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Superconductivity in plutonium compounds

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Abstract

Although the family of plutonium-based superconductors is relatively small, consisting of four compounds all of which crystallize in the tetragonal $HoCoGa_5$ structure, these materials serve as an important bridge between the known Ce- and U-based heavy fermion superconductors and the high-temperature cuprate superconductors. Further, the partial localization of 5f electrons that characterizes the novel electronic properties of elemental plutonium appears to be central to the relatively high superconducting transition temperatures that are observed in $PuCoGa_5$, $PuRhGa_5$, $PuCoIn_5$, and $PuRhIn_5$.

Keywords:
plutonium, superconductivity, $PuCoGa_5$

1. Introduction

$PuCoGa_5$ was the first plutonium-based superconductor, and it displayed the rather high transition temperature, T_c, of 18.5 K [1]. Although this discovery was rather surprising, the initial perspective that $PuCoGa_5$ was related to the previously discovered Ce-115 ($CeCoIn_5$, $CeRhIn_5$, and $CeIrIn_5$) superconductors and reflected the novel properties of elemental plutonium has survived the test of time [2, 3, 4]. In fact, the subsequent discoveries of superconductivity in $PuRhGa_5$, $PuCoIn_5$, and $PuRhIn_5$ and their respective physical properties further confirm this picture.

The $CeMIn_5$ superconductors are composed of layers of $CeIn_3$ and MIn_2, and much of the physics of these materials can be described by considering the materials as layered variants of $CeIn_3$ [5, 2, 6]. In fact, the Ce-115

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family of materials includes examples that have single layers of $CeIn_3$ separated by bilayers of MIn_2 (e.g., superconducting $CePt_2In_7$ [7]) as well as bilayers of $CeIn_3$ separated by single layers of MIn_2 (e.g., Ce_2RhIn_8, which displays pressure-induced superconductivity [8]). Pursuing this metaphor, $PuCoGa_5$ would be described as made up of layers of $PuGa_3$ and $CoGa_2$. Further, $PuGa_3$ is directly related to the high-temperature, face-centered-cubic (fcc) δ-phase of Pu, which is stabilized to room temperature by the addition of small amounts of Ga. Continuing to add Ga to elemental Pu leads one to the binary compound Pu_3Ga and, at least figuratively, to fcc $PuGa_3$ (in reality, $PuGa_3$ crystallizes in either a hexagonal or rhombohedral structure [9]), the parent compound and analog of $CeIn_3$ for $PuCoGa_5$. Figure 1 depicts the Pu-115 related crystal structures that have been synthesized to date[10, 13]. Following the discovery of $PuCoGa_5$, $PuRhGa_5$ (T_c=8.6 K) [11], $PuCoIn_5$ (T_c=2.5 K) [12], and $PuRhIn_5$ (T_c=1.7 K) [3] have also been reported and have been shown to be superconducting. Pu-based 1-2-7 and 2-1-8 compounds have also been reported (e.g., Pu_2PtGa_8 and $PuPt_2In_7$, but superconductivity has yet to be observed in these structures [13].

Plutonium is a fascinating metal. Its 5f electrons straddle the boundary between localized and itinerant behavior. The low-temperature, monoclinic α-Pu displays itinerant f-electron behavior, while the higher-temperature δ-Pu phase displays partially localized f-electron behavior, which, as noted above, can be stabilized to room temperature by the addition of small amounts of impurities such as Ga. This easily perturbed electronic configuration gives rise to an extremely complex metallurgy for plutonium metal[14] and challenges the state of the art in electronic structure calculation methods [15]. The crossover from localized to itinerant f-electron behavior is central to the phenomenology of the broader family of heavy fermion compounds [16] and is also reflective of unconventional superconductors that exist near magnetically ordered phases, as occurs in the Ce-115 compounds [6].

