Progress and Puzzles in Plutonium Superconductors

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Recent progress revealing the unconventional nature of both normal and superconducting states of PuCoGa₅ and PuRhGa₅ 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: PuCoGa₅, PuRhGa₅, pressure, spin-lattice relaxation rate

The discovery of superconductivity in PuCoGa₅ and soon thereafter in isostructural PuRhGa₅ has stimulated experimental and theoretical activity aimed at understanding fundamental electronic and magnetic properties of these and related actinide compounds. The superconducting transition temperatures, 18.5 and 8.7 K for PuCoGa₅ and PuRhGa₅, respectively, are much higher than in any other 5f-electron system, 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.

Developing an appropriate theoretical framework for these 5f electrons, which also experience strong electronic correlations, 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. In spite of these difficulties, remarkable progress has begun to be made. Among several advances, Knight shift, spin-lattice relaxation rate, and muon-spin resonance experiments have shown recently that the superconductivity in both Pu compounds is spin single 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 CeCoIn₅ and CeRhIn₅ which are isostructural with the Pu superconductors, as well as the high Tc cuprates. Indeed, Curro et al. demonstrated that the spin-relaxation rate divided by temperature, 1/TcT, normalized to its value at Tc, scales above and below Tc as a function of T/Tc for PuCoGa₅, CeCoIn₅ and YBa₂Cu₃O₇. Such scaling strongly suggests that pairing in PuCoGa₅ is mediated by antiferromagnetic spin fluctuations, as may be the case in CeCoIn₅ 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 PuCoGa₅ or PuRhGa₅. 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 Tc with increasing ratio of their lattice parameters c/a and a dome-like variation in Tc with applied pressure. The insets of Figs. 1(a) and (b) show the pressure dependences of Tc. Although Tc’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 Tc, normalized by its maximum value Tc,max, can be scaled similarly in both families. Although their shapes are qualitatively similar, the detailed functional form of the scaled Tc’s is not identical for Ce115 and Pu115. As with the dependence of Tc on c/a ratio, where the relative change dlnTc/d(c/a) is the same but Tc/P(c/a) is much larger for the Pu115’s, a comparison of Tc/P and scaled Tc/(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: CeRhIn₅ is an antiferromagnet at ambient pressure and a pressure-induced superconductor, in contrast to PuRhGa₅, which superconducts in the absence of applied pressure. Because pressure favors stronger hybridization in Ce-compounds, a more appropriate comparison should be between CeRhIn₅ under pressure and PuRhGa₅ at atmospheric pressure. A surrogate for CeRhIn₅ under pressure is CeCoIn₅, which has been studied in greater detail. Besides both CeCoIn₅ and PuRhGa₅ being superconductors at atmospheric pressure, the ratio of their upper critical fields Hc₂ along the [100] and [001] directions is comparable, 2.5 and 1.8 for CeCoIn₅ and PuRhGa₅, respectively, and qualitatively reflects their relative structural and electronic anisotropies. Pressure studies of CeCoIn₅ have suggested that at P = 0 it is just beyond the antiferromagnetic/superconducting boundary that is accessed in CeRhIn₅ with applied pressure. Notwithstanding the distinction between 4f- and 5f-electrons, we assume in the following that PuRhGa₅ is analogous to CeCoIn₅, i.e. just beyond a magnetic/superconducting boundary and, as implied by the scaling of data in Fig. 1(b), that PuCoGa₅ is a higher-pressure variant PuRhGa₅.

We examine this assumption by comparing in Fig. 2...
rate of CeRhIn is to 2.1 GPa, induces a temperature dependence of $T_c$ normalized superconducting transition temperature as a function of reduced pressure, where $P_{\text{eff}}$ is the sum of applied and chemical pressures and $P_{\text{eff}}^{\text{max}}$ is $P_{\text{eff}}$ at $T_c^{\text{max}}$. The chemical pressure is taken to be zero for CeRhIn5 and estimated for CeCoIn5 to be 1.3 GPa, which is given by $B\Delta V/V$ where $B$ is the average bulk modulus of CeRhIn5 and CeCoIn5, $\Delta V$ is the difference in unit cell volumes of CeRhIn5 and CeCoIn5 and $V$ is the cell volume of CeCoIn5. The inset is a plot of $T_c$ versus applied pressure. The same procedure as for CeCoIn5 is given by its maximum value $T_c^{\text{max}}$, $\Delta T_c$ is approximately 4 GPa, estimated using the same procedure as for CeCoIn5, but with PuRhGa5 as the reference. The inset gives $T_c$ versus applied pressure, where the data have been adopted from ref. [17].

