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Non-magnetic behaviour of UFe_2Si_2 compound

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Abstract

The work reports the results of measurements of photoemission (XPS) spectra as well as the thermal dependence of the specific heat and electrical resistivity for UFe_2Si_2. The XPS experimental data are compared with the calculations of the density of states using the FPLO methods. The specific heat and electrical resistivity data show no existence of magnetic order to the lowest measurement temperature of 0.2 K. The electronic specific heat coefficient γ is equal to 17.5(1) mJ/mol K^2 excluding the heavy fermion behaviour. According to the XPS data and the band structure calculations there is no energy separation between U 5f and Fe 3d states. This observation implies a strong f–d hybridisation, which may be the reason for non-magnetic behaviour of the studied compound.

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1. Introduction

In recent decades, a great amount of research has been dedicated to the UT_2X_2 compounds (T = transition metal and X = Si or Ge). Most of these compounds crystallize in the tetragonal $ThCr_2Si_2$-type structure. These compounds are attractive because of the great variety of the magnetic properties including the heavy fermion, spin fluctuation, superconductivity and non-Fermi liquid behaviour were observed [1,2]. In majority of these compounds the uranium magnetic moments order ferro- or antiferromagnetically at low temperatures. The different properties are observed in UT_2Si_2 (T = Fe, Re and Os) and UFe_2Ge_2 compounds which were reported to be Pauli paramagnets [3–6].

To shed more light on the magnetic properties of UFe_2Si_2 the results of XPS photoelectron spectroscopy, thermal dependence of the specific heat and electrical resistivity are presented. The band structure calculations are reported as well.

2. Experiment and results

The experiments were carried out on polycrystalline bulk samples that were prepared as reported in our previous paper [4].

The electrical resistivity was measured using a standard four-probe ac method (at 100 Hz with a measuring current of 10 mA) in a ^{4}He gas-flow cryostat. Heat capacity measurements were carried out in a ^{3}He–^{4}He dilution refrigerator, using a semi-adiabatic calorimeter [7].

The XPS spectra were obtained at room temperature using the Leybold LHS10 photoelectron spectrometer with the Mg Kα ($h\nu$ = 1486 eV) radiation in a vacuum of about 10^{-9} mbar. The total energy resolution of the spectrometer with a hemispherical energy analyser was about 0.75 eV for Ag 3d. Binding energies were referred to the Fermi level (E_F = 0). The spectrometer was calibrated using the Cu $2p_{3/2}$ (932.5 eV), Ag $3d_{5/2}$ (368.1 eV) and Au $4f_{7/2}$ (84.0 eV) core-level photoemission lines. The Shirley method [8] was used to subtract background and then the experimental spectra were numerically fitted using the 80% Gaussian and 20% Lorentzian model.

The electronic structure was calculated by full-potential local orbital minimum-basic full-relativistic FPLO scheme including the spin–orbit coupling [9]. The band structure calculations were performed within local spin density approximation (LSDA) for the exchange correlation potential in the form proposed by Perdew and Wang [10]. The FPLO calculations were performed for 460 *k*-points in the irreducible Brillouin zone.

The temperature dependence of the electrical resistivity is shown in Fig. 1a. As one can see there is no anomaly indicative of magnetic ordering, however at temperature of about 8 K a Kondo-like minimum is visible. The latter observation supports data previously reported [6]. The resistivity of UFe_2Si_2 at 2 K can

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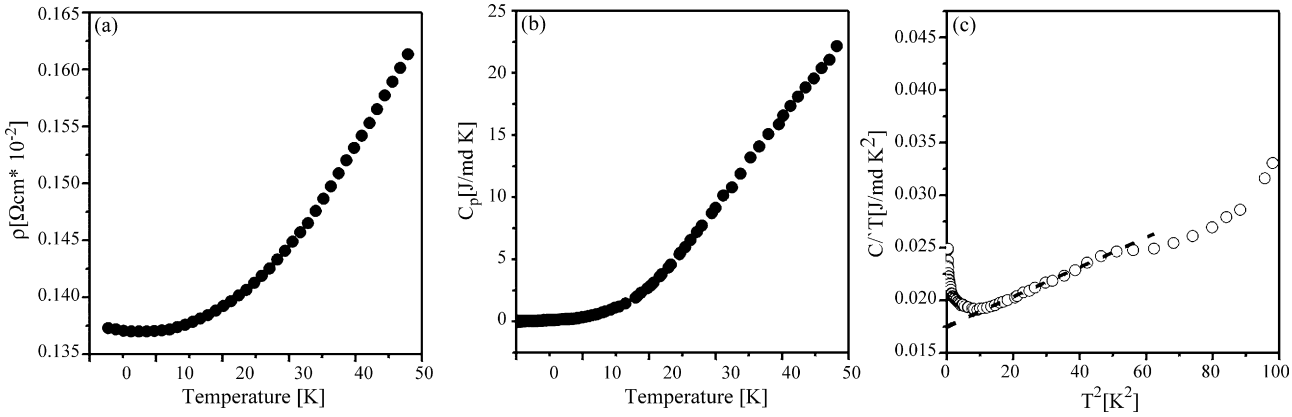