2. Synthesis and Structural Properties

Although relatively few groups can actively work with plutonium because of its radiological hazards, large single crystals of each of the known plutonium superconductors have been grown using molten metal (Ga or In) flux techniques, and single-crystal structural determinations have also been made for each compound. Pictures of representative crystals of these materials are shown in Figure 2 [13]. The structural and ground state properties of these

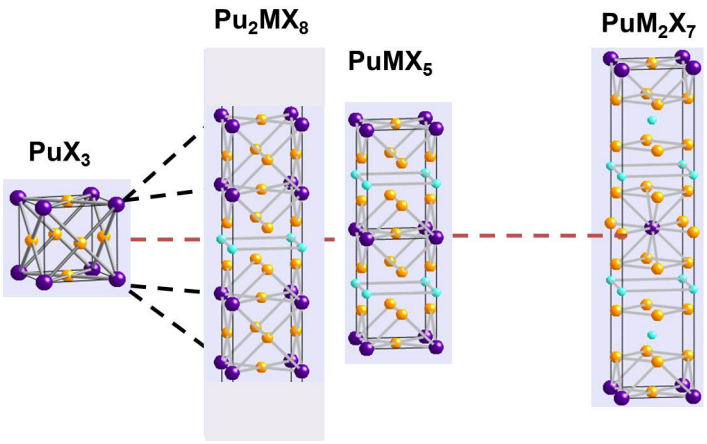

Figure 1: An illustration of the stacking orders that give rise to the Pu-115, Pu-218, and Pu-127 stoichiometries. M is a transtion metal, and X can be Ga or In.

materials are summarized in Table 1. Full structural details of the Pu-based superconductors have been reported by Bauer et al. [13].

As will be discussed below, an interesting feature of the Pu-115 superconductors (as well as the Ce-115 superconductors) is the linear correlation between T_c and the ratio of the tetragonal lattice constants, c/a, independent of chemical composition [17]. It thus appears that the spacing between alternating layers of PuX_3 (X=In or Ga) and MX_2 (M=Co or Rh) is directly correlated with superconducting transition temperature in a way that no other independent structural parameter is. Similarly, intermediate concentrations of Rh in $PuCo_{1-x}Rh_xGa_5$ display the same behavior, indicating that layer spacing is an essential parameter while, in contrast to many other families of superconductors, Rh-Co disorder in the (Rh,Co)Ga_2 layer does not have a deleterious effect on superconducting transition temperature. This is the first of several indications that the superconductivity in Pu-115 materials is qausi-two-dimensional and driven by the behavior of the $PuGa_3$ ($PuIn_3$) layer, both of which display long-range magnetic order as compounds.

3. Superconducting Properties

Bulk superconductivity in $PuCoGa_5$, $PuRhGa_5$, and $PuCoIn_5$ has been demonstrated by heat capacity measurements [1, 18, 12]. Further, the jump ΔC in specific heat at T_c allows an estimate of the Sommerfeld coefficient

	$PuCoGa_5$	$PuRhGa_5$	$PuCoIn_5$	$PuRhIn_5$
T_c(K)	18.5	8.6	2.5	1.7
Lattice Constant($\AA$)	a=4.232 c=6.786	a=4.301 c=6.857	a=4.574 c=7.439	a=4.621 c=7.460
Discovery	Sarrao, 2002 [1]	Wastin, 2003 [11]	Bauer, 2012 [12]	Bauer, 2012 [3]

Table 1: Superconducting transition temperature, lattice constants, and original reference for known plutonium supercondcutors

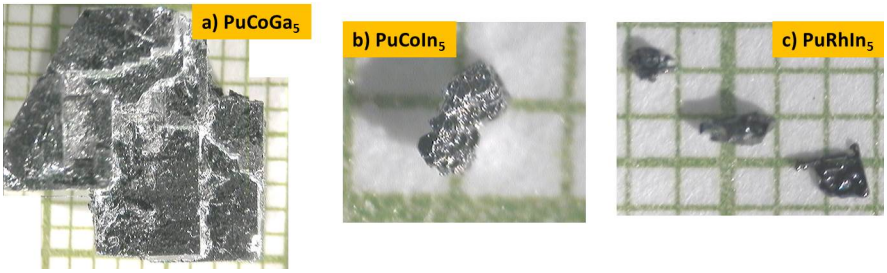

Figure 2: Pictures of representative crystals of known plutonium superconductors. The grid lines in the images are a millimeter scale.

