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LETTER TO THE EDITOR

Electronic band structure of $PuCoGa_5$

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Abstract

The electronic band structure is presented for $PuCoGa_5$, the recently discovered superconductor with $T_C \approx 18$ K. The band structure is calculated by the tight-binding linear muffin-tin orbital method in the atomic sphere approximation.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

It has recently been discovered [1] that $PuCoGa_5$ is superconducting with a transition temperature $T_C \approx 18.5$ K. This transition temperature is much higher than that in known superconducting U compounds, and at ∼18 K it is at the upper limit for phonon-mediated superconductivity. Therefore it has been suggested [1] that the mechanism of superconductivity in $PuCoGa_5$ might be unconventional. To understand the properties of $PuCoGa_5$, information about its electronic structure is important. Since the 5f electrons of Pu are strongly correlated and their behaviour is borderline between a localized and itinerant character [1], results from current methods of computing the electronic structure have to be used with care. An account of many-body effects in the electronic structure is difficult and has only been solved for a limited number of systems. Such an analysis was successful for metallic δ-plutonium [2]. The calculated density of states (DOS) shows a Kondo-like peak at the Fermi level, which is consistent with experimental data from photoemission. It was concluded [2] that the f electron in δ-Pu is slightly on the delocalized side of the localization–delocalization transition.

In $PuCoGa_5$ it is not known to what extent the f electrons are itinerant and therefore whether we can rely on the current calculation methods based upon density functional theory (DFT). However, experience with some uranium compounds, even where is strong evidence of localization of some of the U 5f electrons [3], shows that DFT can account for the observed valence band of the x-ray photoemission spectra (see e.g. [4]).

Therefore it is useful to calculate the band structure of this fascinating compound using a standard method, although one must remember that some many-body features, such as the Abrikosov–Kondo peak at E_F, will not appear.

Table 1. Crystallographic characteristics for $PuCoGa_3$ [1] ($UCoGa_3$, [12]): space group $P4/mmm$; lattice constants $a = 4.232$ Å (4.233 Å), $c = 6.786$ Å (6.723 Å); average Wigner–Seitz radius 1.6016 Å (1.6063 Å).

Atom	Site	x	y	z	Wigner–Seitz radius (Å)
Pu(U)	1a	0	0	0	1.9705(1.9766)
Co	1b	0	0	1/2	1.4912(1.4890)
Ga1	1c	1/2	1/2	0	1.6534(1.6165)
Ga2	4i	0	1/2	0.312(0.3048)	1.5010(1.5004)

Table 2. DOS at the Fermi energy for $PuCoGa_5$ decomposed into contributions from s, p, d and f electrons and from U, Co and Ga atoms. The total DOS gives the number of states per eV and per formula unit (fu) whereas partial DOSs are numbers of states per eV per atom.

	Pu	Co	Ga1	Ga2	Total (per fu)
s	0.007	0.005	0.020	0.013	0.084
p	0.016	0.070	0.282	0.180	1.088
d	0.147	1.665	0.049	0.022	1.949
f	23.462	—	—	—	23.462
Total	23.632	1.740	0.351	0.215	26.583

2. Method of calculation

The electronic band structure of $PuCoGa_5$ was calculated by the tight-binding linear muffin-tin orbital (TB-LMTO) method in the atomic sphere approximation (ASA) [5, 6]. The crystal structure of $PuCoGa_5$ is tetragonal, the space group is $P4/mmm$ and the room-temperature lattice constants are $a = 4.232$ Å and $c = 6.786$ Å [1]. The positions of atoms taken from [1] are collected in table 1 together with the Wigner–Seitz radii S_j used for the band structure calculations. There are two inequivalent positions of Ga atoms in the unit cell. The following atomic configurations were assumed: core $+6p^6 5f^6 7s^2$ for Pu, core $+3d^7 4s^2$ for Co and core $+4s^2 4p^1$ for Ga. The unit cell is filled with Wigner–Seitz spheres, their total volume being equal to the volume of the unit cell; the overlap volume of the muffin-tin spheres is 12.5%. The standard combined corrections for overlapping were used to compensate the ASA errors [5].

Fully relativistic treatments of the core electron and the scalar relativistic approximation for the valence electrons were employed [7]. The exchange-correlation potential of Perdew *et al* [8] with nonlocal corrections was used. The computations were done for 3927 k-points in the irreducible wedge (1/16) of the Brillouin zone. Integration in the k-space was made by the tetrahedron method [9–11]. The self-consistency of the energy values up to the error of 0.01 mRyd was achieved by iterations.

