LA-UR- 02-7370

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Submitted to: Proceedings of ASR-2002 Advances in the Physics of f-electron

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Discovery of Plutonium-Based Superconductivity

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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 behavior. 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 behavior 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. 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 $\AA$ and c=6.786 $\AA$. This is the same crystal structure in which $CeMIn_5$ (M=Co, Rh, Ir), a family of unconventional heavy fermion superconductors[7], and $UMGa_5$[8] also crystallize.

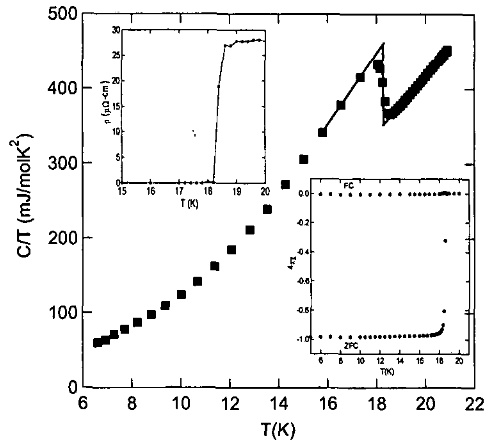

Figure 1. Evidence for superconductivity in $PuCoGa_5$. The main body of the figure shows heat capacity plotted as C/T versus T. The upper and lower insets show electrical resistivity and magnetic susceptibility, respectively, as a function of temperature. In all cases signatures of a phase transition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

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/molK^2. This value of γ is enhanced relative to that expected for normal metals and is suggestive of heavy fermion behavior.

Interestingly, the T_c of $PuCoGa_5$ decreases from its initial value of $\sim$ 18.5 K as a function of time at a rate of approximately 0.2 K/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 dH_{c2}/dT of -59 kOe/K. 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/molK^2 in the free-electron limit. Similarly,

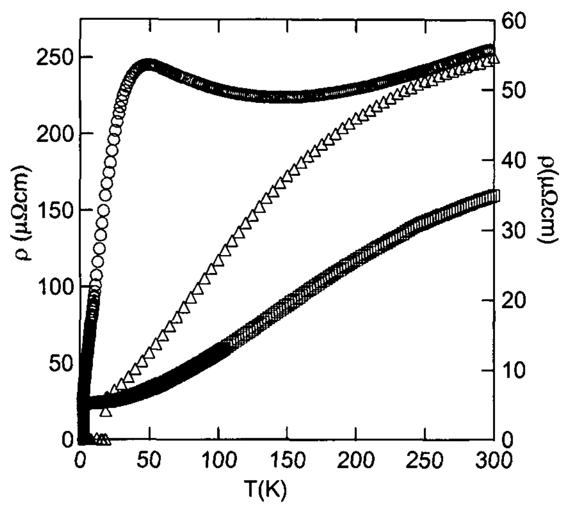

Figure 2. Resistivity as a function of temperature for $CeCoIn_5$ (circles), $PuCoGa_5$ (triangles), and $UCoGa_5$ (squares). The data for $CeCoIn_5$ are plotted on the right axis, whereas $PuCoGa_5$ and $UCoGa_5$ use the left.

from estimates of the thermodynamic critical field, one can estimate $\gamma \sim 70$ mJ/molK^2, assuming the BCS value for the condensation energy. Thus, one has several independent estimates of $\gamma \sim 100$ mJ/molK^2 in $PuCoGa_5$. Although this is a rather small value compared to other heavy fermion superconductors, it is significantly enhanced compared to $UMGa_5$[8], in which no superconductivity is observed.

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 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 in these compounds, which find that the linear-in-temperature coefficient of the low-temperature heat capacity, γ, increases from ~ 10 mJ/molK^2 in $UCoGa_5$ to ~ 100 mJ/molK^2 in $PuCoGa_5$ to ~ 1000 mJ/molK^2 in $CeCoIn_5$[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 in $PuCoGa_5$ 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, by 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. In $PuCoGa_5$, magnetic susceptibility measurements reveal Curie-Weiss behavior consistent with a paramagnetic moment of $\sim 0.7\ \mu_B$/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 unplowed, field for the discovery of additional superconductors.

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

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