(assuming the BCS relation $\Delta C/\gamma T_c = 1.43$) for $PuCoIn_5$, $PuCoGa_5$, and $PuRhGa_5$ of 150, 100, and 50 mJ/mol K^2, respectively, indicating heavy fermion behavior. The inferred values of the Sommerfeld coefficient are also consistent with the value of C/T at temperatures just about T_c given in Table 2 [3]. Although superconductivity in $PuRhIn_5$ has not yet been observed by heat capacity measurements due to its low T_c, thermal expansion as well as resistivity, susceptibility, and NMR measurements indicates bulk, heavy fermion superconductivity in $PuRhIn_5$[13].

An interesting feature of the Pu-115 superconductors is that, due to self-irradiation from radioactive Pu, the superconducting transition temperature decreases as a function of time [19]. This has been studied most carefully in $PuCoGa_5$ and $PuRhGa_5$ because of their relatively higher transition temperatures [20]. The damage created by self-irradiation also produces pinning centers, which cause the upper critical field to increase as a function of time [20]. Jutier et al. [21] have used such data to analyze the nature of the superconducting pairing in $PuCoGa_5$, suggesting a phononic mechanism in contrast to the more common 'dirty d-wave'picture. On the other hand, estimates of the electron-phonon coupling via measurements of the ultrafast

optical response in $PuCoGa_5$ argue against a phonon-based mechanism[22].

The upper critical field $H_{c2}(0)$ as well as the initial temperature derivative of $H_{c2}(T)$, $\partial H_{c2}/\partial T$ @ T_c for $PuMX_5$ is included in Table 2 [3, 23, 24]. For both $PuCoIn_5$ ($H^c_{c2}(0) = 10$ T) and $PuRhIn_5$ ($H^c_{c2}(0) = 7$ T), the measured $H_{c2}(0)$ is much smaller than expected ($0.7T_c$ $\partial H_{c2}/\partial T$ @ T_c =24 and 16 T, respectively) for purely orbital limiting [25], indicating that Pauli limiting may play an important role in determining the value of H_{c2}, as in the $CeMIn_5$ superconductors [26, 6]. $H_{c2}(0)$ has yet to be measured directly in $PuCoGa_5$ and $PuRhGa_5$ because of their higher T_c and correspondingly large H_{c2}; however, both display anomalous behavior, with $PuCoGa_5$'s $H_{c2}(T)$ being less anisotropic than expected, and $PuRhGa_5$'s $H_{c2}(T)$ displaying an anomalous shape [3].

Nuclear magnetic and quadrupole resonance (NMR and NQR) measurements on Pu-115 superconductors provide perhaps the most direct insight into the symmetry of the superconducting order parameter in these materials (Figure 3). Knight shift measurements on $PuCoGa_5$ reveal a spin susceptibility that decreases in the superconducting state, consistent with singlet pairing [27]. A power-law temperature dependence of the spin-lattice relaxation rate $1/T_1 \sim T^3$ below T_c, along with absence of a Hebel-Slichter peak at T_c, provides evidence for d-wave superconductivity with line nodes in the superconducting gap [27]. Point contact spectroscopy studies of $PuCoGa_5$ also find a four-fold modulation of the superconducting gap within the ab-plane, consistent with a $d_{x^2-y^2}$ order parameter symmetry [31]. Spin lattice relaxation measurements for $PuRhGa_5$, $PuCoIn_5$, and $PuRhIn_5$ also find $1/T_1 \sim T^3$ below T_c [28, 29, 30]. For all of the Pu-115 superconductors (Fig. 3), fits of $1/T_1$ to a 'dirty d-wave'model, indicate that $2\Delta/k_BT_c$ = 5-8. Muon spin rotation measurements have also been performed for $PuCoGa_5$ and confirm the temperature dependences measured by NQR [32, 33]. Further, measurements of the in-plane magnetic-field penetration depth suggest d-wave pairing in $PuCoGa_5$ and yield an estimate of zero-temperature penetration depth, $\lambda(0)$ of 241 nm [32]. After approximately one year of aging due to self irradiation, the penetration depth in $PuCoGa_5$ increases to 498 nm [33].

