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Theoretical investigation of the electronic structures and magnetic properties of the bulk and surface (001) of the quaternary Heusler alloy NiCoMnGa

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ABSTRACT

In this paper, we study the electronic structures, magnetic properties, and half-metallicity of the bulk and (001) surface of Heusler alloy NiCoMnGa. Our first-principles calculations exhibit that, within the generalized gradient approximation (GGA) of the electronic exchange–correlation functional, the quaternary Heusler alloy NiCoMnGa is a half-metallic ferromagnet at the equilibrium lattice constant of 5.795 Å with a total spin magnetic moment of 5 μ_B per formula unit. The calculated total atomic magnetic moment follows the Slater–Pauling rule. At the same equilibrium lattice constant, the half-metallicity confirmed in the bulk NiCoMnGa, is destroyed at both MnGa- and NiCo-terminated (001) surfaces and subsurfaces. Based on the magnetic property calculations, the magnetic moments of Co, Mn, and Ga atoms at the NiCo- and MnGa-terminated surfaces increase with respect to the corresponding bulk values while the atomic magnetic moment of Ni at the NiCo-terminated surface decreases.

1. Introduction

Searching for materials with high spin polarization becomes an attractive field for many researchers as an example of spintronic applications [1,2]. It is well known that a high Curie temperature and consistent electronic properties associated with Heusler alloys make these alloys of particular interest [3–5]. The 100% spin-polarization at the Fermi level (E_F) in half-metallic (HM) materials is used as a useful candidate for spin-injection devices. It displays two behaviors at the same time, one as metallic and the other a semiconducting or insulating with an energy gap at the Fermi level following the spin channel. The first time when the HM characteristic in the intermetallic half-Heusler compound NiMnSb, was predicted by de Groot et al. [6]. Many theoretical studies have been conducted to detect the transition-metal-based HM ferromagnets, such as the full-Heusler alloy Co_2CrAl [7], perovskite $La_{0.7}Sr_{0.3}MnO_3$ [8], pyrite-type CoS_2 [9], spinel Fe_3O_4 [10], rutile-type CrO_2 [11], and certain transition metal pnictides and chalcogenides in zinc-blende structure [12–14].

The $L2_1$ Heusler type (space group $Fm3m$) includes four interpenetrating face center cubic (fcc) lattices. If different atoms occupy each of the sublattices we obtain a quaternary Heusler structure with an alternative symmetry, which is the well known LiMgPdSn or Y-type structure (space group $F43m$) [15,16]. Heusler alloys can exhibit tunable electronic and magnetic properties based on their valence-electron count, so this class of materials offer a huge variety of eventualities for the design of rational materials. An important example is the quaternary alloy CoFeMnSi, which has an expected HM band structure [17]. More theoretical works on quaternary Heusler compounds CoFeCrZ done by Gao et al. [18] exposed that among all alloys investigated, both CoFeCrAl and CoFeCrSi are excellent HM ferromagnets with large HM gaps of 0.16 and 0.28 eV, respectively. They also showed that the half-metallicity of CoFeCrAl and CoFeCrSi appears to be severe against the lattice compression (till up to 7% and 4%, respectively), and the half-metalicity is preserved for CoFeCrSi and CoFeCrGe but destroyed for CoFeCrAl and CoFeCrGa when the Coulomb interaction is considered.

Recently, Felser's research group [17,19–21] found the HM ferromagnetism in many quaternary Heusler alloys such as CoFeMnZ (Z=Al, Si, Ga, Ge), NiFeMnGa, and NiCoMnGa based on some ternary Heusler compounds. They also observed high Curie temperatures (from 326 to 711 K) after fruitfully synthesizing these alloys [20,21]. Alijani et al. [21] fabricated NiFeMnGa, NiCoMnGa, and CuCoMnGa quaternary Heusler compounds by arc-melting the stoichiometric mixtures of high-purity elements in an argon

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atmosphere. They have proved experimentally that NiCoMnGa keeps its half-metallicity and high Curie temperature, high spin polarization and high magnetic moment were observed, which makes it important as a new material for spintronic applications.

There are no theoretical or experimental studies on the thin films or multilayers of NiCoMnGa. These properties are very important to the practical spintronic implementations, because the half-metallicity of bulk may be lost at the surfaces and interfaces. For example, recent studies demonstrated that the half-metallicity verified in the bulk Mn_2CoSn is destroyed at its surfaces [22]. Very recently, the half-metallicity of Mn_2CoAl (001) surface [23] was investigated via first-principles calculations. It is found that because of the strong surface potential effect on the surface of Co atom, the CoMn-terminated (001) surface fails to preserve the half-metallicity, which can be observed in the bulk. Motivated by the above, we apply here the density functional theory to investigate the structural, electronic, and magnetic properties of the bulk and all possible (001) surfaces of NiCoMnGa.

2. Computational method

In this work, we use the full-potential linearized augmented-plane wave method to investigate the electronic, magnetic and structural properties of the bulk and NiCoMnGa (001) surface by applying the Wien2K designed by Blaha, Schwarz, and co software provided-workers [24–26]. The electronic exchange correlation functional is obtained within the generalized gradient approximation in the scheme of Perdew, Burke, and Enzerhof [27]. We select the k meshes of $15\times15\times15$ for the bulk and use the $15\times15\times1$ k meshes for the (001) surface in the Brillouin zone. The number of plane waves is restricted by $R_{mt}\times K_{max}=8.5$ and the expansion is made up to $l=10$ in the muffin tins. In the present calculations, both in the bulk and surface systems, the muffin-tin radii are set to 2.3 a.u. for the transition metals in addition to the main group elements. The self-consistency calculations are considered to be achieved when the total energy difference between succeeding iterations is less than 10^{-5} Ry per formula unit. We know that the crystal structure of the quaternary Heusler alloy NiCoMnGa has a highly ordered $CuHg_2Ti$-type structure. The type of atom arrangement in the quaternary Heusler compound is X_1X_2YZ: X_1 (3/4, 3/4, 3/4), X_2 (1/4, 1/4, 1/4), Y (1/2, 1/2, 1/2), and Z (0, 0, 0). Many studies on the Heusler compounds used the $CuHg_2Ti$-type structure [28,29].

