The magnetic ground state and anisotropic property of UCoGa and UCoAl

Ying Xu, Zhi Zeng *

Institute of Solid State Physics, Chinese Academy of Sciences, Hefei 230031, China

Abstract

The first-principles electronic structure calculations for the uranium intermetallic compounds UCoGa and UCoAl are performed by means of scalar-relativistic full-potential linear augmented plane wave (FP-LAPW) and fully relativistic FP-LAPW method taking into account of spin–orbit coupling. The itinerant ferromagnet of UCoGa is obtained, which is in accordance with the experiments. The hybridization of 5f–p and 5f–d as well as bonding properties of conductive electron bands are discussed by comparing the density of states and charge density of these two isoelectronic compounds.

Both UCoGa and UCoAl belong to the ternary uranium equiatomic compounds of the UTX (X = a metal of third or fourth group of the periodic table, and T a transition metal) and crystallize in ZrNiAl structure. The ZrNiAl structure has layered structure constructed by two types of basal plane alternating along the c-axis. One layer consists of uranium atoms and one third of T atoms while the other layer consists of the remaining T atoms and X atoms. This layered structure is indeed ready for strong anisotropy. UCoAl has received many attentions due to the meta-magnetic transition with the very low critical field ($B_c = 0.7$ T) and this metamagnetism is uniaxial [1,2]. Experiments demonstrate that in the UCoAl$_{1-x}$Ga$_x$ compounds ferromagnetism has been achieved by appropriate Ga doping ($x \geq 0.2$) [3]. As an isoelectronic compound of UCoAl, UCoGa orders ferromagnetically at low temperature with magnetic moments stacked along the c-axis [4]. Electronic structure analysis for UCoAl has been described in [5]. In this paper we will present the new results of the first principles calculations for UCoGa and compare it with that of UCoAl. Our attention will be focused on the 5f-ligand hybridization and spin–orbit coupling (SOC) upon them which brings out a great difference between UCoGa and UCoAl.

We’ve used the FP-LAPW method as implemented in WIEN2k code [6], in which the spin–orbit coupling is treated by the second variational method [7]. In both compounds the nearest U atoms are within the basal plane and the distances are 3.458 and 3.466 Å for UCoGa and UCoAl, respectively. In 5f-electron metallic systems, actinide–actinide spacing $d_{AC}$ is the principal parameter influencing anomalous properties. Hill has shown that, for $d_{AC} < 3.5$ Å, local moments are quenched due to interatomic f–f overlap. Here, that both the nearest interuranium distances are of the order of Hill limit indicates that in both compounds the 5f states are at the border of itinerant and localization.

Our calculation for UCoGa is firstly performed by scalar-relativistic spin-polarized self-consistence where SOC is ignored. Spin moments for U, Co1, Co2 are 1.39, 0.39, 0.25, respectively. There are 0.57 μB spin moments in the interstitial region. The total and partial density of states at Fermi level $N(E_F)$/f.u. of 17 states/eV

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*Corresponding author.
E-mail address: zzeng@theory.issp.ac.cn (Z. Zeng).

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mainly (76%) result from U-5f states, which is in the same magnitude with $N_c(\varepsilon_F)$ of 20 states/eV measured from electronic heat coefficient. In order to probe the role of SOC, fully relativistic calculations are performed. When the magnetization direction is allowed to be parallel to c-axis, calculated spin and orbital moments are listed in Table 1. The smaller orbital moments 1.12\(\mu_B\) of U than free U\(^{3+}\) and U\(^{4+}\) reflect the itinerancy of 5f states. This is in accordance with the trend that strong 5f-ligand hybridization leads to delocalized 5f states. When the magnetization direction lies in the a-b plane, about 8.6 meV magnetocrystalline anisotropy energy is gained which is consistent with the experiments that c-axis is the easy magnetism direction. Orbital moments reduce to 0.77\(\mu_B\) and spin moments remain the same.

Table 1
The spin and orbital moments as the results of fully relativistic calculations, experiment values also shown

<table>
<thead>
<tr>
<th></th>
<th>U</th>
<th>Co1</th>
<th>Co2</th>
</tr>
</thead>
<tbody>
<tr>
<td>(M_S) ((\mu_B/f.u.))</td>
<td>1.11</td>
<td>-0.26</td>
<td>-0.11</td>
</tr>
<tr>
<td>(M_L) ((\mu_B/f.u.))</td>
<td>-1.23</td>
<td>-0.04</td>
<td>-0.03</td>
</tr>
<tr>
<td>(M_{\text{interstitial}}) ((\mu_B/f.u.))</td>
<td>0.45</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(M_{\text{Net}}) ((\mu_B/f.u.))</td>
<td>0.47</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Exp ((\mu_B/f.u.))</td>
<td>0.78</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

To investigate the nature of the states around \(E_F\), the charge density contours of UCoGa and UCoAl are compared. In the basal plane containing U and Co1 atoms, the charge density contours of UCoGa and UCoAl are almost the same, which reflects the interaction between U and Co atoms has the similar behavior both for UCoAl and UCoGa. While in [010] plane involving Co2 and Ga (or Al) atoms, we find that the hybridization of f-p in UCoGa is stronger than that in UCoAl. The nearest distances of U-X are 3.00 Å in UCoGa and 3.03 Å in UCoAl, which means the difference of distance is negligible. Therefore the strong f-p hybridization of UCoGa is attributed to the large atom sphere and strong electronegetivity of Ga atom. The Ga-doping-induced meta-magnetic transition in UCoAl can be viewed as strong hybridization of Ga-p states with U-5f states which leads to the reduction of density of states at Fermi energy. This situation results in the resolvent of strong spin-fluctuation in UCoAl and the strong correlated f states are weakened by energy gain from hybridization. We think this is the reason why the ground state of UCoGa is stable ferromagnetic.

In summary, we have performed the electronic structure calculations for UCoGa and UCoAl. The ferromagnetic ground state for UCoGa is obtained. By comparing the charge density of these two compounds, it is found that the strong U-5f and Ga-p hybridization reduces the density of states at \(E_F\) and results in the resolvent of spin fluctuation in UCoAl.

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References