5$ provides additional evidence for electronic anisotropy. That $CeRhIn_5$ can be tuned with pressure from an antiferromagnetic ground state to a superconducting ground state (T_c=2.17 K) is the best evidence for a tunable carrier density.[12] It is seemingly fortuitous that $CeIrIn_5$ and $CeCoIn_5$ naturally produce carrier densities which yield ambient-pressure superconducting ground states. Lastly, one has the optimization of T_{sf}. An estimate of T_{sf} comes from γ, which typically is assumed to scale as $1/T_{sf}$.[6] Among the $CeMIn_5$ family, γ at T_c increases monotonically from 290 to 400 to 750 $mJ/molK^2$ in the sequence M=Co, Rh, Ir, and T_c decreases as expected in the same sequence. Thus one seems to have found at least a local optimization for magnetically mediated superconductivity in $CeCoIn_5$ intrinsically, whereas tuning with pressure is required for $CeRhIn_5$ to reach a comparable T_c. Finally, we conclude on a speculative note. Another common empirical observation is that in related structural families of superconductors, members with the highest T_c often display unstable superconductivity (e.g., the series Nb_3Sn, Nb_3Ge, Nb_3Si).[18] That the properties of $CeCoIn_5$ are so simple perhaps suggests that even higher T_c members of this family remain to be found.

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Acknowledgements
We thank A.V. Balatsky, L.P. Gor'kov, M.J Graf, and G.G. Lonzarich for valuable conversations. This work was supported in part by the NSF and FAPESP, and was performed under the auspices of the U.S. Dept. of Energy.

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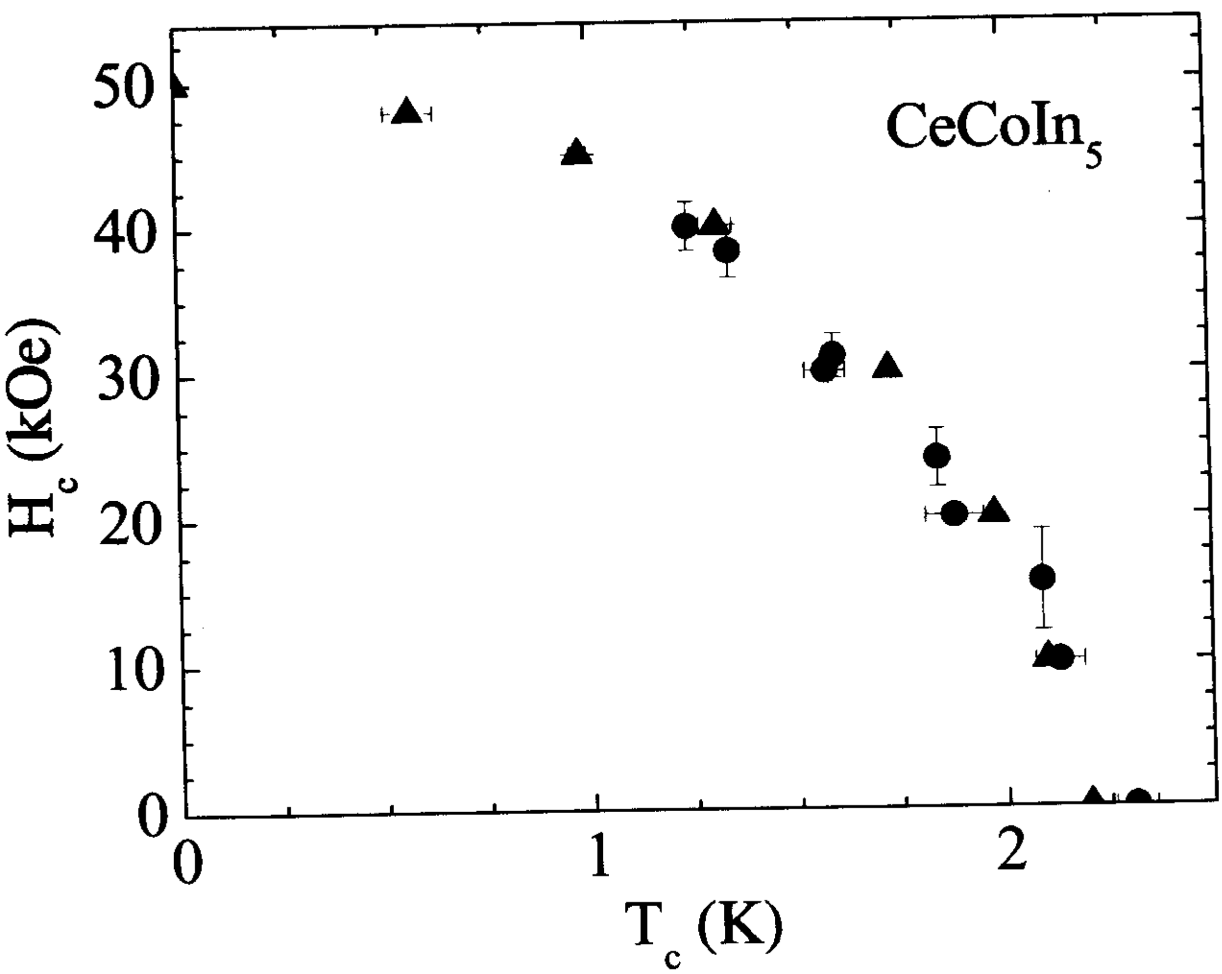
Figure Captions

Figure 1. Magnetic susceptibility and electrical resistivity of $CeCoIn_5$. Susceptibility is measured in a 1-kOe field applied parallel (circles) or perpendicular (squares) to the c axis of $CeCoIn_5$ using a SQUID magnetometer. The inset shows zero-field-cooled magnetic susceptibility (circles) as a fraction of $1/4\pi$ measured in 10 Oe and resisitivity (triangles) in the vicinity of the superconducting transition.

Figure 2. Specific heat divided by temperature versus temperature for $CeCoIn_5$. For both the zero-field (open squares) and 50-kOe (solid circles)

data, a nuclear Schottky contribution, due to the large nuclear quadrupole moment of In, has been subtracted. The inset shows the entropy recovered as a function of temperature in the superconducting (open squares) and field-induced normal (solid circles) states.

Figure 3. Magnetic field-temperature phase diagram of $CeCoIn_5$. The triangles correspond to specific heat measurements, and the circles were determined by electrical resistivity. In both cases magnetic field was applied along the c axis of the sample. For comparison, with a field of 90-kOe applied perpendicular to the c axis, T_c=1.3 K.



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