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Interplay of magnetism, structure and superconductivity in heavy-fermion systems $CeMIn_5$ and $PuMGa_5$

J.D. Thompson [a,*], M. Nicklas [a,b], V.A. Sidorov [a,c], E.D. Bauer [a],
R. Movshovich [a], N.J. Curro [a], J.L. Sarrao [a]

[a] Los Alamos National Laboratory, Condensed Matter and Thermal Physics, MS K764, Los Alamos, NM 87545, USA
[b] Max-Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany
[c] Institute for High Pressure Physics, 142190 Troitsk, Moscow Region, Russia

Abstract

Experiments that probe the normal and superconducting states of the heavy-fermion compounds $CeMIn_5$, M═Co, Rh, Ir, consistently point to the dependence of superconductivity on magnetism and, consequently, to the conclusion that soft magnetic excitations produce Cooper pairing. A more limited number of experiments on the isostructural superconductors $PuMGa_5$, M═Co, Rh, also are consistent with a magnetic pairing mechanism, but in these cases, the relationship between magnetism and superconductivity is less obvious. A connection between the Ce- and Pu-based families of unconventional superconductors is provided through a common dependence of their superconducting transition temperatures on lattice anisotropy and a weak structural distortion that may reflect a hybridization-induced change in magnetic fluctuations.
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1. Introduction

Magnetism and unconventional superconductivity are interdependent in strongly correlated electron systems, such as heavy-fermion materials. The relationship between these two broken symmetries is explicit in some examples, e.g., $CeIn_3$ [1], $CePd_2Si_2$ [1], $CeRh_2Si_2$ [2] and $CeCu_2Ge_2$ [3]. In each of these, antiferromagnetic order at atmospheric pressure evolves with applied pressure toward an antiferromagnetic quantum-critical point where unconventional superconductivity emerges. A ‘dome’ of superconductivity exists in a narrow range of pressures around the quantum-critical point, suggesting that soft antiferromagnetic fluctuations are most effective in mediating Cooper pairing in this narrow pressure window. A rather different relationship between antiferromagnetism and superconductivity appears in members of the $CeMIn_5$ family of heavy-fermion compounds in which M is a transition metal belonging to the Co-column, M═Co, Rh or Ir. $CeRhIn_5$ is the only member of this family to order antiferromagnetically. Its response to pressure [4,5] is similar to, but also distinctly different from, that of the examples listed above. Instead of superconductivity existing in a narrow pressure range, there is approximately an order of magnitude broader range of pressures in which long-ranged antiferromagnetic order coexists homogeneously with unconventional superconductivity [6,7], provided the Néel temperature $T_N(P)$ remains higher than the superconducting transition temperature $T_c(P)$. At pressures above about 1.9 GPa, T_c reaches a maximum of ≈ 2.3 K and exceeds $T_N(P)$; so far, there is no evidence for antiferromagnetic order below the $T_c(P)$ boundary for $P > 1.9$ GPa [5]. A similar relationship between magnetism and superconductivity also is found at atmospheric pressure in the substitutional series $CeRh_{1-x}Ir_xIn_5$: antiferromagnetic order and unconventional superconductivity coexist for $0.3 \leq x \leq 0.6$ where $T_N > T_c$; whereas, there is only superconductivity for $x > 0.6$ [8,9].

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[*] Corresponding author. Tel.: +1 505 667 6416; fax: +1 505 665 7652.
E-mail address: jdt@lanl.gov (J.D. Thompson).

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2. CeMIn$_5$ compounds

In CeIrIn$_5$ and CeCoIn$_5$, the relationship between some form of magnetism and superconductivity is less explicit. These members of the CeMIn$_5$ family are superconductors below $T_c = 0.4$ [10] and 2.3 K [11], respectively, at atmospheric pressure and display power-laws in physical properties below T_c [12], as expected for an unconventional superconductor with line nodes in the superconducting gap. Like all the examples mentioned above, they also exhibit a 'dome-shaped' evolution of T_c versus pressure [13,14], consistent with pressure tuning of the underlying pairing mechanism. Because antiferromagnetic fluctuations are known to favor d-wave superconductivity, which is strongly suggested from the four-fold modulation of the thermal conductivity [15] and specific heat [16] of CeCoIn$_5$ as a magnetic field is rotated in its tetragonal basal plane, a reasonable assumption is that the boson mediating Cooper pairing in CeCoIn$_5$ is antiferromagnetic fluctuations. However, no long ranged magnetic order has been found under ambient conditions or at pressures below 2.5 GPa or in fields less than 17 T.