Fig. 1. (a) Superconducting transition temperature, normalized by its maximum value $T_c^{\text{max}}$, as a function of reduced pressure, where $P_{\text{eff}}$ is the sum of applied and chemical pressures and $P_{\text{eff}}^{\text{max}}$ is $P_{\text{eff}}$ at $T_c^{\text{max}}$. The chemical pressure is taken to be zero for CeRhIn5 and estimated for CeCoIn5 to be 1.3 GPa, which is given by $B\Delta V/V$ where $B$ is the average bulk modulus of CeRhIn5 and CeCoIn5, $\Delta V$ is the difference in unit cell volumes of CeRhIn5 and CeCoIn5 and $V$ is the cell volume of CeCoIn5. The inset is a plot of $T_c$ versus applied pressure. The chemical pressure experienced by PuCoGa5 is approximately 4 GPa, estimated using the same procedure as for CeCoIn5, but with PuRhGa5 as the reference. The inset gives $T_c$ versus applied pressure, where the data have been adopted from ref. [17].

Rather different interpretations have been suggested for the Körringa-like $1/T_cT$ behavior found above $T_c$ in CeRhIn5 and PuRhGa5. In CeRhIn5, it has been attributed to the competing effects of a decrease in $1/T_cT$ due to the development of a presumed pseudogap, which appears first at lower pressures where antiferromagnetism and superconductivity coexist, and an increase in $1/T_cT$ due to the development of antiferromagnetic fluctuations. On the other hand, the constant $1/T_cT$ in PuRhGa5 has been argued to reflect Fermi-liquid behavior. 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 PuRhGa5 increases approximately as $T^{4/3}$ and its static susceptibility is Curie-Weiss-like in the temperature range where $1/T_cT$ is a constant, behaviors not characteristic of a Landau Fermi liquid.

If we adopt the view that Körringa behavior in PuRhGa5 is not due to the formation of a Fermi-liquid state, then it is possible to map Ce115’s and Pu115’s onto a generic temperature-pressure phase diagram, given in Fig. 3, that is explicitly valid for CeRhIn5. De Haas-van Alphen measurements on CeRhIn5 find that the effective quasiparticle mass diverges as $P^2$ is approached and that the Fermi-surface volume expands at $P^2$ to become comparable to that of CeCoIn5 at $P = 0$. Recent measurements of the specific heat of CeRhIn5 under pressure have discovered that magnetic order is induced with the application of a magnetic field when

Fig. 2. Normalized spin-lattice relaxation rate divided by temperature, $1/T_cT$; versus temperature divided by $T_c$. The normalizing factor $1/T_cT_0$ was determined as in ref. [8]. The same scaling form collapses $T_1$ data for Yb123CuOy onto curves for CeCoIn5 and PuCoGa5, and the temperature dependence of the scaled data above $T_c$ are consistent with nuclear relaxation being dominated by antiferromagnetic spin fluctuations. Although data for CeRhIn5 at 2.1 GPa do not fall exactly on the curve for CeCoIn5, 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: PuCoGa5, PuRhGa5, CeCoIn5, and CeRhIn5 at 2.0 and 2.1 GPa.
Fig. 3. Schematic temperature-pressure phase diagram based on extensive pressure studies of CeRhIn$_5$.\(^\text{12,15}\) AFM: antiferromagnetic; SC: superconducting with no evidence for antiferromagnetism in zero magnetic field; AFM+SC: coexistence of antiferromagnetism and superconductivity; $T_{P\text{G}}$: 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.

$P_1 < P < P_2$.\(^\text{21}\) These results validate an extrapolation of the $T_N(P)$ phase boundary to $T = 0$ at $P_2$ and provide the previously missing rationale for a diverging quasiparticle mass and change in Fermi-surface volume at $P_2$. CeCoIn$_5$ at $P = 0$ exhibits pronounced non-Fermi-liquid properties that are expected if it were close to a quantum-critical point.\(^\text{16}\) On the basis of this, its Fermi-surface volume being close to that of CeRhIn$_5$ just beyond $P_2$ and its smaller cell volume relative to CeRhIn$_5$, it appears that CeCoIn$_5$ at atmospheric pressure is, in effect, very close to $P_2$ 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 $P_1$, i.e., there is no evidence for long range magnetic order in zero-field and $1/T_1 T$ is constant from $T_\text{c}$ to a few times $T_\text{c}$. 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 $P_2$. Of course, the generic pressures $P_1$ and $P_2$ 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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