ELECTRONIC STRUCTURE OF $YbMn_2X_2$ (X=Si,Ge) COMPOUNDS *

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Electronic structure of the $YbMn_2X_2$ (X=Si, Ge) compounds with the $ThCr_2Si_2$-type structure have been investigated. The valence band and the core levels states are analyzed. The XPS valence band spectra are compared with the results of *ab initio* electronic structure calculations using the Tight-Binding Linear Muffin-Tin Orbital method (TB LMTO). The obtained results indicate that in $YbMn_2Ge_2$ the Yb ion is 2+ while in $YbMn_2Si_2$ is 3+. For both compounds the Mn3*d* states are close to the Fermi level. The calculated Mn moments are 1.93 μ_B for $YbMn_2Si_2$ and 2.30 μ_B for $YbMn_2Ge_2$.

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Rare-earth intermetallic compounds containing ytterbium exhibit a wide range of interesting and unusual physical and magnetic properties. This occurs mainly as a result of their mixed valence state (II/III) or changes from one valence state to another. The YbT_2X_2 (T is a *nd* transition metal, X is Si or Ge) compounds of the tetragonal $ThCr_2Si_2$-type structure (space group I4/mmm) are of particular interest with a range of effects reported

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earlier [1]. $YbMn_2Si_2$ is an antiferromagnet with ordered Mn sublattice (T_N=520 K) and an additional transition at ~30 K related to ordering in the Yb sublattice [2]. In $YbMn_2Ge_2$ at high temperatures the Mn magnetic moments order with a planar antiferromagnetic structure (T_N=510 K) while below T_t=185 K the change of the magnetic structure to the canted one is observed. There is no evidence for ordering in the Yb lattice down to 1.5 K [3]. Nowik *et al.* [4] have recently investigated magnetic phase transitions in the $YbMn_2Si_{2-x}Ge_x$ series by magnetisation and ^{57}Fe Mössbauer effect studies on ^{57}Fe doped samples.

This work reports the results of X-ray photoemission spectroscopy (XPS) measurements for $YbMn_2X_2$ (X=Si,Ge) compounds and pure Yb and Mn. The XPS valence band spectra are compared with *ab-initio* electronic structure calculations using the Tight-Binding Linear Muffin-Tin Orbital method (TB LMTO) [5]. On the basis of these results the electronic structure of the compounds have been determined.

The XPS spectra were obtained at room temperature using the Leybold LHS10 photoelectron spectrometer with the MgK_α radiation ($h\nu$ =1253.6 eV) and energy resolution about 0.9 eV for Ag3d. The vacuum during the measurements was about 10^{-9} Torr. Surfaces of the compounds were mechanically cleaned by scraping with diamond file in a preparation chamber under high vacuum. These procedure of cleaning was performed directly before each XPS measurement. The Shirley method [6] used to subtract the background from the experimental data and then the data were refined assuming 80% Gaussian and 20% Lorentzian contribution for each peak.

The electronic structure was calculated by *ab-initio* self-consistent Tight Binding Linear Muffin Tin Orbital method (TB LMTO) [5] within the framework of the local spin density approximation (LSD). The scalar-relativistic approximation for band electrons and the fully-relativistic treatment of the frozen core electrons were used. The exchange correlation potential was assumed according to von Barth and Hedin [7] with gradient corrections [8]. The self-consistent calculations were performed in the atomic sphere approximation (ASA) for the experimental values of the lattice parameters. The values of the atomic sphere radii were chosen in such a way that the sum of all the atomic sphere volumes was equal to the volume of the unit cell. In the band calculations, the initial atomic configurations were taken according to the Periodic Table of Elements. The energy was calculated for 817 k-points in the irreducible wedge of the Brillouin zone. The theoretical photoemission spectra were obtained from the calculated density of states (DOS) convolved by the Lorentzian with the half-width equal to 0.4 eV and scaled using the proper photoelectric cross-sections for partial states [9]. The XPS valence bands (VBs) between the Fermi energy (E_F=0 eV) and the binding energy of both compounds are shown in Fig. 1.

