

The Electronic and Magnetic Properties of UGe Compound

M. WERWIŃSKI AND A. SZAJEK

Institute of Molecular Physics, Polish Academy of Sciences, M. Smoluchowskiego 17, 60-179 Poznań, Poland

The UGe system crystallizes in the orthorhombic ThIn-type structure. The uranium atoms occupy three crystallographic sites with interatomic U–U distances like below and above the Hill limit. The band structure has been calculated by using the density functional theory and generalized gradient approximation. *Ab initio* calculations were performed based on the full-potential local-orbital minimum-basis code. Calculations showed that all three types of uranium atoms are magnetically ordered with antiparallel alignment of the magnetic moments. For uranium atoms with an interatomic distance below the Hill limit magnetic moments are significantly reduced due to hybridization effects.

PACS numbers: 71.20.–b, 75.25.–j

1. Introduction

The recent reinvestigation of the U–Ge system has finally established existence of six compounds with the chemical formulas U_5Ge_4 , UGe, U_3Ge_5 , UGe_2 , UGe_3 and UGe_{2-x} (existing in a homogeneity range of $0.30 < x < 0.35$) [1]. The existence of the uranium monogermanide UGe was first time reported by Alcock and Grievson [2]. The UGe compound crystallizes in the orthorhombic ThIn-type structure (Pbnm space group) [3]. Both uranium and germanium atoms occupy three crystallographic positions: U1(4d), U2(4d), U3(4c), and Ge1(4d), Ge2(4d), Ge3(4c). The U3–U3 distance between uranium U3 atoms equal to 2.92 Å is below the Hill limit (~ 3.4 Å). The U3 atoms form metallic chains along the *c* axis. Magnetic measurements indicate the Curie–Weiss type behavior down to 1.7 K [3]. The average effective moment is equal to $2.47 \mu_B/U$ atom. There is no magnetic ordering transition down to 1.7 K. Earlier calculations [4] showed that all three types of uranium atoms are magnetically ordered.

The aim of this work is to give insight into the magnetic structure of the UGe compound based on the modern *ab initio* spin polarized band structure calculations.

2. Details of calculations

The full potential local-orbital minimum-basis code (FPLO [5], v. 9.00-33) was employed. The DFT calculations were performed without and with the orbital polarization (OP) correction [6]. We used experimental lattice constants and the Wyckoff positions [3]. The unit cell contains 24 atoms. The generalized gradient approximation (GGA) [7] was used to the potential parameterized in the form proposed by Perdew et al. [7]. The tetrahedron method was employed for the Brillouin-zone integration [8]. The self-consistent criterion for the total energy was equal to 10^{-8} Ha, a criterion for charge was equal to 10^{-6} electrons.

3. Results and discussion

The results of calculations without spin polarization are shown in Fig. 1. The valence bands have gap between -4.7 and -6.7 eV. The subbands between -10.8

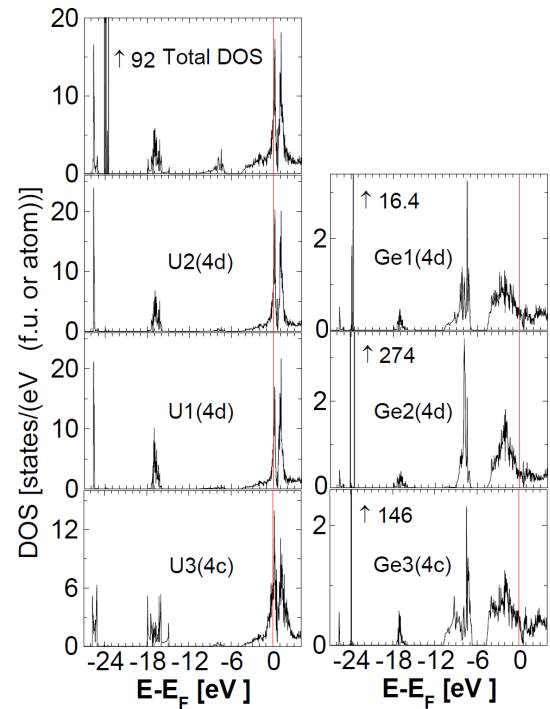


Fig. 1. The DOS plots without spin polarization (total and site projected).

and -8 eV are formed mainly by Ge(4*s*) electrons. Above the gap the main contribution is provided by Ge(4*p*) and U(7*s*, 6*d*, 5*f*) electrons. The higher binding spectra are formed by U(6*p*_{1/2}) and U(6*p*_{3/2}) electrons located at -25.5 and -17 eV. Because of stronger hybridization the U(6*p*_{3/2}) peak is more broadened than U(6*p*_{1/2}). At -24 eV are two very narrow and high peaks: Ge(3*d*_{3/2}) and Ge(3*d*_{5/2}). U(5*f*) electrons provide the main contribution to the total DOS at the Fermi level (about 95%) (see Table). The Sommerfeld coefficient γ_0 is equal to 19.4 mJ/(mol K²). Unfortunately experimental value is not known.

The spin polarized values of γ_0 are much lower. Total energy calculations showed that magnetic solution is more stable by about 46.4 meV/f.u. Spin projected DOS

TABLE
DOS at the Fermi level [states/(eV spin (f.u. or atom))] and local magnetic moments m [μ_B /atom]. Calculations performed without (NSP) and with (SP) spin polarization, the later also with orbital corrections (OP) applied to $5f$ states. The Sommerfeld coefficient γ_0 [mJ/(mol K²)] in the linear term of the specific heat.

