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Electronic Structure and Magnetic Properties of the $UCoAs_2$ Compound

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The $UCoAs_2$ compound crystallizes in the tetragonal $HfCuSi_2$ type structure with space group $P4/nmm$. The compound orders ferromagnetically at 150 K with a spontaneous magnetic moment of about 1.8 μ_B per formula unit. We present results of fully relativistic band structure calculations based on the full-potential local-orbital minimum-basis scheme (FPLO-5.10-20) and compare magnetic moments obtained from calculations without and with orbital polarization corrections. The magnetic behavior of the Co atoms remains unknown.

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1. Introduction

Preparation and first experimental studies of novel compound $UCoAs_2$ were reported by Kaczorowski et al. [1]. $UCoAs_2$ is a representative of ternary pnictides with chemical formula UTX_2, where T stands for a transition metal atom and X stands for pnictogen (N, P, As, Sb, Bi). UTX_2 compounds crystallize in the tetragonal $HfCuSi_2$ type structure with space group $P4/nmm$. Most of UTX_2 order ferro- or antiferromagnetically at relatively high temperatures 100–250 K [2–4] and have strongly anisotropic magnetic properties. These compounds behavior is predominated especially by the uranium $5f$ electrons, which show character between the localized $4f$ and itinerant $3d$ ones. Ternary uranium compounds have also demonstrated range of unique properties such as spin fluctuations, heavy fermions or superconductivity.

$UCoAs_2$ orders ferromagnetically at 150 K with a spontaneous magnetic moment of about 1.8 μ_B per formula unit [1]. The question is the division of partial magnetic moments. Contribution from $5f$ electrons of uranium atoms seems to be undisputed, but the character of Co atoms remains unknown. Mostly in ternary uranium compounds which contain the transition metal atoms only the uranium atoms carry magnetic moments [2]. However, like for example in $UCo_{2-x}P_2$ [5, 6] and $UCo_{2-x}Sn_2$ [7] also the Co atoms have contribution to magnetization.

2. Details of the calculations

In order to study electronic structure of $UCoAs_2$ we used improved version of the full-potential local-orbital minimum-basis scheme (FPLO-5.10-20) [8] based on the local spin density approximation (LSDA) [9]. The spin polarized fully relativistic calculations were carried out for the tetragonal structure with 8 atoms per unit cell (two formula units). We used the Wyckoff positions and the lattice constants measured by using the single-crystal X-ray diffraction method [1]. We assumed the basis set definition like listed below. For U atoms: core (electrons up to $5p$) + semi core ($5d6s6p$) + valence ($7s7p6d5f$); Co: $(1s2s2p) + (3s3p) + (4s4p3d)$; As: $(1s2s2p3s) + (3p) + (4s4p3d)$. The calculations were performed for the reciprocal space mesh containing 2176 points (division $30 \times 30 \times 30$) within the irreducible wedge of the Brillouin zone using the tetrahedron method for integrations [10]. The LSDA exchange-correlation potential was used in the Perdew and Wang form [11]. Computations were done without and with orbital polarization corrections [12, 13]. The self-consistent criterion was equal to 10^{-8} Ry for the total energy.

3. Results and discussion

We present results of fully relativistic band structure calculations based on the FPLO code. The spin and partial resolved densities of states (DOS) are presented in Figs. 1 and 2. We tried several different procedures to obtain results in best agreement with the experiment. We present results which are obtained without and with orbital polarization corrections (OPC), and also without and with the initial spin splitting (IS). An important determinant of correctness was the total magnetic moment, which was estimated experimentally from the field dependence of the magnetization on pressed powder sample [1] and is equal to 1.8 μ_B per formula unit.

Calculations without OPC and without IS are presented on the left sides in Fig. 1 and Table I. After the comparison of received total magnetic moment equal to

0.36 μ_B and experimental one 1.8 μ_B, we decided to discard these outcomes.

3.1. The initial spin splitting

Ab initio calculations sometimes lead to local energy minimum (like in case presented above). To find global solution we started to deal with parameter called "initial spin-split". Spin polarized calculations were initialized with unbalanced spin occupation on particular atoms, but without "fixed spin moments". We have tried several configurations. We got the best results (minimum total energy) from starting point: +1.5 μ_B on U and −1.5 μ_B on Co atoms. These results are presented on

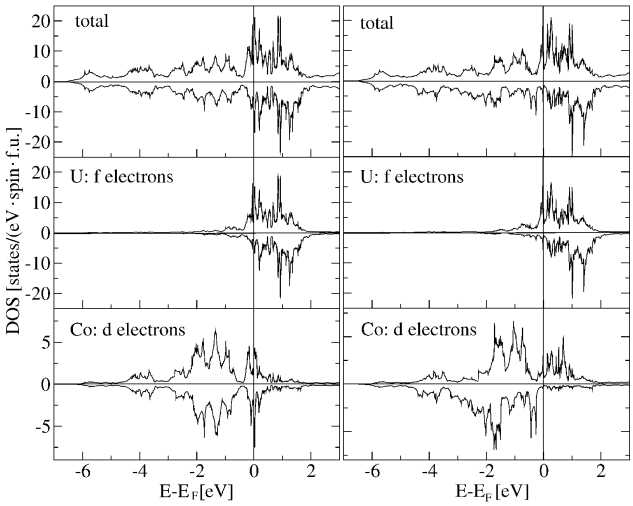

Fig. 1. Spin and component resolved densities of states for $UCoAs_2$ obtained by LSDA calculations, without (left) and with (right) the initial spin splitting.