3. Results and discussion

3.1. Electronic structure of bulk $CuHg_2Ti$–NiCoMnGa

We start with the geometry optimization of the lattice constants of bulk NiCoMnGa in $CuHg_2Ti$ phase, by using the ferromagnetic calculations of respective total energy-versus-lattice constants curves, as indicated in Fig. 1. The equilibrium lattice constant is thus obtained from the lowest energy. Our calculated equilibrium lattice constant of $CuHg_2Ti$–NiCoMnGa is 5.795 Å. One can see that, the result is in agreement with the previously available experimental result (5.803 Å) and theoretical value (5.784 Å) [21].

More significantly, the total spin magnetic moment of NiCoMnGa is 5 μ_B per formula unit that is integer-valued. This integer value is a very important condition for a compound to be a HM ferromagnet. The total magnetic moment of NiCoMnGa consists of five parts: the magnetic moment of the Ni atom (0.593 μ_B), the magnetic moment of the Co atom (1.225 μ_B), the magnetic moment of the Mn atom (3.253 μ_B), the magnetic moment of the Ga atom (-0.050 μ_B), and the magnetic moment of interstitial area (-0.021 μ_B). It means that the main contribution to the total magnetic moment of $CuHg_2Ti$–NiCoMnGa comes from the 3d electrons in the transition metals. The calculated total atomic

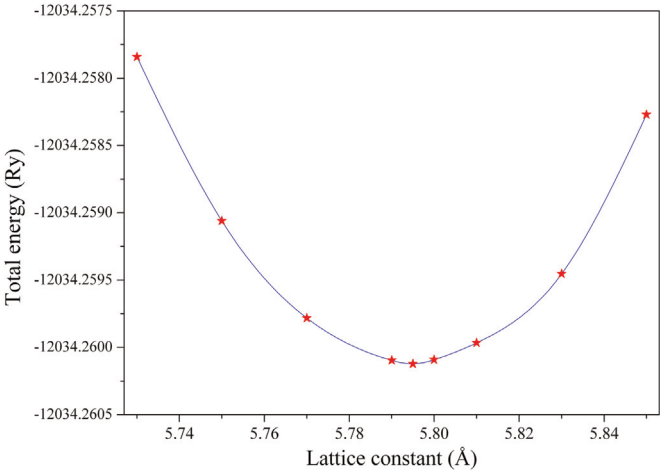

Fig. 1. The total energy versus lattice constant for NiCoMnGa in the $CuHg_2Ti$ structure.

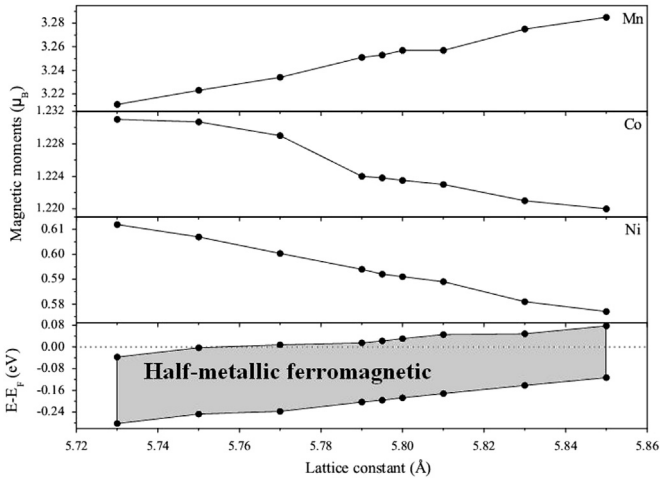

Fig. 2. The calculated Mn, Co and Mn magnetic moments, and half-metallic ferromagnetism as functions of lattice constant.

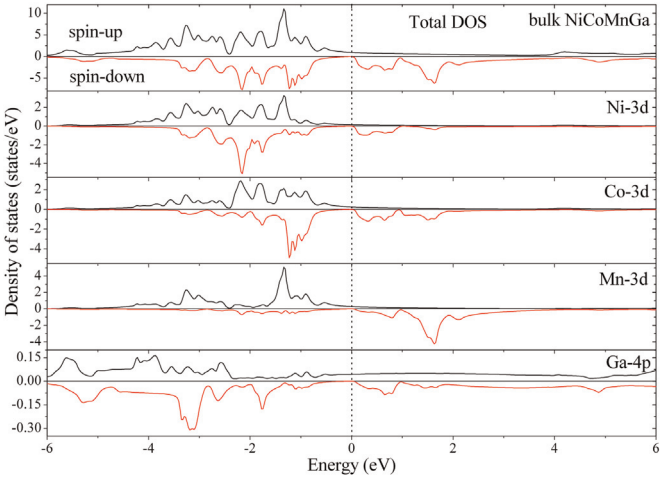

Fig. 3. The calculated total and partial densities of states of the NiCoMnGa quaternary Heusler compound. The zero energy value corresponds to the Fermi level E_F.