Atom (site)	NSP DOS	SP DOS	m	SP+OP DOS	m
U1(4d)	4.68	3.32/0.99	0.05	2.22/0.71	-1.42
U2(4d)	4.31	1.18/4.36	-0.29	0.75/3.70	1.48
U3(4c)	2.77	3.39/2.31	-0.02	3.21/0.92	-0.38
Ge1(4d)	0.20	0.14/0.18	-0.02	0.12/0.18	-0.08
Ge2(4d)	0.16	0.15/0.14	0.03	0.17/0.16	-0.03
Ge3(4c)	0.24	0.19/0.24	-0.01	0.17/0.22	-0.01
total (per f.u.)	4.12	2.79/2.75	-0.09	2.21/1.96	-0.15
γ_0	19.4	13.1	-	9.8	-

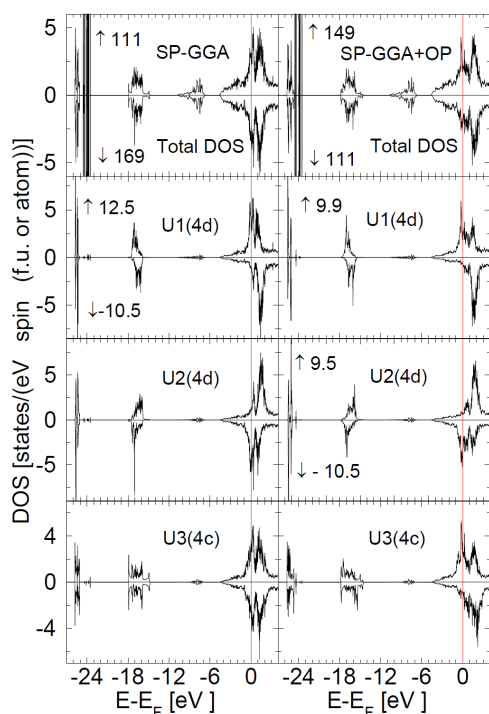


Fig. 2. The spin polarized (SP-GGA) DOS plots (total and site projected) within the GGA approach, also including OP corrections to the $5f$ states (SP-GGA+OP).

plots within GGA approach are presented in Fig. 2. The large spin magnetic moments are located on two uranium atoms: U1(4d) and U2(4d). The total magnetic moments on these uranium atoms are equal to only 0.05 and $-0.29 \mu_B$ /atom. The short U3–U3 distance (below the Hill limit) indicates strong overlap of the corresponding $5f$ orbitals and significantly reduces the magnetic moment on U3 atoms. The U3($5f$) and U3($6p$) peaks are more broadened than for the U1 and U2 atoms.

The most important *ab initio* approach, applied by us to improve discrepancy between the GGA magnetic moments and experimental ones, takes into account so-called orbital polarization term as proposed by Eriksson et al. [6], implemented in FPLO code. Results are presented in the right part in Fig. 2. The OP corrections were added to the $5f$ states on uranium atoms. In all uranium atoms magnetic moments increased. The resultant moments are collected in Table. The OP approach gives moments on U3 atom much lower than for U1 and U2.

In spite of use more advanced FPLO method than in our previous work [4] we have to be aware of static DFT limitations in describing real ground state, which can be very much influenced by the pronounced fluctuations of magnetic moments (see for example paper by Prokeš et al. [9]). Such complex ground state can be better described by the dynamical mean field theory (DMFT) [10].

4. Conclusions

Presented results of the spin polarized band structure calculations, within GGA and GGA+OP approach, for UGe compound showed that on all three types of uranium atoms magnetic moments are formed. The moments on U1 and U2 atoms have an antiparallel alignment. The moment on U3 atom is reduced because of the short U3–U3 distance, which leads to the overlap of the corresponding $5f$ orbitals. The results can clarify the low value of the effective magnetic moment obtained by the susceptibility measurements.

Acknowledgments

This work was supported by MNiSW within grants No. NN202134933 and No. NN202288338.

References

- [1] R. Troć, H. Noël, P. Boulet, *Philos. Mag. B* **82**, 805 (2002).
- [2] D. Alcock, P. Grieverson, *J. Instrum. Methods* **90**, 304 (1961).
- [3] P. Boulet, A. Daoudi, M. Potel, H. Noël, *J. Solid State Chem.* **129**, 113 (1997).
- [4] A. Szajek, *J. Magn. Magn. Mater.* **272-276**, e347 (2004).
- [5] FPLO code by K. Koepnik, H. Eschrig, *Phys. Rev. B* **59**, 1743 (1999); <http://www.fplo.de>.
- [6] O. Eriksson, B. Johansson, M.S.S. Brooks, *J. Phys. C* **1**, 4005 (1989).
- [7] J.P. Perdew, K. Burke, M. Ernzerhof, *Phys. Rev. Lett.* **77**, 3865 (1996); *ibid.*, **78**, 1396 (1997).
- [8] P. Blöchl, O. Jepsen, O.K. Andersen, *Phys. Rev. B* **49**, 16223 (1994).
- [9] K. Prokeš, A. de Visser, Y.K. Huang, B. Fåk, E. Ressouche, *Phys. Rev. B* **81**, 180407(R) (2010).
- [10] G. Kotliar, S.Y. Savrasov, K. Haule, V.S. Oudovenko, O. Parcollet, C.A. Marianetti, *Rev. Mod. Phys.* **78**, 865 (2006).