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Progress and Puzzles in Plutonium Superconductors

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Recent progress revealing the unconventional nature of both normal and superconducting states of $PuCoGa_5$ and $PuRhGa_5$ has cast these materials in a broader context of strongly correlated materials and phenomena. In this regard, a comparison of the Pu-based superconductors to their isostructural Ce-based counterparts suggests a set of experiments that might lead to a clearer definition of the puzzles they present.

KEYWORDS: $PuCoGa_5$, $PuRhGa_5$, pressure, spin-lattice relaxation rate

The discovery of superconductivity in $PuCoGa_5$[1)] and soon thereafter in isostructural $PuRhGa_5$[2)] has stimulated experimental and theoretical activity aimed at understanding fundamental electronic and magnetic properties of these and related actinide compounds. The superconducting transition temperatures T_c of 18.5 and 8.7 K for $PuCoGa_5$ and $PuRhGa_5$, respectively, are much higher than in any other 5f-electron system,[3)] which suggests that superconductivity might be influenced by the unusual 5f-electron configuration of Pu that appears to be poised delicately at a localized/delocalized border.[4)] Developing an appropriate theoretical framework for these 5f electrons, which also experience strong electronic correlations,[5,6)] is particularly challenging but important for establishing a starting point for understanding the superconducting mechanism. Likewise, the radioactivity of Pu constrains what experiments can be done and how they are conducted as well as induces time-dependent disorder due to self-radiation damage.[7)] In spite of these difficulties, remarkable progress has begun to be made. Among several advances, Knight shift,[8)] spin-lattice relaxation rate,[8,9)] and muon-spin resonance[10)] experiments have shown recently that the superconductivity in both Pu compounds is spin singlet and that the superconducting energy gap is unconventional, probably with d-wave symmetry. This is the same gap symmetry found in several heavy-fermion compounds, such as $CeCoIn_5$[11)] and $CeRhIn_5$[12)] which are isostructural with the Pu superconductors, as well as the high T_c cuprates.[13)] Indeed, Curro *et al.*[8)] demonstrated that the spin-relaxation rate divided by temperature, $1/T_1T$, normalized to its value at T_c, scales above and below T_c as a function of T/T_c for $PuCoGa_5$, $CeCoIn_5$ and $YBa_2Cu_3O_7$. Such scaling strongly suggests that pairing in $PuCoGa_5$ is mediated by antiferromagnetic spin fluctuations, as may be the case in $CeCoIn_5$ and the cuprates. In the latter two materials, there is evidence for 'nearby' antiferromagnetism that could serve as the origin of fluctuations as these systems are tuned away from long-range magnetic order and into a superconducting state by hole doping or pressure, but so far, this has not been established for either $PuCoGa_5$ or $PuRhGa_5$. As we will discuss, there are similarities between the Pu superconductors and their Ce-based analogs that hint to the origin of antiferromagnetic fluctuations in these compounds.

In addition to their common tetragonal crystal structure, both Pu115 and Ce115 families exhibit a linear increase in T_c with increasing ratio of their lattice parameters c/a[14)] and a dome-like variation in T_c with applied pressure.[15–17)] The insets of Figs. 1(a) and (b) show the pressure dependences of T_c. Although T_c's of the Ce115s are a factor of four or more lower and the dome-width correspondingly narrower than those of their Pu analogs, the main panels of these figures show that T_c, normalized by its maximum value T_c^{max}, can be scaled similarly in both families. Although their shapes are qualitatively similar, the detailed functional form of the scaled T_c's is not identical for Ce115 and Pu115. As with the dependence of T_c on c/a ratio, where the relative change $dlnT_c/d(c/a)$ is the same but $dT_c/d(c/a)$ is much larger for the Pu115's,[14,18)] a comparison of $T_c(P)$ and scaled $T_c(P)$ curves emphasize not only differences in characteristic electronic energy scales but also more subtle distinctions between roles played by less spatially extended wavefunctions of Ce's 4f electrons and those of the more extended and strongly hybridizing 5f-electrons of Pu.