Measurements of the electrical resistivity of CeCoIn$_5$ show that $\rho(T)$ is linear in temperature from just above T_c to nearly 20 K [13]. In this same temperature range, the inverse of the Hall coefficient, $-1/R_H$, also is T-linear [17], and, consequently, the cotangent of the Hall angle, $\cot(\theta_H) \equiv \rho_{xx}/R_H H$, is proportional to T^2 [17,18]. None of these temperature dependences are expected of a Landau Fermi liquid but imply that CeCoIn$_5$ may be near a magnetic quantum-critical point. The smaller unit cell volume of CeCoIn$_5$ compared to that of CeRhIn$_5$ is consistent with CeCoIn$_5$ being under a relative effective chemical pressure of about 1.6 GPa, which is smaller than but close to the pressure of ∼2.5 GPa where the Néel temperature of CeRhIn$_5$ extrapolates to $T = 0$. This, combined with scaling of the $T_c(P)$ phase diagrams for CeRhIn$_5$ and CeCoIn$_5$ when this chemical pressure is taken into account, led to the conclusion [13] that the nearby antiferromagnetism in CeCoIn$_5$ was at an inaccessible negative pressure of about −1.6 GPa. More recent measurements of the magnetic field dependence of the specific heat [19] and electrical resistivity [20] of CeCoIn$_5$ at atmospheric pressure have revealed non-Fermi liquid temperature dependences emanating from the $T = 0$ value of the upper critical magnetic field $H_{c2}(0)$. Increasing the field beyond $H_{c2}(0)$ recovers Fermi-liquid temperature dependences in both specific heat and resistivity. These observations prompted the suggestion [19] that the nearby antiferromagnetic order was 'hidden' by superconductivity but manifested itself in quantum-critical fluctuations precisely at the point where superconductivity was destroyed by a magnetic field. This remarkable coincidence requires that the magnetism and superconductivity be tuned simultaneously to $T = 0$ by the same value of the magnetic field. This coincidence is emphasized further by specific heat studies of CeCoIn$_{5-x}$Sn$_x$ in which small amounts of Sn linearly suppress superconductivity to $T_c = 0$ at a value of $x \approx 0.18$ [21]. For $x < 0.18$, non-Fermi-liquid temperature dependences also emerge at $H_{c2}(x,0)$, but with $C/T = $ constant, i.e., Fermi-liquid like, as the magnetic field in increased above $H_{c2}(0)$. These experiments imply either that the magnetic order and superconductivity have both the same field and x dependence or alternatively that it is a superconducting quantum-critical point [22] and not hidden antiferromagnetism that produces the non-Fermi-liquid behavior at $H_{c2}(0)$. The first possibility, though conceivable, seems extremely fortuitous, and the second scenario is highly unusual.

