Discovery of plutonium-based superconductivity

2003 J. Phys.: Condens. Matter 15 S2275

(http://iopscience.iop.org/0953-8984/15/28/368)

J. Phys.: Condens. Matter **15** (2003) S2275–S2278 PII: S0953-8984(03)62549-X

Discovery of plutonium-based superconductivity

J L Sarrao[1], J D Thompson[1], N O Moreno[1], L A Morales[1], F Wastin[2], J Rebizant[2], P Boulet[2], E Colineau[2] and G H Lander[1,2]

[1] Los Alamos National Laboratory, Los Alamos, NM 87545, USA
[2] European Commission, JRC, Institute for Transuranium Elements, Postfach 2340, 76125 Karlsruhe, Germany

E-mail: sarrao@lanl.gov

Received 12 November 2002
Published 4 July 2003
Online at stacks.iop.org/JPhysCM/15/S2275

Abstract
The discovery of superconductivity in single crystals of $PuCoGa_5$ with transition temperature $T_c = 18.5$ K is discussed. The existing data lead to the speculation that the superconductivity in $PuCoGa_5$ may be unconventional. In such a scenario the properties of $PuCoGa_5$ would be intermediate between those of isostructural $UCoGa_5$ and $CeCoIn_5$, more heavily studied f-electron materials.

1. Introduction

Plutonium is a fascinating metal whose 5f electrons are poised on the boundary between localized and itinerant behaviour. This instability gives rise to an extremely complex metallurgy [1] and challenges the state of the art in electronic structure calculations [2]. The crossover from localized to itinerant f-electron behaviour is also central to the phenomenology of heavy-fermion compounds [3].

Here, we discuss a recently discovered microcosm of the fascinating properties of plutonium: the discovery of superconductivity in $PuCoGa_5$ at 18.5 K [4]. Not only is this a rather high T_c for an intermetallic compound, but also there is at least the suggestion that this superconductivity may be unconventional and, perhaps, spin-fluctuation mediated [5, 6].

2. Evidence for superconductivity in $PuCoGa_5$

Large single crystals of $PuCoGa_5$ have been grown from an excess Ga flux. Further and independently, single-crystal platelets have been obtained by arc-melting and subsequent annealing. The physical properties of these materials are identical and reveal bulk superconductivity near 18.5 K [4]. In both cases, single-crystal structural determinations have been made. One finds that $PuCoGa_5$ crystallizes in the $HoCoGa_5$ crystal structure with tetragonal lattice constants $a = 4.232$ Å and $c = 6.786$ Å. This is the same crystal

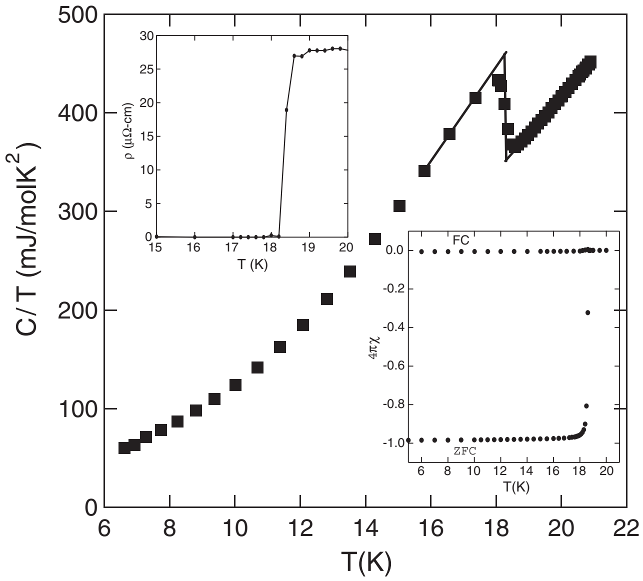

Figure 1. Evidence for superconductivity in $PuCoGa_5$. The main body of the figure shows heat capacity plotted as C/T versus T. The upper and lower insets show electrical resistivity and magnetic susceptibility, respectively, as a function of temperature. In all cases signatures of a phase transition are observed in the vicinity of 18.5 K.

structure in which $CeMIn_5$ (M = Co, Rh, Ir), a family of unconventional heavy-fermion superconductors [7], and $UMGa_5$ [8] also crystallize.

