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ELECTRONIC BAND STRUCTURE OF USi_3[1]

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The electronic band structure of USi_3 is calculated by TB LMTO-ASA method. The 5f electrons form a rather broad band with the center above the Fermi level. The d and p electrons contribute considerably to the density of states at the Fermi level.

1. Introduction

In contrast to 4f electrons in lanthanides, which in most cases are well localized the 5f electrons in actinides exhibit a diversified behaviour. The overlap of the more extended 5f wave functions of neighbouring actinide atoms in solids leads to the delocalization of 5f states. In elemental actinide metals structural and electronic properties can be understood assuming increasing degree of localization with the increasing 5f occupancy. In an early systematics known as the Hill rule it was conjectured that for a spacings between actinide atoms lower than a certain critical value direct 5f-5f overlap results in delocalization of 5f electrons. However, there exist numerous compounds of actinides in which 5f electrons are delocalized even if the separation between actinide atoms exceeds the critical values and there is no direct 5f-5f overlap. In such cases delocalization of 5f electrons is the result of hybridization of the 5f orbitals of actinide atoms with the valence states of the ligand atoms.The interplay between these two main effects, direct 5f-5f overlap and 5f-ligand hybridization decides on a broad variety of behaviours in actinide compounds from normal, broad-band non-magnetic metals, to superconductors, heavy fermion systems and localized moment magnetic systems (see e.g. [1]). The knowledge of the band structure is thus essential to characterize and understand a system in question.

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2. Method

The band structure of USi_3 was computed using the Linear Muffin Tin Orbital method in the Tight Binding Atomic Sphere Approximation (TB LMTO-ASA) [2], with the spin-orbit interactions taken into account. The Perdew-Wang [2] potential with non-local corrections was used. The standard [2] combined corrections terms were included to compensate for errors due to the atomic spheres approximation. The compound USi_3 crystallizes in the Cu_3Au type structure (see e.g. [3] and references therein). The lattice constant is a=4.0348(8)Å [4]. The band structure calculations were performed for the average Wigner-Seitz radius S taken from the experimental value of the lattice constant ($(4\pi/3)S^3 = a^3/4$ for the fcc type unit cell), i.e. for S≅2.9797 a.u. Different values for the Wigner-Seitz radii S for U and Si atoms were assigned by the program, satisfying the general condition $4S^3 = 3S_{Si}^3 + S_U^3$. The values were $S_U = 3.3973 a.u., S_{Si} = 2.8107 a.u.$ giving the overalp volume of muffin-tin spheres 8.9%. The input electronic configurations were taken as: core $+5f^3 6p^6 6d^1 7s^2$ for U and core + $3s^2 3p^2$ for Si. Computations were done for 286 k-points in the 1/48 wedge of the first Brillouin zone for the fcc system. For integration over the Brillouin zone the modified tetrahedron method [5] was employed. The iterations were repeated until the energy eigenvalues of the consecutive iteration steps were the same within an error of 0.01 mRy.

3. Results

The band structure data calculated for the experimental value of the lattice parameter are summarized in the fig.1. The 5f atomic levels of U split into a band of about 0.5 Ry wide with its gravity center lying about 1eV above the Fermi level. The density of states at the Fermi level is dominated by the f electrons on U atoms and the p-electrons on Si atoms. The high peak appears in the partial density of p states on U, indicating that p electrons on U occupy essentially the level lying deeply below the Fermi level. We extended the band structure calculations of USi_3 with spin polarization taken into account. No net magnetic polarization and no magnetic moments either on U or Si atoms appeared within the numerical accuracy of $10^{-4}\ \mu_B$/atom. The system behaves as a metallic, Pauli type paramagnet.

4. Discussion

The band structure of USi_3 was calculated a decade ago by a self-consistent APW method [6] and by the LMTO method [7]. Recent experimental studies [8] required more specific information on the electronic band structure than available in [7] and [8]. The present results obtained by the TB LMTO method with improved accuracy (for 286-k points in the irreducible wedge of the Brillouin zone vs. 84 in [7]) although consistent with the general shape of the spectrum with [6] and [7] revealed significant differences in details. The density of states at the Fermi level, $DOS(E_F)$= 44.56 states/(Ry cell), is lower here than the value 74 states/(Ry cell) reported in [6] and 65.08 states/(Ry f.u.) given in [7]. The high value of the $DOS(E_F)$ can be considered consistent with the value $(1+\lambda)DOS(E_F)$ = 80 states/(Ry cell) determined from the electronic specific heat [9] provided in the phonon enhancement parameter λ, not known for USi_3 from independent measurements, would be small.