4. Normal State Properties

Because of their relatively high T_c and the general interest in the electronic structure of plutonium, a number of groups using a variety of theoretical and computational approaches have explored the electronic structure of

	T_c (K)	γ_n (mJ/mol K^2)	$H_{c2}^{a,b}$ (T)	H_{c2}^{c} (T)	$\partial H_{c2}^{a,b}/\partial T$ (T/K)	$\partial \mathrm{H}_{c2}^{c}/\partial T$ (T/K)	$\xi_0^{a,b}$ ($\mathring{A}$)	ξ_0^{c} ($\mathring{A}$)
$PuCoGa_5$	18.5	~ 80	~ 120	~ 100	-10	-8	16	18
$PuRhGa_5$	8.6	~ 50	31	17	-3.5	-2	35	45
$PuCoIn_5$	2.5	200	~ 35	9.5	-17.6	-13.2	33	38
$PuRhIn_5$	1.7	350	~ 23	6.5	-20.4	-13	37	46

Table 2: Superconducting properties of plutonium superconductors. Notes: γ_n: Sommerfeld coefficient at T_c; $H_{c2}^{a,b}$ (H_{c2}^{c}): upper critical field in the a-b plane (parallel to the c-axis) extrapolated to $T = 0$; $\partial H_{c2}/\partial T$: slope of the upper critical field near T_c; ξ_0: Ginzburg-Landau superconducting coherence length at $T = 0$. Data from [3, 13, 34] and references therein.

the Pu-based superconductors [35, 36, 37]. The general picture that emerges from this diversity of approaches is i) a Fermi surface that is quasi two-dimensional, although not as cylindrical as the Ce-115 materials, and ii) an f-electron configuration that is partially localized and of mixed valence character. The normal state physical properties of the Pu-115 superconductors measured experimentally are consistent with this picture.

Early photoemission spectroscopy measurements on $PuCoGa_5$ showed that Pu's 5f electrons occurred in two configurations, the first being itinerant and occurring at the Fermi energy and the second being localized a little more than 1 eV below the Fermi energy [38]. This behavior is consistent with that which has been observed in Ga-stabilized δ-Pu. More recent resonant x-ray emission spectroscopy measurements on $PuCoGa_5$ and $PuCoIn_5$ confirm this view and further reveal an f electron configuration that includes an admixture of $5f^5$, $5f^4$, $5f^6$ states, with $PuCoGa_5$ displaying greater mixed valence character than $PuCoIn_5$ [39].

Significant efforts have been made to experimentally observe the Fermi surface of the Pu-115 superconductors, especially because of the insights that have been revealed from such measurements in the Ce-115 materials [40]. Unfortunately, thesel efforts have not yet been successful, due in part to the ingrowth of irradiation damage that begins upon initial solidification of the material, shortening the time available to conduct such measurements before disorder obscures the desired quantum oscillations.

Consistent with this picture of the f-electron configuration of the Pu-115 materials, the electrical resistivity of $PuCoGa_5$ is weakly temperature dependent at high temperature before developing a curvature consistent with mixed

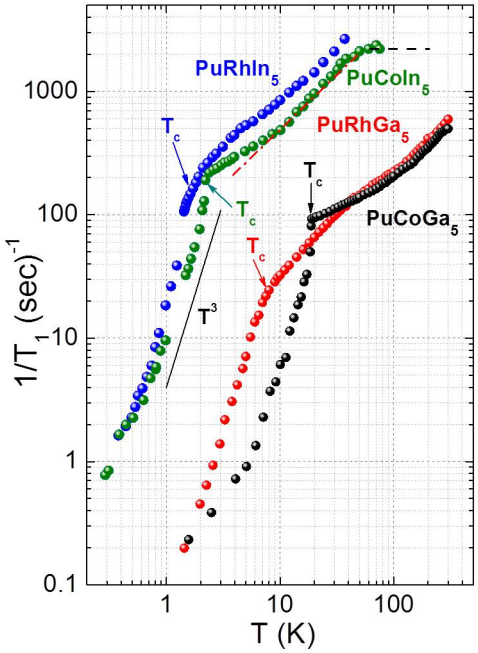

Figure 3: Spin-lattice relaxation rate $1/T_1$ versus T for each of the known plutonium superconductors. The solid black line is a T^3 power law, reflective of the observed behavior below T_c. The normal state behavior is approximately Korringa-like at intermediate temperatures above T_c.