The superconducting properties of CeIrIn$_5$ also are consistent with d-wave pairing, but so far there is no evidence of nearby antiferromagnetic order. Superconductivity in this case emerges from a normal state in which C/T weakly increases as temperature approaches T_c, which could be consistent with Fermi-liquid behavior; however, in this temperature range, the electrical resistivity in not quadratic in temperature, as expected for a Landau Fermi liquid, but increases as T^{α}, where $\alpha \approx 1.25$ [10]. In a magnetic field much larger than $H_{c2}(0)$, C/T tends to approach a logarithmic divergence at low temperatures [23]. This systematic study of the field dependence of C/T suggests that there should be a field-induced quantum-critical point near $H = 25$ T, which is where a metamagnetic transition [24] in CeIrIn$_5$ also extrapolates to $T = 0$. This value of 25 T substantially exceeds $H_{c2}(0)$, which attains a maximum value of 1 T for H parallel to the tetragonal basal plane. Further, as shown in Fig. 1, pressure studies of CeRh$_{1-x}$Ir$_x$In$_5$ alloys find [5] that superconductivity in CeIrIn$_5$ becomes separated from the Rh-rich compositions with T_cs that track the pressure evolution of antiferromagnetism. For $x \leq 0.25$, the T–P phase diagrams scale onto each other with a simple shift of the pressure axis, $P_{\text{eff}} = P + P_{\text{Ir}}$, as if the introduction of Ir acts as an effective chemical pressure $P_{\text{Ir}}(\text{GPa}) \approx 10x^2$. For these samples, the maximum T_c as a function of pressure also exceeds 2 K, as it does in pure CeRhIn$_5$ and CeCoIn$_5$. This simple scaling, plotted in Fig. 2, breaks down for larger Ir concentrations, implying additional effects of the Ir substitutions for Rh.

In spite of the apparent separation of CeIrIn$_5$ from antiferromagnetic order and its lower superconducting transition temperature, the pressure response [14] of its superconductivity is similar to that of other members of the CeMIn$_5$ family. This similarity is shown in Fig. 3 where we plot $T_c(P)$ normalized by its maximum value as a function of pressure for CeRhIn$_5$, CeCoIn$_5$ and CeIrIn$_5$.This figure shows a common evolution of T_c for each CeMIn$_5$ compound, and from results plotted in Fig. 2, is valid as well for CeRh$_{1-x}$Ir$_x$In$_5$ alloys with $x \leq 0.25$. Together, Figs. 1–3 help establish a common underlying relationship between antiferromagnetism and superconductivity in this family of materials. However, this relationship would not be so apparent if nature had not provided CeRhIn$_5$ against which these other members could be compared. That is, if we only knew CeCoIn$_5$ and CeIrIn$_5$, experimental results for these materials would argue for some form of unconventional pairing mechanism, but its tie to magnetic fluctuations certainly would be more tenuous.

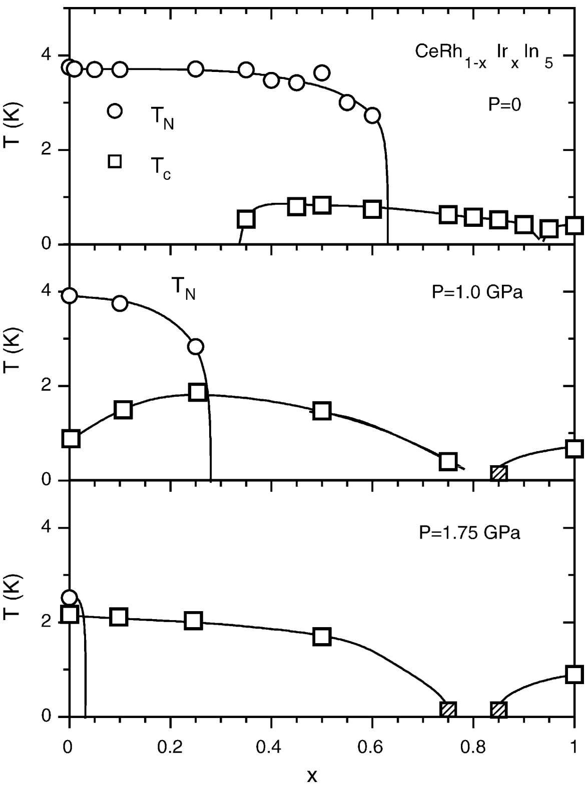

Fig. 1. Pressure-induced evolution of the temperature-doping phase diagram of CeRh$_{1-x}$Ir$_x$In$_5$. The isobaric phase diagrams at elevated pressures were constructed from a series of pressure measurements of the electrical resistivity and ac susceptibility on selected compositions. Details are given in Ref. [5]. Circles denote the onset of antiferromagnetic order and squares give the bulk superconducting transition temperature. Note that with increasing pressure, the end compound $x = 1$ becomes increasingly separated from antiferromagnetism.