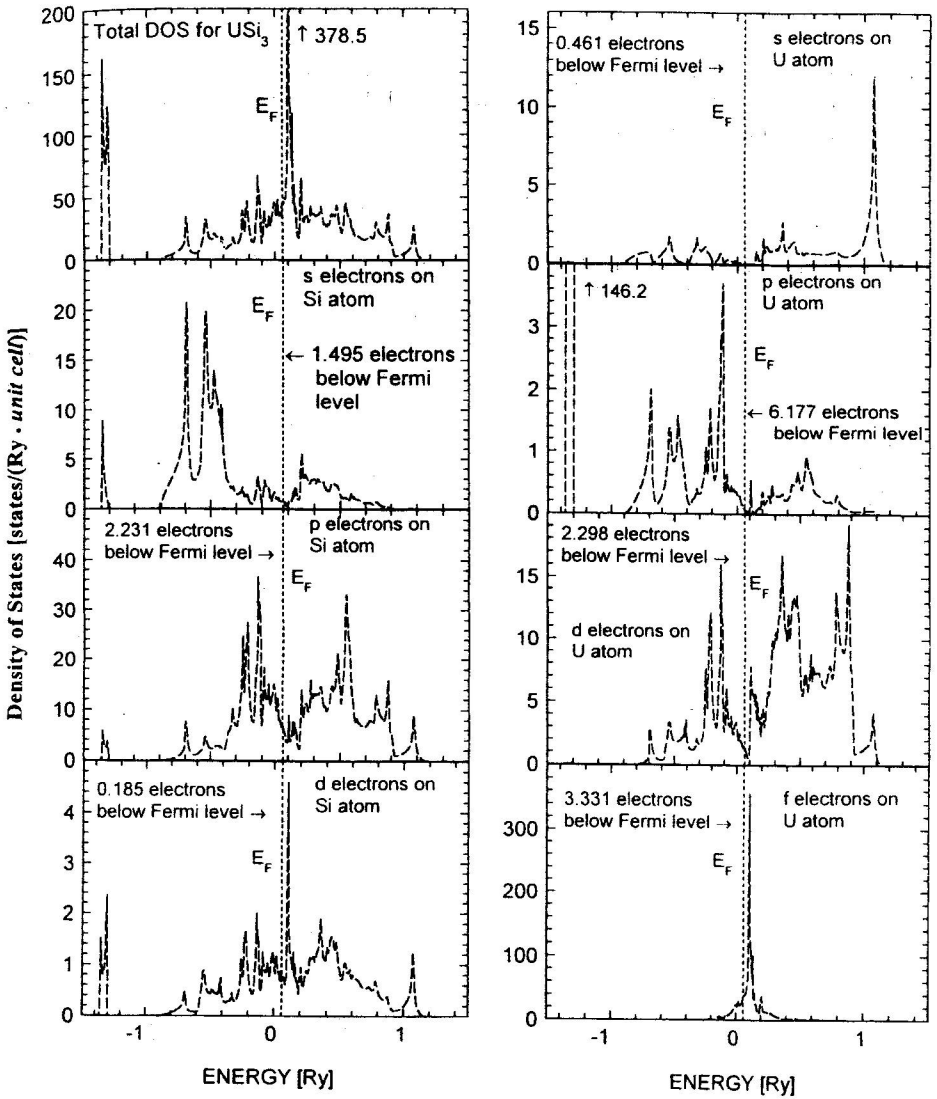

Fig.1. Partial densities of states for USi_3 for s, p, d and f orbitals from U (right panel) and Si atoms. The DOS values for Si are given for three atoms in the cell. Numbers at the arrows pointing up give heights of the corresponding peaks. The information on the total number of electrons (per atom) below the Fermi level is also provided.

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