3. Results and conclusions

The calculated electronic structure is summarized in the form of the DOS plots in figures 1 and 2(a)–(c). Values of the DOS at the Fermi level (E_F) are collected in table 2.

The total DOS for $PuCoGa_5$ shows features common to many UTX_5 compounds: a narrow peak of width ∼0.5 eV from 5f electrons and a band about 3 eV wide from d and p electrons. What is unusual is the deep minimum at E_F of the DOS from s electrons. It is thus striking that s electrons contribute only 0.3% to the total DOS (E_F).

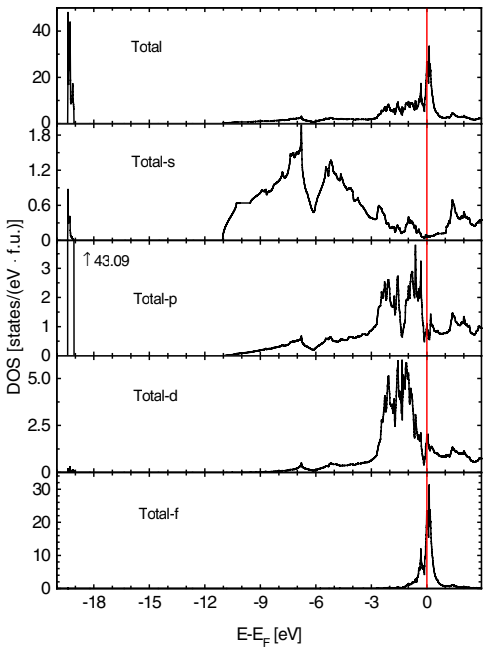

Figure 1. Calculated total and partial DOSs for $PuCoGa_5$.

Table 3. Number of states for $PuCoGa_5$ and its projections on s, p, d and f states and types of atoms.

	Pu	Co	Ga1	Ga2	Total (per fu)
s	0.549	0.687	1.425	1.262	7.709
p	6.294	0.938	1.513	1.353	14.157
d	1.994	7.888	0.151	0.112	10.481
f	5.653	—	—	—	5.653
Total	14.490	9.513	3.089	2.727	38.000

The projected DOSs for Pu, Co and Ga atoms (in both crystallographic positions) are presented in figures 2(a), (b) and (c) respectively.

The numbers of states provided in table 3 reveal large charge transfers between states, which can be understood as effects of strong hybridization of s p d atomic levels.

Calculations give low values of DOS for the presumably most mobile s and p electrons so the number of electric current carriers is relatively low. This observation is compatible with rather high normal state resistivity of $PuCoGa_5$ [1]. The total DOS $(E_F) = 26.583$ states (eV fu)$^{-1}$ can be used to find the bare value of the Sommerfeld coefficient $\gamma_{th} = 62$ mJ mol^{-1} K^{-2}. This value is smaller than the experimental value $\gamma = 77$ mJ mol^{-1} K^{-2} reported in [1].

The fundamental question of how well the true energy structure of a system with correlated electrons is described by band structure calculations based on the DFT does not have a universal answer. To shed some light on the validity of the present results we calculated the electronic structure by the same method for the compound $UCoGa_5$ which is isostructural to $PuCoGa_5$ and has very similar values for the lattice constants. The crystallographic data taken from [12]