valence materials below 150 K. At lowest temperatures, $\rho(T)$ assumes an unusual power-law T-dependence: $\rho \sim T^{4/3}$ [1] that also appears in $PuRhGa_5$ [11]. $\rho(T)$ of $PuCoIn_5$ and $PuRhIn_5$ is nearly temperature independent above 150 K before passing through a maximum around 100 K and then decreasing rapidly at lower temperatures, again consistent with mixed valence or Kondo lattice behavior [3]. At lowest temperature, $\rho(T) \sim T^n$ with n = 1 and 4/3 for $PuRhIn_5$ and $PuCoIn_5$, respectively, power-laws that are again distinctly different from the expected T^2 Fermi liquid dependence. Insufficient data exist to make strong statements about the implied non-Fermi liquid behavior in all of the Pu-115 superconductors. However, such behavior often occurs in nearly-magnetic materials, as these plutonium materials appear to be, given their normal state electronic configuration and potentially d-wave superconducting state.

Bulk magnetic susceptibility has been measured in $PuRhIn_5$, $PuRhGa_5$, and $PuCoGa_5$. In $PuRhIn_5$, $\chi(T)$ follows a modified Curie-Weiss behavior above 100 K with an effective moment $\mu_{eff} = 0.85\ \mu_B$ ($0.3\ \mu_B$) and an antiferromagnetic Weiss temperature Θ = -120 K (-7 K) for a magnetic field perpendicular (parallel) to the c-axis [3]. In contrast to $PuRhIn_5$, $\chi(T)$ is only weakly anisotropic in $PuRhGa_5$ [9] despite a Curie-Weiss behavior that is consistent with a Pu^{3+} configuration. In $PuCoGa_5$ polarized neutron measurements of the microscopic magnetization reveal a small and weakly temperature dependent magnetic moment [41], in contrast to initial bulk measurements that revealed Curie-Weiss behavior [1], perhaps consistent with the mixed valence configuration inferred from x-ray emission spectroscopy.

NMR and NQR measurements have also been performed in the normal state of each of the Pu-115 superconductors (Figure 3). In $PuCoIn_5$ $1/T_1$ is essentially temperature independent above 60 K, indicating the presence of weakly coupled 5f magnetic moments and anticipating that the bulk magnetic susceptibility will display Curie-Weiss behavior as in $PuRhIn_5$ [29]. Between 10 to 40 K, Korringa-like relaxation, $1/T_1 \sim T$, appears and is followed at lower temperatures by a faster relaxation rate, perhaps due to the presence of antiferromagnetic spin fluctuations. This scenario could also explain the anomalous low-temperature behavior of the electrical resistivity discussed above. $PuRhIn_5$ spin-lattice relaxation behavior is similar to that of $PuCoIn_5$ [30]. $1/T_1$ in $PuCoGa_5$ and $PuRhGa_5$ are similar in temperature dependence and notably slower in magnitude than their In-based counterparts. Below $\sim$ 30 K, $PuCoGa_5$ displays a temperature dependence that is decidedly not Korringa-like [27].

5. Perspective

By most accounts, the superconductivity observed in the Pu-115 materials is not conventional, i.e., an explanation of observed properties in terms of phonon-mediated pairing consistent with the BCS theory is incomplete. As discussed above, the temperature dependence of the spin-lattice relaxation rate and the magnetic penetration depth below T_c yield power-law behavior consistent with d-wave pairing. Further, point-contact spectroscopy reveals a four-fold modulation of the superconducting gap. However, relative to e.g., the high-T_c cuprates, the direct evidence of d-wave superconductivity is rather limited.

In the case of the Ce-115 materials, the evidence for d-wave superconductivity is quite strong, and the numerous similarities between the Ce-115 and Pu-115 materials at least suggest a similar origin of superconductivity. In both the Ce-115 and Pu-115 materials, a universal structural tuning is found in which T_c varies linearly with the c/a, the ratio of the tetragonal lattice constants, even in heavily alloyed materials in which the naive expectation would be that superconductivity is destroyed [17]. Further, the individual Pu-115 superconductors can be mapped, based on their unit cell volumes, onto a generalized Doniach phase diagram that is analogous to that observed in Ce-115 and suggests that the superconducting compounds exist in close proximity to magnetic order [12]. This proximity to magnetism is also often used to explain the unit cell volume of elemental plutonium

If one assumes that the superconductivity is mediated by spin fluctuations, a comparison can be made between superconducting T_c and a characteristic spin fluctuation temperature T_{sf} inferred from normal state properties [27, 44]. In this scenario, a linear relationship between T_c and T_{sf} is observed among the plutonium superconductors, but more dramatically (Figure 4), the Pu-115 materials appear to bridge a gap in T_c and T_{sf} between more traditional heavy fermion superconductors and the cuprates.