Fig. 1. Temperature dependence of the (a) electrical resistivity; (b) specific heat $C(T)$ and (c) C/T vs. T^2 plot for UFe_2Si_2.

be estimated to be 0.00137 Ω cm. In Fig. 1b the specific heat versus temperature is shown. No sign of magnetic ordering down to our lowest measurement temperature of 0.2 K was found. In the low temperature range (3.5–7 K) the specific heat can be approximated as

$$C = \gamma T + \beta T^3 \quad (1)$$

where γ is the Sommerfeld coefficient and β is a constant including the Debye temperature. For estimating the Debye temperature a plot of C/T versus T^2 in Fig. 1c is presented. As one can see a linear part within 12 and 45 K^2 is clearly visible. The γ parameter was found to be 17.5(2) mJ/mol K^2 on the other hand the β parameter was estimated to be about $1.41(3) \times 10^{-4}$ J/mol K^4. The latter value corresponds to the Debye temperature of about 410 K. This value derived from low temperature data seems to be overestimated when compared to values close to 200 K reported for isostructural compounds [11]. In Ref. [11] as many as 12 optical and 3 acoustic branches were taken into account, while in our case only a mean value of Debye temperature was estimated. When taking all 15 branches the mean Debye temperature can be calculated, this temperature is close to 323 K.

Similar values of the γ coefficient and Debye temperature have been observed also in isostructural UT_2Ge_2 (T = 3d elements) compounds [12]. In isostructural $LaFe_2Si_2$ the γ coefficient is equal to 22 mJ/mol K^2 [13].

The band structure calculations were based on the crystal structure determined previously [2]. Accordingly, in the crystal unit cell the atoms occupy the following Wyckoff positions:

- U atoms at 2a site: 0, 0, 0; 1/2, 1/2, 1/2;
- Fe atoms at 4d site: 0, 1/2, 1/4; 1/2, 0, 1/4; 0, 1/2, 3/4; 1/2, 0, 3/4;
- Si atoms in at site: 0, 0, z; 0, 0, $\bar{z}$; 1/2, 1/2, 1/2 + z; 1/2, 1/2, 1/2 − z with z equal to 0.376.

The calculated band structure is shown in Fig. 2. The inset of this figure shows a part of the Brillouin zone with characteristic points.

In Fig. 3 we present the total and partial density of states (DOS) for the particular atoms: U, Fe and Si, respectively. By inspecting Fig. 3 we can see that the occupied part of the DOS can be decomposed in the region near to the Fermi level into the 5f, 6d and 7s states of the uranium atoms, Fe 3d, 3p and 4s and Si 3p and 3d states. The U 5f states form a sharp peak on the Fermi level where the Fe 3d and U 6d states form a broad band. These results suggest possible hybridisation of U 5f and Fe 3d bands. The intensities of 7p and 7s uranium states as well as 4s iron and 3p silicon states are very small.

The XPS valence band of the UFe_2Si_2 compound is presented in Fig. 4. The experimental data can be directly compared to the calculated DOS by FPLO method.

The determined electronic structure of UFe_2Si_2 is different from those observed in isostructural UT_2Si_2 (T = Co, Ni, Cu) compounds [13]. For these compounds the U 5f level lies near the Fermi level whereas T 3d bands lie considerably below E_F. Consequently, between U 5f and T 3d states the energy separation exists. In all UT_2Si_2 (T = Co, Ni, Cu) compounds the uranium ions possess localized magnetic moments, since they order ferro- or antiferromagnetically with transitions temperatures near 100 K [2].