3. PuMGa$_5$ superconductors

Another family of superconductors PuMGa$_5$, M=Co and Rh, crystallize in the same HoCoGa$_5$ structure as the CeMIn$_5$ compounds. The superconducting transition temperatures in the Pu-based materials, however, are substantially higher than in their Ce counterparts, exceeding 18.5 K in PuCoGa$_5$ [25] and reaching 8.7 K in PuRhGa$_5$ [26]. The transition temperatures in these 5f-electron materials far exceed those of any other correlated f-electron material and raise the possibility that superconductivity may be conventional. In both Pu-compounds, superconductivity develops out of a normal state in which the static magnetic susceptibility follows a Curie–Weiss dependence from room temperature to T_c with an effective moment of about $0.65\mu_B$, which is slightly reduced from the Hund's rule value of $0.84\mu_B$ for Pu^{3+} [25,26]. The absence of any evidence for a magnetic transition above 1 K from specific heat measurements of PuCoGa$_5$ [27] suggests that the magnetic 5f electrons of Pu may be involved in superconductivity. This conclusion is supported by the Ga-Knight shift of PuCoGa$_5$ that shows no local spin susceptibility below T_c and is consistent with spin-singlet pairing [28]. The most compelling evidence against a conventional electron–phonon pairing mechanism comes from spin-lattice relaxation measurements [28]. In addition to the lack of a Hebel–Schlicter peak in $1/T_1$ near T_c, $1/T_1$ follows the same T^3 dependence below T_c that is observed in CeMIn$_5$

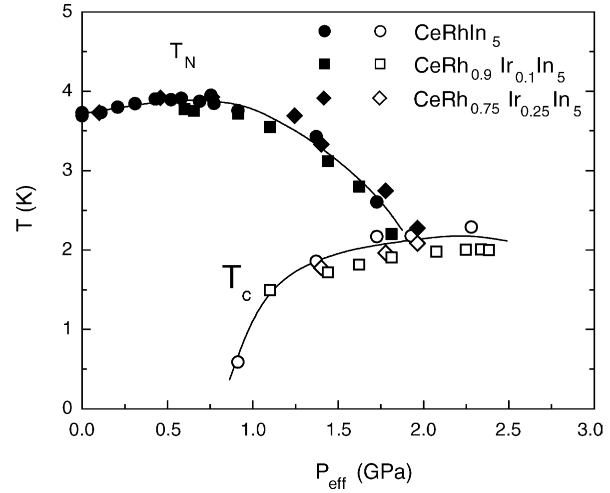

Fig. 2. Temperature–pressure phase diagram for CeRh$_{1-x}$Ir$_x$In$_5$ samples with $x = 0$, 0.1 and 0.25. The pressure axis P_{eff} is the sum of applied pressure and an effective chemical pressure P_{Ir} produced by the Ir substitutions. Values of P_{Ir} are 0, 0.1 and 0.6 GPa for $x = 0$, 0.1, and 0.25, respectively. Even though the unit-cell volumes of CeRh$_{1-x}$Ir$_x$In$_5$ increase weakly at atmospheric pressure with increasing x, the empirically observed scaling of physical properties implies that Ir acts effectively as a positive chemical pressure, i.e., as though Ir substitution decreases the cell volume. In view of results shown in Fig. 4 this apparent inconsistency might be resolved if, for $x \leq 0.25$, physically applied pressure and Ir substitution for Rh similarly change the ratio of tetragonal lattice parameters c/a. This remains to be established.