Stronger hybridization of 5f- versus 4f-electrons is obvious in the Rh members: $CeRhIn_5$ is an antiferromagnet at ambient pressure and a pressure-induced superconductor,[19)] in contrast to $PuRhGa_5$, which superconducts in the absence of applied pressure.[2)] Because pressure favors stronger hybridization in Ce-compounds, a more appropriate comparison should be between $CeRhIn_5$ under pressure and $PuRhGa_5$ at atmospheric pressure. A surrogate for $CeRhIn_5$ under pressure is $CeCoIn_5$,[16)] which has been studied in greater detail. Besides both $CeCoIn_5$ and $PuRhGa_5$ being superconductors at atmospheric pressure, the ratio of their upper critical fields H_{c2} along the [100] and [001] directions is comparable, 2.5[20)] and 1.8[21)] for $CeCoIn_5$ and $PuRhGa_5$, respectively, and qualitatively reflects their relative structural and electronic anisotropies. Pressure studies of $CeCoIn_5$ have suggested that at $P = 0$ it is just beyond the antiferromagnetic/superconducting boundary that is accessed in $CeRhIn_5$ with applied pressure.[16)] Not withstanding the distinction between 4f- and 5f-electrons, we assume in the following that $PuRhGa_5$ is analogous to $CeCoIn_5$, i.e. just beyond a magnetic/superconducting boundary and, as implied by the scaling of data in Fig. 1(b), that $PuCoGa_5$ is a higher-pressure variant $PuRhGa_5$.

We examine this assumption by comparing in Fig. 2

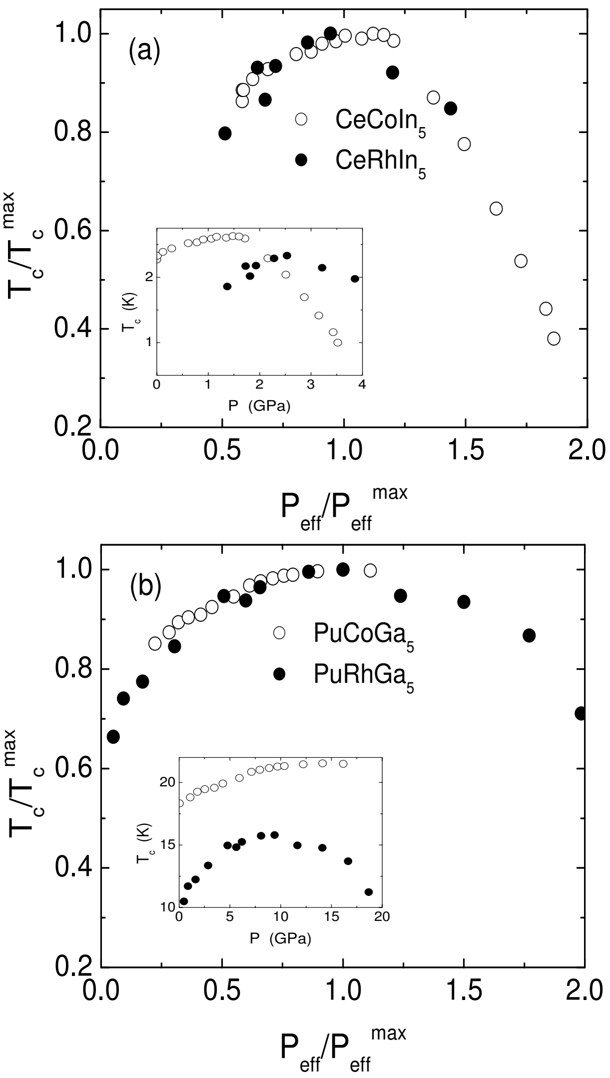

Fig. 1. (a) Superconducting transition temperature, normalized by its maximum value T_c^{max} as a function of reduced pressure, where P_{eff} is the sum of applied and chemical pressures and P_{eff}^{max} is P_{eff} at T_c^{max}. The chemical pressure is taken to be zero for $CeRhIn_5$ and estimated for $CeCoIn_5$ to be 1.3 GPa, which is given by $B\Delta V/V$ where B is the average bulk modulus of $CeRhIn_5$ and $CeCoIn_5$, ΔV is the difference in unit cell volumes of $CeRhIn_5$ and $CeCoIn_5$ and V is the cell volume of $CeCoIn_5$.[18] The inset is a plot of T_c versus applied pressure.[15,16] (b) Normalized superconducting transition temperature as a function of reduced pressure for $PuRhGa_5$ and $PuCoGa_5$. T_c^{max}, P_{eff} and P_{eff}^{max} are defined as in panel (a). The chemical pressure experienced by $PuCoGa_5$ is approximately 4 GPa, estimated using the same procedure as for $CeCoIn_5$ but with $PuRhGa_5$ as the reference.[18] The inset gives T_c versus applied pressure, where the data have been adopted from ref. [17].