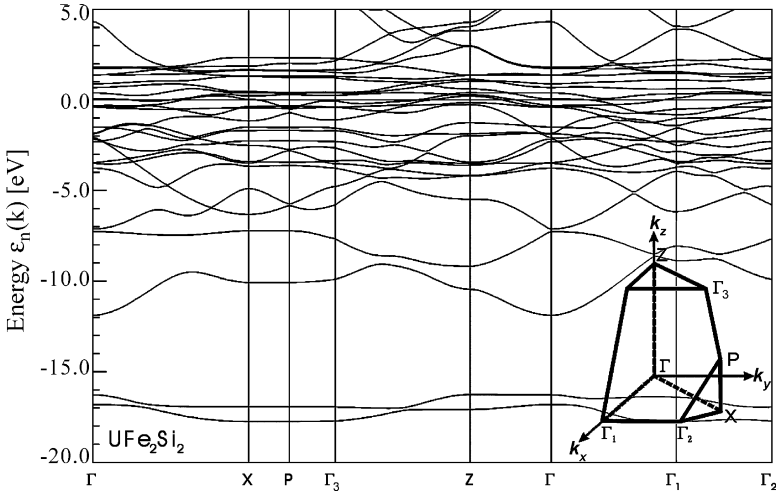

Fig. 2. Energy bands $\varepsilon_n(k)$ for UFe_2Si_2. High-symmetry directions in the Brillouin zone are marked. The Fermi energy is set to zero. The inset shows 1/16 of the Brillouin zone.

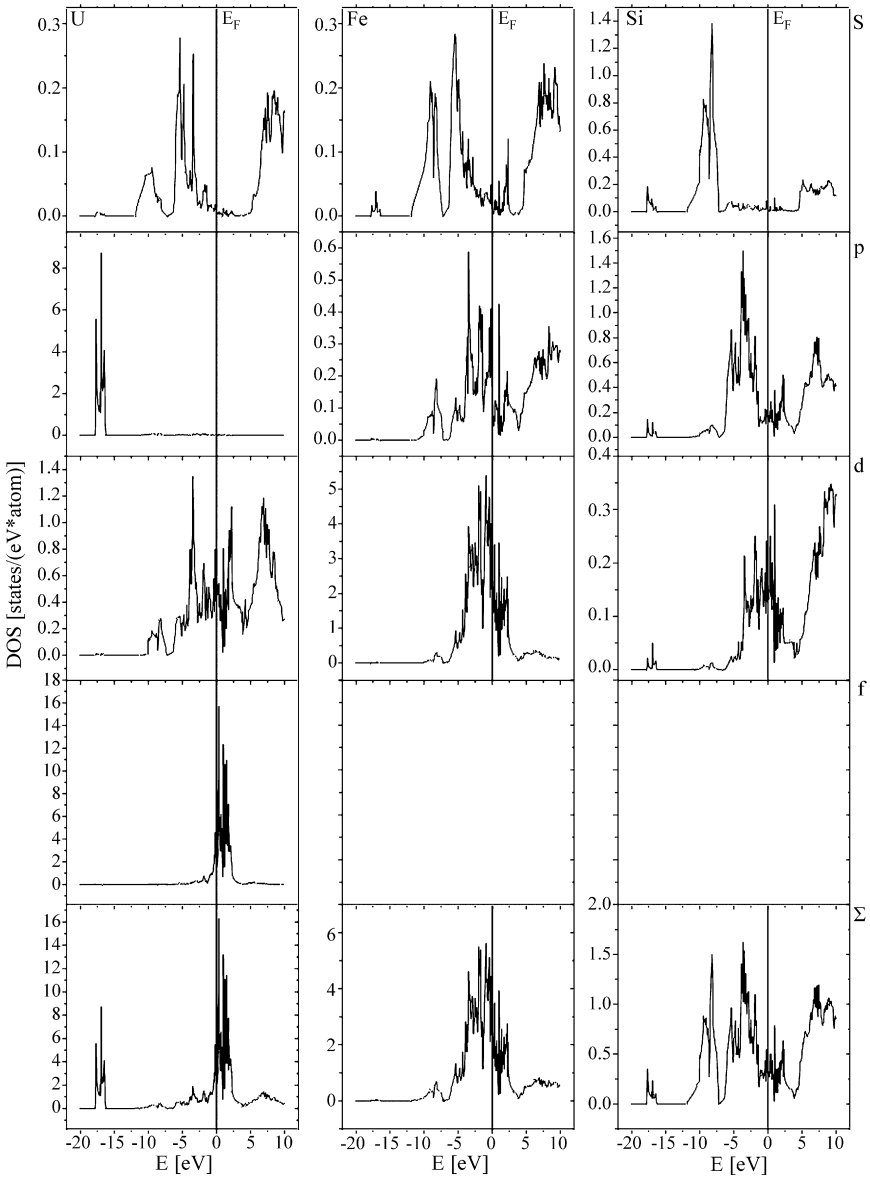

Fig. 3. Partial and total contributions to DOS for UFe_2Si_2.