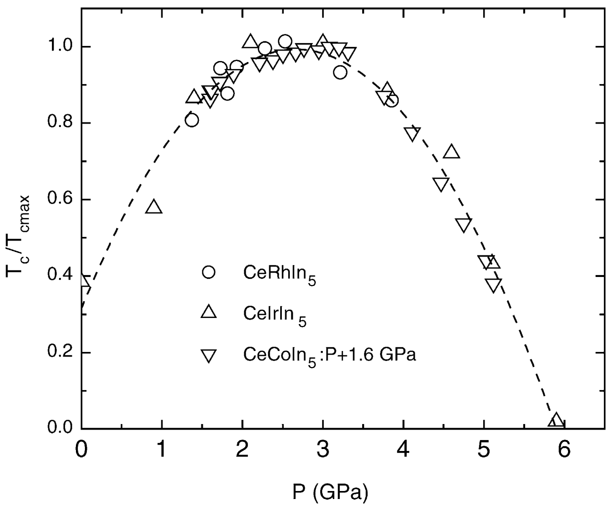

Fig. 3. Superconducting transition temperatures, normalized by the maximum value induced by pressure, as a function of pressure for CeRhIn$_5$, CeIrIn$_5$ and CeCoIn$_5$. The pressure axis for CeCoIn$_5$ has been shifted by 1.6 GPa to reflect its smaller unit cell volume relative to that of CeRhIn$_5$. $T_c(P)$ data for CeIrIn$_5$ are taken from reference [14]. The overall shape of these curves is very similar for each compound as shown by the dashed curve which is a quadratic fit to all of the data and is given by $T_c/T_{\mathrm{cmax}} = 0.32 + 0.51P - 9.5 \times 10^{-2}P^2$.

superconductors [12]. In contrast to a conventional superconductor where $1/T_1$ decreases exponentially below T_c due to a finite superconducting gap over the entire Fermi surface, the experimental temperature dependences are consistent with the presence of line nodes in the gap. These results strongly support the interpretation that $PuCoGa_5$ is an unconventional d-wave superconductor.

As with $CeCoIn_5$ and $CeIrIn_5$, there is no evidence for antiferromagnetic order in either $PuCoGa_5$ or $PuRhGa_5$, which also exhibits a dome-shaped dependence of $T_c(P)$ [29] and, by analogy to $PuCoGa_5$, also is probably a d-wave superconductor. So far, there is no antiferromagnetic Pu-analog of $CeRhIn_5$. But like the $CeMIn_5$ family, a comparison of these Pu superconductors to other examples strengthens an argument that soft magnetic fluctuations may mediate superconductivity. This comparison is made in Fig. 4 where we plot the superconducting transition temperatures of $CeMIn_5$ and $PuMGa_5$ as a function of their structural parameters. The upper plot shows T_c versus the ratio of tetragonal lattice parameters c/a. In both families of compounds, T_c is a linear function of structural anisotropy, and, further, the relative rate of change of T_c with increasing c/a ratio, $\partial \ln T_c/\partial(c/a) \approx 80$, is nearly identical. Although the origin of this empirical relationship remains to be established, it suggests that the magnetic mechanism of superconductivity in $CeMIn_5$ also may determine T_c in the Pu compounds. This suggestion is supported from the correlation plotted in the lower panel of Fig. 4. The structural parameter $(2zc - a)/a$ measures a weak tetragonal distortion of face-centered cubic units of the $HoCoGa_5$ structure type [30] that are stacked sequentially along the c-axis and separated by an intervening layer of $MIn(Ga)_2$. As argued recently in the context of isostructural and antiferromagnetic $UTGa_5$ [31] and $NpTGa_5$ [32] compounds, where T is an isoelectronic transition metal from either the Fe, Ni or Co columns, a weak distortion measured by $(2zc - a)/a$ in these compounds tunes magnetic interactions through a change in spin hybridization of the 5f element with the transition metal element. Extending these arguments to the Ce- and Pu-based superconductors suggests that the increase in T_c with increasing local tetragonal distortion also reflects a hybridization-induced change in soft magnetic excitations that mediate Cooper pairing in these materials.