the temperature dependence of the spin-lattice relation rate of $CeRhIn_5$ under pressure with that of $PuRhGa_5$ and $CeCoIn_5$. Somewhat counter to our assumption, the normalized relaxation rate of $PuRhGa_5$ evolves with temperature much more like that of $CeRhIn_5$ at 2.0 GPa, and below $T/T_c < 3$, it deviates qualitatively from the temperature dependence of $CeCoIn_5$. However, increasing the applied pressure on $CeRhIn_5$ by just 5%, that is to 2.1 GPa, induces a temperature dependence of T_1 closely resembling that of $CeCoIn_5$ and $PuCoGa_5$.

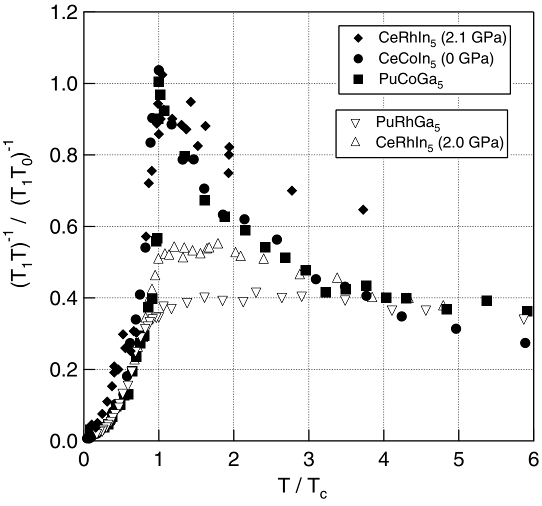

Fig. 2. Normalized spin-lattice relaxation rate divided by temperature, $1/T_1T$, versus temperature divided by T_c. The normalizing factor $1/T_1T_0$ was determined as in ref. [8]. The same scaling form collapses T_1 data for $YBa_2Cu_3O_7$ onto curves for $CeCoIn_5$ and $PuCoGa_5$, and the temperature dependence of the scaled data above T_c are consistent with nuclear relaxation being dominated by antiferromagnetic spin fluctuations.[8] Although data for $CeRhIn_5$ at 2.1 GPa do not fall exactly on the curve for $CeCoIn_5$, its systematic evolution with pressure suggests that this will happen if measurements were made at a slightly higher pressure. T_1 data are taken from the literature: $PuCoGa_5$,[8] $PuRhGa_5$,[9] $CeCoIn_5$,[11] and $CeRhIn_5$ at 2.0 and 2.1 GPa.[12]

Rather different interpretations have been suggested for the Korringa-like $1/T_1T$ behavior found above T_c in $CeRhIn_5$ and $PuRhGa_5$. In $CeRhIn_5$, it has been attributed to the competing effects of a decrease in $1/T_1T$ due to the development of a presumed pseudogap, which appears first at lower pressures where antiferromagnetism and superconductivity coexist, and an increase in $1/T_1T$ due to the development of antiferromagnetic fluctuations.[12] On the other hand, the constant $1/T_1T$ in $PuRhGa_5$ has been argued to reflect Fermi-liquid behavior.[9] With information presently available, it is, unfortunately, not possible to distinguish unambiguously between these two very different scenarios. We note, however, that the resistivity of $PuRhGa_5$ increases approximately as $T^{4/3}$ and its static susceptibility is Curie-Weiss-like in the temperature range where $1/T_1T$ is a constant,[2] behaviors not characteristic of a Landau Fermi liquid.