In figure 1 we show the evidence for bulk superconductivity in $PuCoGa_5$. A transition to zero resistance, coincident with full-shielding diamagnetism, is observed near 18.5 K. At this same temperature, a step-like transition in heat capacity is observed. If one assumes the BCS value of $\Delta C/\gamma T_c = 1.43$, then one infers from these data that γ, a measure of the conduction electron contribution to the low-temperature heat capacity, is 77 mJ mol^{-1} K^{-2}. This value of γ is enhanced relative to that expected for normal metals and is suggestive of heavy-fermion behaviour.

Interestingly, the T_c of $PuCoGa_5$ decreases from its initial value of ∼18.5 K as a function of time at a rate of approximately 0.2 K per month. This decrease would seem to be a result of radiation-induced self-damage associated with the spontaneous decay of ^{239}Pu. This mechanism is further indicated by the fact that the critical current, J_c, actually increases with time over the same period [4].

A correspondingly large value of the upper critical field H_{c2} in $PuCoGa_5$ has been inferred [4]. In particular, field-dependent resistivity data yield an initial slope of $\mathrm{d}H_{c2}/\mathrm{d}T$ of −59 kOe K^{-1}. From this value, one can estimate an upper critical field of 740 kOe. Further, one can estimate the BCS coherence length and therefore the Fermi velocity, and find that $\gamma \sim 60$ mJ mol^{-1} K^{-2} in the free-electron limit. Similarly, from estimates of the thermodynamic critical field, one can estimate $\gamma \sim 70$ mJ mol^{-1} K^{-2}, assuming the BCS value for the condensation energy. Thus, one has several independent estimates of $\gamma \sim 100$ mJ mol^{-1} K^{-2} in $PuCoGa_5$. Although this is a rather small value compared to those for other heavy-fermion superconductors, it is significantly enhanced compared to that for $UMGa_5$ [8], in which no superconductivity is observed.

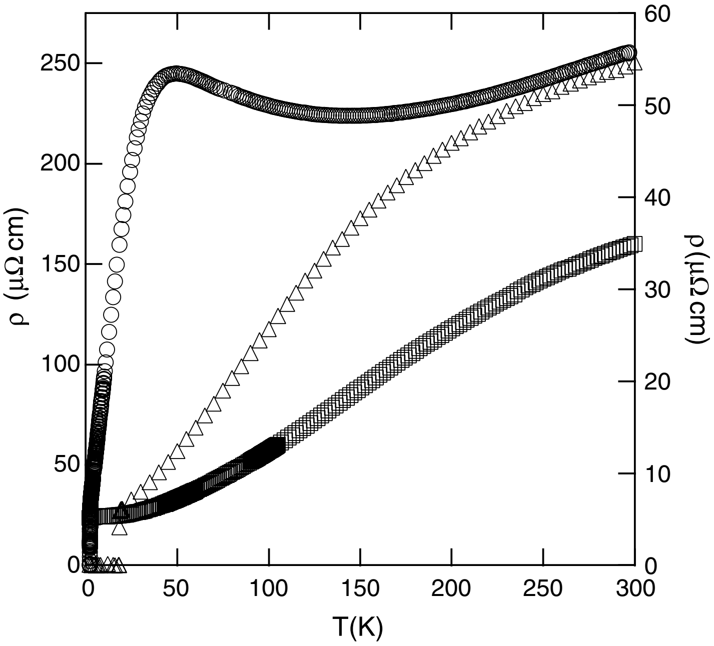

Figure 2. Resistivity as a function of temperature for $CeCoIn_5$ (circles), $PuCoGa_5$ (triangles), and $UCoGa_5$ (squares). The data for $CeCoIn_5$ are plotted using the right axis, whereas $PuCoGa_5$ and $UCoGa_5$ use the left.