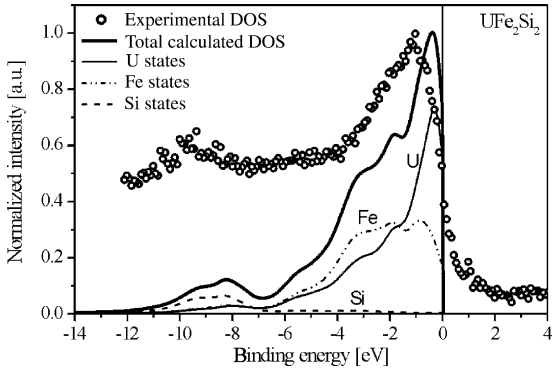

Fig. 4. Comparison of the measured and calculated XPS spectra of valence band at room temperature for UFe_2Si_2. The Fermi level located at $E = 0$ eV is marked by the vertical solid line. The calculations of the XPS spectra were performed using convolution of calculated DOS with Lorentzian profile of 0.4 eV FWHM. Appropriate cross-sections for photoionization has been taken into account [15].

3. Summary

The electrical resistivity and specific heat data, presented in this work, indicate that in the case of UFe_2Si_2 both U and Fe sublattices are nonmagnetic. Results of XPS measurements of the valence band and calculations of the density of states reveal that the U 5f and Fe 3d states are not separated from each other and both lie near the Fermi level. This seems to be indicative of a strong hybridisation of these states and could be the reason for the non-magnetic behaviour of the UFe_2Si_2 compound. Our conclusion may be additionally supported by means of the hybridisation matrix V_{df} calculated by Endstra et al. for the UT_2Si_2 series of compounds [14]. The authors estimated value of V_{df} for UFe_2Si_2 compound to be about 10% higher than for other isostructural UT_2Si_2 compounds [14].

References

[1] V. Sechovsky, L. Havela, in: E.P. Wohlfarth, K.H.J. Buschow (Eds.), Ferromagnetic Materials, vol. 4, North-Holland, Amsterdam, 1989.
[2] J. Leciejewicz, A. Szytuła, Magnetism of Ternary AuT_2X_2 Actinide Intermetallics, vol. 35, Folia Physica, Universitatis Jagellonica, 1993.
[3] T.T.M. Palsatra, A.A. Menovsky, G.J. Nieuwenhuys, J.A. Mydosh, J. Magn. Magn. Mater. 54–57 (1986) 435.
[4] A. Szytuła, S. Siek, J. Leciejewicz, A. Zygmunt, Z. Ban, J. Phys. Chem. Solids 49 (1988) 1113.
[5] A.J. Dirkmat, T. Endstra, E. Knetsh, A.A. Menovsky, J.A. Mydosh, J. Phys. 49 (1988) 475.
[6] A. Szytuła, M. Ślaski, B. Dunlap, Z. Sungaila, A. Umezawa, J. Magn. Magn. Mater. 75 (1988) 71.
[7] H.A. Kierstead, B.D. Dunlap, S.K. Malik, A.M. Umarjii, G.K. Shenoy, Phys. Rev. B 32 (1985) 135.
[8] D.A. Shirley, Phys. Rev. B 5 (1972) 391.
[9] K. Koepernik, H. Eschrig, Phys. Rev. B 59 (1999) 1743.
[10] J.P. Perdew, Y. Wang, Phys. Rev. B 45 (1992) 13244.
[11] P. Svoboda, J. Vejpravova, F. Honda, E. Santava, O. Schneeweiss, T. Komatsubara, Physica B 328 (2003) 139.
[12] T. Endstra, A.J. Dirkmaat, S.A.M. Mentink, A.A. Menovsky, G.J. Nieuwenhuys, J.A. Mydosh, Physica B 163 (1990) 309.
[13] M. Mihalik, V. Sechovsky, Physica B 359–361 (2005) 163.
[14] T. Endstra, G.J. Nieuwenhuys, J.A. Mydosh, Phys. Rev. B 48 (1993) 9595.
[15] J.J. Yeh, I. Lindau, Atomic Data Nuclear Data Tables 32 (1985) 1.