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Superconductivity in plutonium compounds

J.L. Sarrao *, E.D. Bauer, J.N. Mitchell, P.H. Tobash, J.D. Thompson

Los Alamos National Laboratory, Los Alamos, NM 87545, United States

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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 $PuRhIn_5$.

Contents

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 [2–4]. 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 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 $PuCoGa_5$. Fig. 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].

* Corresponding author. Tel.: +1 505 667 6645.
E-mail address: sarrao@lanl.gov (J.L. Sarrao).

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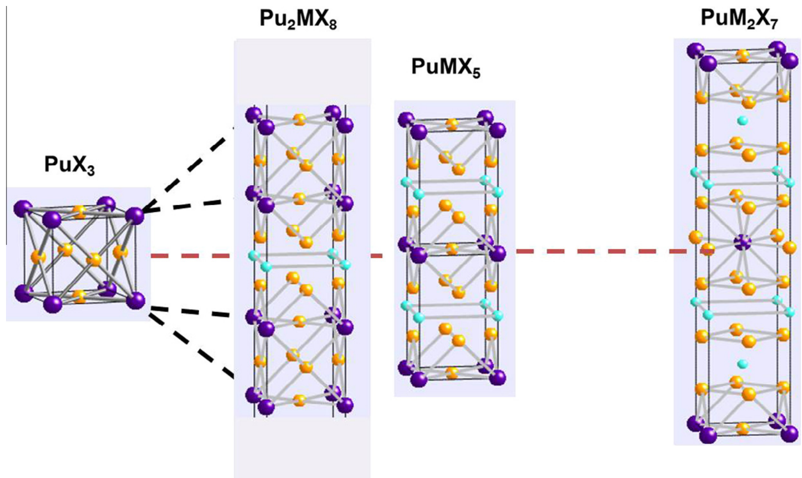

Fig. 1. An illustration of the stacking orders that give rise to the Pu-115, Pu-218, and Pu-127 stoichiometries. M is a transition metal, and X can be Ga or In.

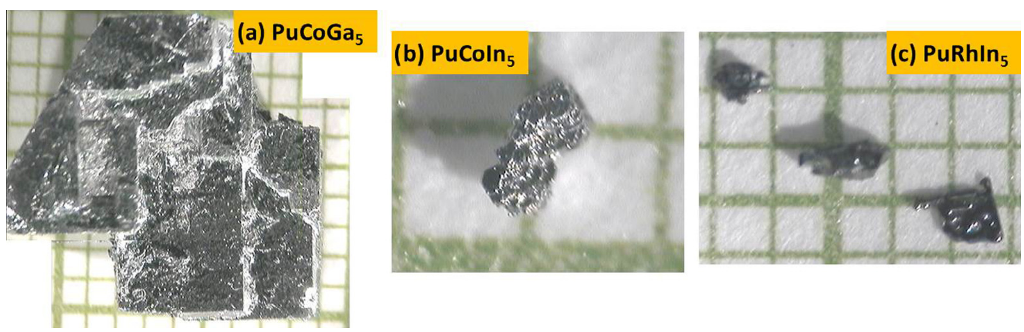

Fig. 2. Pictures of representative crystals of known plutonium superconductors. The grid lines in the images are a millimeter scale.

Table 1
Superconducting transition temperature, lattice constants, and original reference for known plutonium superconductors.

	$PuCoGa_5$	$PuRhGa_5$	$PuCoIn_5$	$PuRhIn_5$
T_c (K)	18.5	8.6	2.5	1.7
Lattice constant (Å)	a = 4.232	a = 4.301	a = 4.574	a = 4.621
	c = 6.786	c = 6.857	c = 7.439	c = 7.460
Discovery	Sarrao et al. [1]	Wastin et al. [11]	Bauer et al. [12]	Bauer and Thompson [3]

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 Fig. 2 [13]. The structural and ground state properties of these 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 quasi-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].