On the stability of low-symmetry phases in Zr_2CoSi

M. Pugaczowa-Michalska[1]

[1]*Institute of Molecular Physics Polish Academy of Sciences, M. Smoluchowskiego 17, 60-179 Poznan, Poland*

Several proposed Zr_2-based Heusler compounds have attracted interest due to their promising half-metallic properties in inverse cubic Heusler structure. Since ideal full Heusler alloys can crystallize in two different well-ordered cubic structures (Hg_2CuTi-type and Cu_2MnAl-type), this implies diverse behavior of the magnetic moments on the transition metal atoms and also a difference in electronic structure. Therefore, it is important to determine which of the possible structures is energetically more favorable from a finite number of possible candidate structures in the new proposed materials.

Here, with the full potential local orbital (FPLO) minimum basis method and the Vienna ab-initio simulation package (VASP), we present a predictive study of the structural and magnetic properties of Zr_2CoSi. The spin-polarized calculations of the electronic structure of Zr_2CoSi involved both above-mentioned cubic Heusler structures. In our calculations, Zr_2CoSi in the Hg_2CuTi structure has a higher total energy of about 1.4 eV/f.u. than that in the Cu_2MnAl one (or $L2_1$). Additionally, it seems that the negative value of formation energy may indicate the possibility of crystallization of the Zr_2CoSi compound in the $L2_1$ cubic structure. Unfortunately, the density of states (DOS) do not support the half-metallic state of Zr_2CoSi. Van Hove singularity involving partially occupied Co 3d e_g states occurs in minority states of the Zr_2CoSi at the Fermi level. The studied compound satisfies the preconditions to exhibit a cubic-to-tetragonal instability according to a band-Jahn-Teller mechanism. On the other hand, the calculated elastic constants of Zr_2CoSi in the $L2_1$ structure for both exchange-correlation functionals used in the calculations: generalized gradient approximations (GGA) and local density approximations (LDA), shows that the system exhibits mechanical instability due to negative value of the tetragonal shear modulus C'. Our analysis of the total energy landscapes of the system shows that there are two tetragonal and one orthorhombic distorted phases of Zr_2CoSi that are energetically favored over the undistorted cubic $L2_1$ structure. However, only one tetragonal phase of Zr_2 CoSi with $c/a = 1.22$ fulfills necessary and sufficient elastic stability conditions in both GGA and LDA calculation. The calculated values of elastic constants of Zr_2CoSi in the second tetragonal phase (with $c/a < 1.0$) and orthorhombic one (with parameter $\delta = 0.89$) have not satisfied the Born inequalities. The calculations show that the lowering of the symmetry and the increase of the tetragonal distortion parameter is responsible for the decrease in the magnetic moment. In the tetragonal phase ($c/a = 1.22$) the total magnetic moment of Zr_2CoSi falls to 1.268 μ_B.