While the strong analogies with the Ce-115 materials and the overall correlation with other heavy fermions and cuprates are suggestive, it would seem premature to conclude that superconductivity in the Pu-115 materials is mediated solely by spin fluctuations. Similarly, one should not assume that all of the Pu-115 materials have the same pairing mechanism given that, e.g., $PuCoIn_5$ and $PuCoGa_5$ have rather different normal state magnetic susceptibilities. In particular, the fact that Pu has multiple 5f electrons and they appear to exist in an admixture of 5f states suggests that more complex valence fluctuations might be responsible for mediating superconductivity [39, 42]. Theoretical work is ongoing to explore such scenarios, including critical valence fluctuations and/or density fluctuations as the origins of pairing [44, 43, 45, 46, 47]. In these scenarios, the characteristic temperature in Figure 4 may be more general than a spin fluctuation scale and characterize a Kondo or valence fluctuation energy. A full exploration of these theoretical approaches and their normal- and superconducting-state signatures might provide insight not only to plutonium superconductors but also to broader classes of strongly correlated materials.

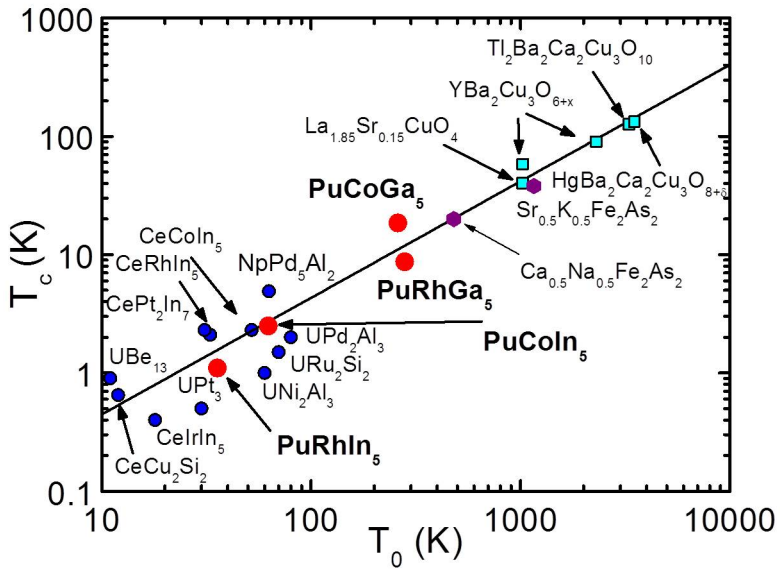

Figure 4: Superconducting transition temperature T_c versus a characteristic temperature T_0 for several families of superconductors. The plutonium superconductors occupy a region of T_c and T_0 that are intermediate between heavy fermion superconductors and high-T_c cuprates.

6. Conclusion

The superconductivity that has been observed in the Pu-115 family of compounds raises as many questions as it answers. Qualitatively, the physical properties of these materials display many attributes that are consistent with other strongly correlated superconductors and also embody many of the mysteries of elemental plutonium and its allotropes. A scenario in which unconventional superconductivity emerges from a strongly-correlated mixed valent normal state at relatively high temperature given measured fluctuation energies is plausible. However, quantitatively, much more work remains to definitively prove a pairing mechanism, e.g., via a full suite of phase-sensitive measurements, in these materials. Further, the phase space of plutonium intermetallic compounds has only just begun to be explored. While only four plutonium-based superconductors are known, they represent a larger fraction of all known Pu-based intermetallic compounds than e.g., in transition metal or rare earth compounds. Fundamentally, this should leave one optimistic

that further discoveries await that will increase the number of known unconventional superconductors and clarify the enigma that is the electronic and metallic properties of elemental plutonium.

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