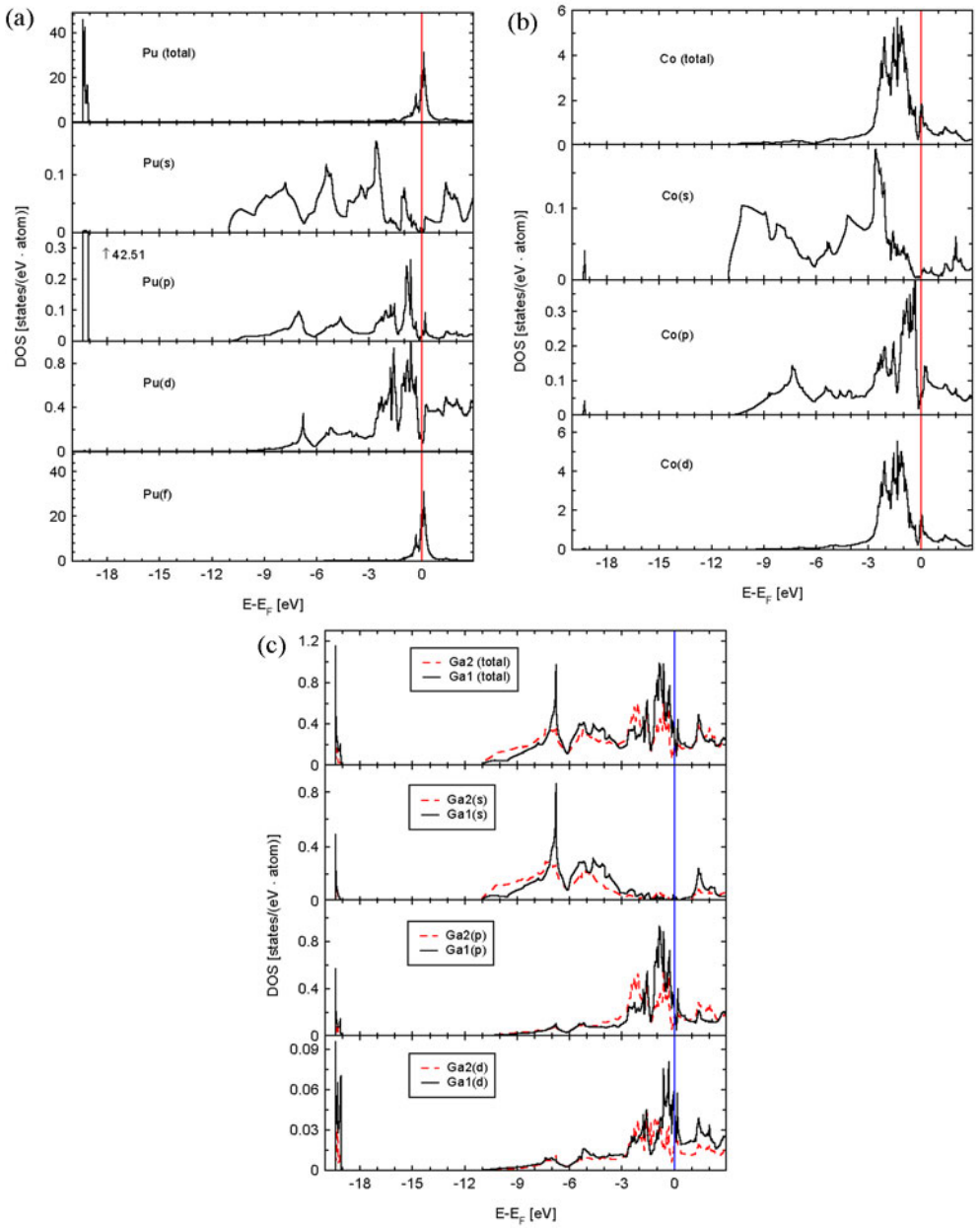

Figure 2. (a) Contributions from Pu atoms to DOS. (b) Contributions from Co atoms to DOS. (c) Contributions to DOS from Ga atoms in the positions (1c) (Ga1, full curve) and (4i) (Ga2, broken curve).

are collected in table 1 in parenthesis. The total DOSs of $UCoGa_5$ and $PuCoGa_5$ are compared in figure 3 in the region near E_F. The significant differences are, understandably, only in the positions of the 5f bands, lying higher in energy in the uranium compound and being mostly unoccupied.

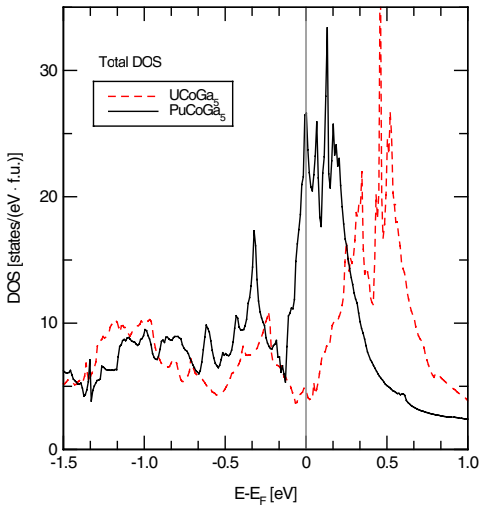

Figure 3. Comparison of calculated total DOSs for $PuCoGa_5$ and isostructured $UCoGa_3$ in the vicinity of the Fermi energy.

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