If we adopt the view that Korringa behavior in $PuRhGa_5$ is not due to the formation of a Fermi-liquid state, then it is possible to map Ce115's and Pu115s onto a generic temperature-pressure phase diagram, given in Fig. 3, that is explicitly valid for $CeRhIn_5$.[12] De Haas-van Alphen measurements on $CeRhIn_5$ find that the effective quasiparticle mass diverges as $P2$ is approached and that the Fermi-surface volume expands at $P2$ to become comparable to that of $CeCoIn_5$ at $P = 0$.[22] Recent measurements of the specific heat of $CeRhIn_5$ under pressure have discovered that magnetic order is induced with the application of a magnetic field when

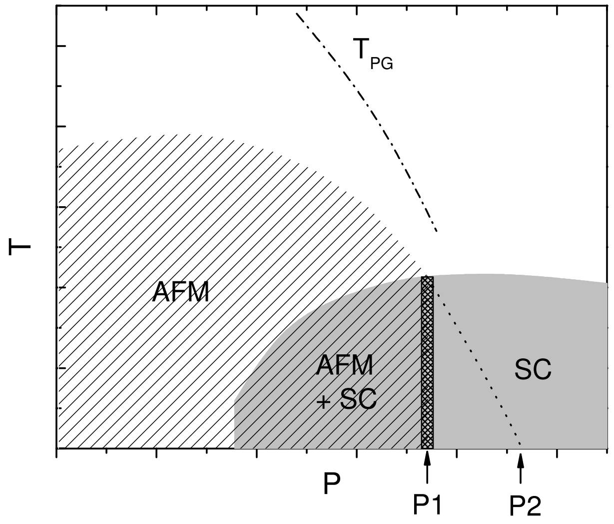

Fig. 3. Schematic temperature-pressure phase diagram based on extensive pressure studies of $CeRhIn_5$.[12,15] AFM: antiferromagnetic; SC: superconducting with no evidence for antiferromagnetism in zero magnetic field; AFM+SC: coexistence of antiferromagnetism and superconductivity; T_{PG}: temperature-pressure boundary below which there is evidence for a pseudogap in $CeRhIn_5$. The pressure and temperature axes are arbitrary but relative for $CeCoIn_5$ and Pu115's, as discussed in the text.

$P1 < P < P2$.[23] These results validate an extrapolation of the $T_N(P)$ phase boundary to $T = 0$ at $P2$ and provide the previously missing rationale for a diverging quasiparticle mass and change in Fermi-surface volume at $P2$. $CeCoIn_5$ at $P = 0$ exhibits pronounced non-Fermi-liquid properties that are expected if it were close to a quantum-critical point.[16] On the basis of this, its Fermi-surface volume being close to that of $CeRhIn_5$ just beyond $P2$ and its smaller cell volume relative to $CeRhIn_5$, it appears that $CeCoIn_5$ at atmospheric pressure is, in effect, very close to $P2$ in Fig. 3. From the comparison of relaxation data in Fig. 2, $PuRhGa_5$ at $P = 0$ resembles $CeRhIn_5$ at a pressure just beyond $P1$, i.e., there is no evidence for long range magnetic order in zero-field and $1/T_1T$ is constant from T_c to a few times T_c. This suggests that magnetic order also might be 'hidden' by $PuRhGa_5$'s superconductivity and be a source of magnetic fluctuations needed for Cooper pairing. It further follows that $PuCoGa_5$ at atmospheric pressure might be located, like $CeCoIn_5$, on this generic phase diagram at a pressure close to $P2$. Of course, the generic pressures $P1$ and $P2$ are expected to be quantitatively different for the Ce115's and Pu115's.

To the extent these analogies are valid, we would expect: (1) the spin-lattice relaxation rate of $PuRhGa_5$ evolves with applied pressure to become similar to that of $CeCoIn_5$ and $PuCoGa_5$ at atmospheric pressure; (2) to find evidence for field-induced magnetic order in $PuRhGa_5$; (3) that the Fermi surfaces of $PuRhGa_5$ and $PuCoGa_5$ are similar to each other, differing primarily in the volume they enclose, and similar to those of their Ce115 counterparts; and (4) that an isoelectronic Pu115 compound with a larger unit cell than $PuRhGa_5$ should be antiferromagnetic. Should these expectations eventually be confirmed in future experiments, some pieces of the puzzles presented by unconventional superconductivity in Pu115 compounds may begin to fall in place or at least become better defined. Irrespective of the outcome, these superconductors offer fertile ground for further experimental and theoretical research.

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