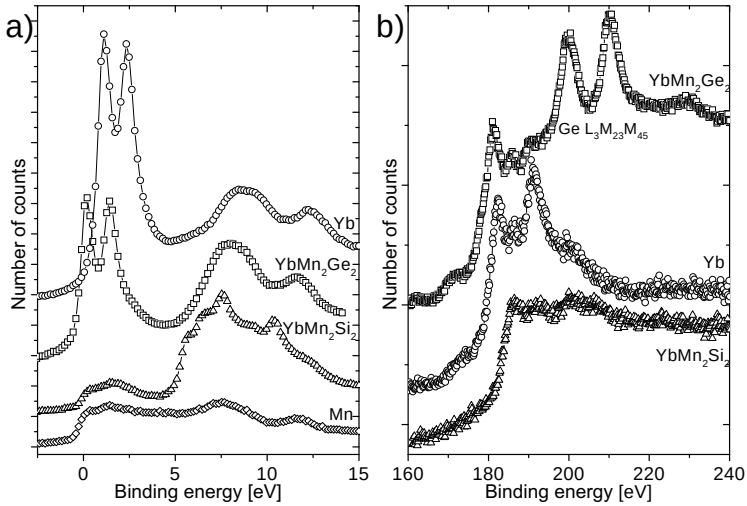

Fig. 1. XPS spectra of valence band (a) and Yb $4d$ core level (b) of $YbMn_2X_2$ compounds and the appropriate spectra for metallic Yb and Mn.

The obtained experimental data are compared with the calculated ones. The calculated partial and total density of states (DOS) for both compounds is presented in Fig. 2.

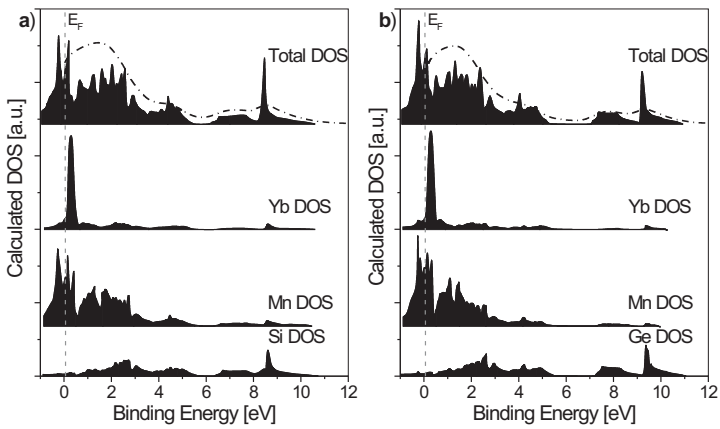

Fig. 2. Total density of states and the contribution from Yb, Mn and Si or Ge to the total density of states of paramagnetic $YbMn_2Si_2$ (a) and $YbMn_2Ge_2$ (b) (the Fermi level is located at $E = 0$ eV). The dash dot line presents the density of states convolved by Lorentzians of the half width of 0.4 eV, multiplied by the appropriate cross sections.

For $YbMn_2Si_2$ the calculated data (Fig. 2a) indicate that Yb $4f$ state is close to the Fermi level (0.2 eV). Close to the Fermi level are also two broad peaks: the first one intersects the Fermi level and the second one, between 0.5 and 3.0 eV, is connected with Mn $3d$ state. The Si $3p$ and Si $3s$ states are represented by two broad peaks in the energy region between 0.5 and 5.5 eV and between 6.5 and 10.5 eV, respectively. For $YbMn_2Ge_2$ the calculated data (Fig. 2(b)) give similar results for the Yb $4f$ and Mn $3d$ states. The Ge $4p$ state forms a broad peak between 0.5 and 5.5 eV while Ge $4s$ state between 7.5 and 11 eV. The experimental data give slightly different valence band structure for these compounds. In the case of $YbMn_2Si_2$ near the Fermi level only the broad peak corresponding to the Mn $3d$ states is detected between E_F and 4.5 eV. The spectrum of $YbMn_2Si_2$ between 5 and 12.5 eV is dominated by $4f^{13}$–$4f^{12}$ peaks corresponding to the Yb^{3+}. For $YbMn_2Ge_2$ two peaks, similar to these corresponding to Yb^{2+} in metallic Yb, are observed close to the E_F. Fig. 1(b) illustrates the Yb $4d$ XPS spectra of the investigated $YbMn_2X_2$ (X=Si,Ge) compounds and metallic Yb. For metallic Yb and $YbMn_2Ge_2$ two peaks corresponding to the Yb $4d_{5/2}$ and $4d_{3/2}$ spin–orbit doublet are observed. In the case of $YbMn_2Ge_2$, the Ge L3M23M45 Auger peak also appears in this region, as indicated in the figure. The XPS spectrum of $YbMn_2Si_2$ shown in Fig. 1 has very complex structure due to the $4d^9 4f^{13}$ multiplets. The calculated values of the Mn magnetic moments are equal to 1.93 μ_B for $YbMn_2Si_2$ and 2.34 μ_B for $YbMn_2Ge_2$. These values are in good agreement with the ones determined in the neutron diffraction experiments (2.0(1) μ_B [2] and 2.55(5) μ_B [3], respectively).

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