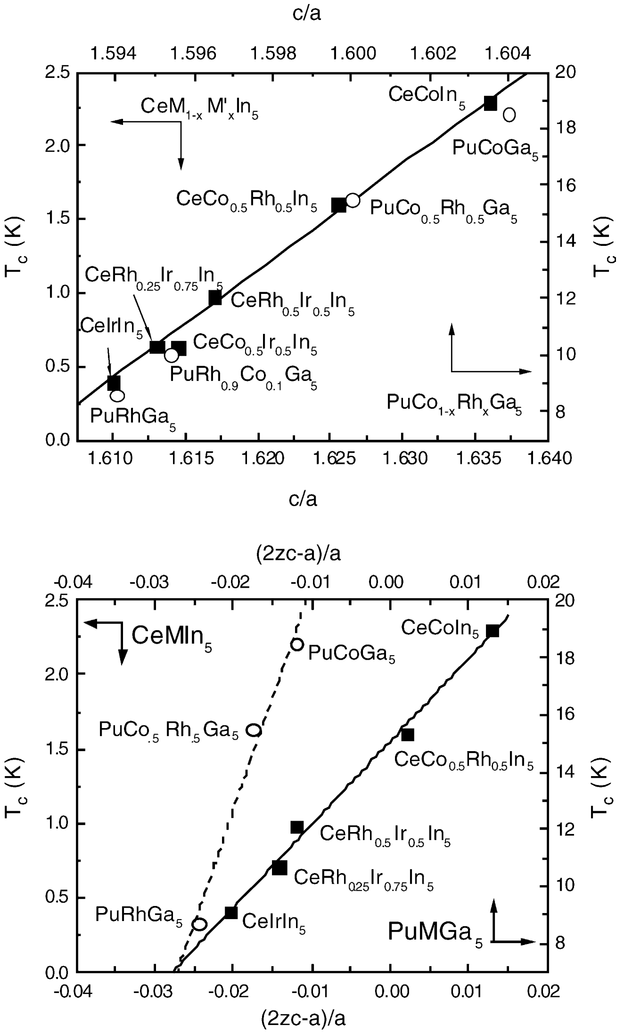

Fig. 4. Upper panel: superconducting transition temperatures as a function of tetragonal lattice parameters c/a for various Ce- and Pu-members of the $CeMIn_5$ and $PuMGa_5$ families. The relative dependence of T_c on c/a is nearly identical for both families. Lower panel: T_c vs. the local structure parameter $(2zc - a)/a$, where c and a are the tetragonal lattice parameters and z locates the position of the unique In(Ga) atom along the c-axis of the $HoCoGa_5$ structure type. $(2zc - a)/a$ measures the magnitude of a weak tetragonal distortion of the face-centered building block of these compounds.

4. Summary

In summary, a relationship between magnetism and superconductivity is clear in some examples of strongly correlated electron materials and less so in others. Pressure studies have been particularly useful in revealing this relationship in $CeRhIn_5$, and comparing its pressure response to those of other $CeMIn_5$ compounds has allowed an extension of this relationship to those materials as well. Had the $CeMIn_5$ systems not been found before the Pu-based superconductors, their relationship to magnetism would not have been as apparent. In this case, though, the relationship is not established from pressure measurements alone but has come through a common dependence of T_c on structural parameters, which, together with other properties of these Pu superconductors, strongly suggests that their superconductivity also is mediated by magnetic fluctuations. In the absence of a theory of magnetically mediated superconductivity equivalent to that for conventional superconductors, it is difficult to make a definitive test that unambiguously rules out alternative pairing mechanisms; for example, there is no isotope-effect equivalent type of experiment for magnetically mediated superconductivity. Consequently, the pairing mechanism must be inferred from a body of experiments that are consistent with broad expectations. The $CeMIn_5$ and $PuMGa_5$ superconductors are not alone in this respect, and to our knowledge, magnetically mediated superconductivity has not been proven in any material. In spite of this situation, it is highly likely that there are several examples of magnetically mediated superconductors. Here, we have discussed some of the evidence that points consistently toward an unconventional pairing mechanism in these two isostructural families. If, in fact, $PuMGa_5$ materials are spin-mediated superconductors, their transition temperatures are far higher than any other examples, except the cuprates, and, in this regard, may be a valuable link in bridging our understanding of superconductivity in correlated f-electron systems and the high T_c cuprates.

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