3. Trends in normal-state properties

Normal-state properties provide further evidence that $PuCoGa_5$ displays stronger electron correlation effects than $UCoGa_5$. Figure 2 displays the electrical resistivity for $CeCoIn_5$, $PuCoGa_5$, and $UCoGa_5$. Judging from the characteristic change in curvature of the temperature dependence of the resistivity, one can see in figure 2 that this temperature scale is higher in $PuCoGa_5$ than in $CeCoIn_5$, but not as high as in $UCoGa_5$. This trend can be confirmed from heat capacity measurements for these compounds, which find that the linear-in-temperature coefficient of the low-temperature heat capacity, γ, increases from $\sim$10 mJ mol^{-1} K^{-2} for $UCoGa_5$ to $\sim$100 mJ mol^{-1} K^{-2} for $PuCoGa_5$ to $\sim$1000 mJ mol^{-1}K^{-2} for $CeCoIn_5$ [7].

From these data one is led to the conclusion that the superconductivity in $PuCoGa_5$ may be unconventional. In such a scenario, the order-of-magnitude-higher T_c for $PuCoGa_5$ as compared to $CeCoIn_5$ ($T_c = 2.3$ K) [7] would be expected from the increase in bandwidth in going from 5f electrons to 4f electrons [9, 10]. It is generally understood that 4f electrons have a greater degree of localization than do 5f electrons, as deduced, for example, from the evolution of the Wigner–Seitz radius as a function of atom across the lanthanide/actinide families [11].

Although the suggestion of unconventional superconductivity in $PuCoGa_5$ may seem implausible, the alternative, an 18 K conventional, phonon-mediated superconductor, is equally challenging. For $PuCoGa_5$, magnetic susceptibility measurements reveal Curie–Weiss behaviour consistent with a paramagnetic moment of $\sim$0.7 μ_B/Pu [4]. The pair-breaking tendency of magnetic moments would suggest that $UCoGa_5$, a temperature-independent paramagnet, would have a higher T_c than $PuCoGa_5$, in contrast to what is observed.

4. Summary

We have discussed the observation of superconductivity above 18 K in $PuCoGa_5$. The speculation that this might be unconventional, spin-fluctuation-mediated superconductivity raises the possibility that $PuCoGa_5$ could be an intellectual bridge between the known heavy-fermion superconductors (with characteristic $T_c \sim 1$ K) and the high-T_c cuprates (with characteristic $T_c \sim 100$ K). Thus, the transuranics may represent a particularly fertile, if unploughed, field for the discovery of additional superconductors.

Acknowledgments

We thank Z Fisk for fruitful discussions. Work at Los Alamos was performed under the auspices of the US Department of Energy.

References

[1] Hecker S S 2001 *MRS Bull.* **26** 672
[2] Savrasov S Y, Kotliar G and Abrahams E 2001 *Nature* **410** 793
[3] Smith J L, Fisk Z and Hecker S S 1985 *Physica* B + C **130** 151
[4] Sarrao J L, Morales L A, Thompson J D, Scott B L, Stewart G R, Wastin F, Rebizant J, Boulet P, Colineau E and Lander G H 2002 *Nature* **420** 297
[5] Opahle I and Oppeneer P M 2002 *Phys. Rev. Lett.* **90** 157001
[6] Hotta T and Ueda K 2002 *Preprint*
[7] Petrovic C, Pagliuso P G, Hundley M F, Movshovich R, Sarrao J L, Thompson J D, Fisk Z and Monthoux P 2001 *J. Phys.: Condens. Matter* **13** L337
[8] Grin Yu, Rogl P and Hiebl K 1986 *J. Less-Common Met.* **121** 497
[9] Mathur N D, Grosche F M, Julian S R, Walker I R, Freye D M, Haselwimmer R K W and Lonzarich G G 1998 *Nature* **394** 39
[10] Monthoux P and Lonzarich G G 2001 *Preprint* cond-mat/0207556
Monthoux P 2002 *J. Phys.: Condens. Matter* **15** S1973
[11] Wills J M and Eriksson O 2002 *Los Alamos Sci.* **26** 128