TABLE I

The magnetic moments obtained by LSDA calculations, without (left) and with (right) the initial spin splitting. The spin, orbital and total local magnetic moments in the Bohr magnetons per atom. The spin, orbital and total magnetic moments in the Bohr magnetons per formula unit are presented.

Atom	m_{spin}	m_{orb}	m_{tot}	m_{spin}	m_{orb}	m_{tot}
U	0.72	–1.08	–0.35	1.12	–1.68	–0.55
Co	0.02	0.01	0.03	–0.95	–0.16	– 1.10
As	–0.02	0.00	–0.02	–0.03	–0.01	–0.03
As	–0.01	–0.01	–0.01	–0.10	0.01	–0.09
formula	0.71	–1.07	–0.36	0.06	–1.83	–1.80

the right sides in Fig. 1 and Table I. The total magnetic moment 1.78 μ_B is close to experimental one 1.8 μ_B. The main difference from results without the initial spin splitting is rather big magnetic moment on Co atoms 1.1 μ_B. The magnetic behavior of the Co atoms seems to be a key to proper theoretical description of $UCoAs_2$.

3.2. The orbital polarization corrections

Corresponding results obtained using OPC [14] are presented. Like in previous calculations we performed two cases: without and with the IS. Results of calculations with IS are presented on the right sides of Fig. 2 and Table II. LSDA+OPC+IS configutration leads to the total magnetic moment equal to 3.2 μ_B which disagree with the experimental value. We decided to discard also this outcome.

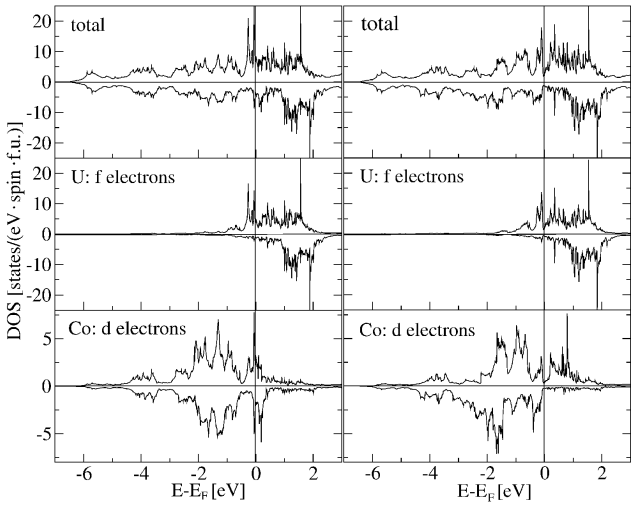

Fig. 2. Spin and component resolved densities of states for $UCoAs_2$ obtained by LSDA+OPC calculations, without (left) and with (right) the initial spin splitting.

TABLE II

The magnetic moments obtained by LSDA+OPC calculations, without (left) and with (right) the initial spin splitting. The spin, orbital and total local magnetic moments in the Bohr magnetons per atom. The spin, orbital and total magnetic moments in the Bohr magnetons per formula unit are presented.

Atom	m_{spin}	m_{orb}	m_{tot}	m_{spin}	m_{orb}	m_{tot}
U	1.57	–3.39	–1.83	1.49	–3.29	–1.80
Co	0.10	0.01	0.11	–1.00	–0.30	–1.30
As	–0.03	–0.02	–0.05	–0.03	–0.02	–0.06
As	0.00	–0.02	–0.02	–0.11	0.00	–0.11
formula	1.63	–3.42	–1.78	0.35	–3.61	–3.26

On the left side in Fig. 2 and Table II we present results with OPC and without IS (LSDA+OPC). OPC increased the local magnetic moments both U and Co atoms, and, as a result, for the second time we got the total magnetic moment of about 1.78 μ_B almost equal to the experimental value. The correctness of results cannot be longer distinguished upon the value of the total magnetic moment. In good agreement with the

experiment we got also moments from LSDA+IS and LSDA+OPC calculations. From the comparison of total energies we have known however that the lowest energy value is for LSDA+OPC calculations (without the initial splitting). The total energy difference between LSDA+IS and LSDA+OPC is 20 mHa, which means that the OPC result is energetically the most stable, which also means that we will look closer on this outcome. On the left side in Fig. 2 spin and partial resolved plots of densities of states (DOS) are presented. Relatively wide band has formed up to 6 eV below the Fermi level. The contribution from U f electrons and Co d electrons is the most important for this band, contribution from As electrons is minor. The total DOS on the Fermi level is equal to 300 states/Ha which leads to theoretical Sommerfeld coefficient equal to 26 mJ mol^{-1} K^{-2}, the main contribution is provided by U ($5f$) and Co ($3d$) electrons. The total magnetic moment is predominantly located on U atoms 1.83 μ_{B} (including the orbital contribution), much weaker and opposite moment is on Co atoms 0.1 μ_{B}, contributions from As atoms are even smaller.

4. Summary and conclusions

We selected the result without IS and with OPC as the most reasonable describes properties of $UCoAs_2$. It is shown that applying the orbital polarization corrections to spin polarized band structure calculations leads to good agreement with the experimental values of the magnetic moment. The magnetic contribution from $5f$ electrons of uranium atoms seems to be clear, but the character of Co atoms remains unknown. The X-ray or neutron diffraction spectroscopy could confirm presence or absence of magnetic moment on Co atoms.

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