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Influence of varying Germanium content on the optical function dispersion of $Fe_2MnSi_xGe_{1-x}$: An *ab initio* study

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ABSTRACT

The optical dielectric functions of $Fe_2MnSi_{1-x}Ge_x$ alloys for selected concentrations (x=0.0, 0.25, 0.5, 0.75 and 1.0) were investigated. The ferromagnetic $Fe_2MnSi_xGe_{1-x}$ is semiconducting with optical band gaps 0.507, 0.531, 0.539, 0.514 and 0.547 eV for the minority spin and is metallic for the majority spin. From the calculated results the half-metallic character and stability of ferromagnetic state for $Fe_2MnSi_xGe_{1-x}$ is determined. The total magnetic moment is found to be $3.0\mu_B$ for all alloys with the most contribution from Mn local magnetic moments. Iron atoms however exhibit much smaller spin moments, about 10% of the bulk value, and the *sp* atoms have induced magnetic moments due to the proximity of Fe first nearest neighbors, which couple antiferromagnetically with Fe and Mn spin moments. We have employed full-potential linearized augmented plane wave method based on spin-polarized density functional theory. The generalized gradient approximation exchange-correlation potential was used. The edge of optical absorption for $\varepsilon_2(\omega)$ of spin-down varies between 0.507 (Fe_2MnGe) and 0.547 eV (Fe_2MnSi). Since the spin-up shows metallic nature, the Drude term was included in the spin-up optical dielectric functions. This confirms our finding that these materials are half-metallic. Furthermore, the reflectivity, refractivity and the absorption coefficient were calculated. These results show that the materials possess half-metallic character.

1. Introduction

For about three decades the study of half metallic compounds has received a great deal of attention [1–13]. The interest in these compounds is driven by the spin degree of freedom in electronics [14–17]. This rapidly expanding field of spin based electronics (spintronics) has exploited a large class of emerging materials, such as high temperature superconductors [18], ferromagnetic semiconductors [19,20], organic ferromagnets [21,22], organic semiconductors [23], and carbon nanotubes [24,25] based devices, to bring about revolution and novel functionalities to the traditional devices. In other areas within spintronics a degree of spin polarization as high as possible is required, preferably 100%. Half metals are considered in this area. These materials are metals for one spin direction, while being semiconductors or insulators for the opposite spin direction.

Half-metallic ferromagnetic (HMF) compounds were discovered by de Groot et al. in 1983 [26,27], till date a lot of them have been theoretically predicted and some of them furthermore have been confirmed experimentally [28–31]. Much interest has been paid to understand the mechanism behind the half-metallic magnetism and to study its implication on various physical properties [32,33]. However, it is highly desirable to explore new half-metallic ferromagnetic materials which are compatible with important III–V and II–VI semiconductors. Among the most cited candidates for 100% spin polarization are semi-Heusler alloys [26,27,33–36], Full Heusler alloys [9,37], zinc-blende structure materials [38,39], and semimetallic magnetic oxides CrO_2 and Fe_3O_4 [11,40,41]. A number of other materials have also been suggested as half metallic ferromagnets. Among them, $Fe_2MnSi_{1-x}Ge_x$ compounds. For offering insights into their physical properties, we have studied the optical properties of $Fe_2MnSi_{1-x}Ge_x$ compounds using the linearized augmented plane waves in the frame of density functional theory. Optical techniques have established themselves as providing key insights into the band structure of materials. In fact, the electronic structure of many semiconductors has been determined by comparing theoretical calculations to optical results [42].

Although Fe_2MnSi is studied extensively and is predicted to be an HMF [43–45], it is anticipated that this material can be

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successfully applied to highly efficient spin injection and detection through Schottky tunnel barriers in group-IV semiconductor devices. Also epitaxial Fe_2MnSi thin films have a Curie temperature of about 210 K, which is much lower than room temperature [46], however, there is a limited experimental work and no theoretical work relating directly to the electronic structure, optical and magnetic properties of $Fe_2MnSi_{1-x}Ge_x$ compounds. In hope of achieving a high magnetization by exploiting Mn and a giant magnetocaloric effect (GMCE) by combining a magnetic transition with a structural transition, Zhang et al., have studied the $Fe_2MnSi_{1-x}Ge_x$ compound series and they found that the Curie temperature, as well as the spontaneous magnetization, does not significantly change with Ge content in the single-phase $L2_1$. There is no field-induced transition and the magnetocaloric effect is rather small throughout the $Fe_2MnSi_{1-x}Ge_x$ series [47]. In addition, in the $L2_1$-type of structure, both Fe_2MnGe and Fe_2MnSi have a Curie temperature around 250 K [47]. Further insight into the electronic structure of the HMF compounds can be obtained from the calculation of optical functions, by summing the transitions from occupied to unoccupied states over whole Brillouin zone (BZ) points. In this work we have investigated the optical properties of $Fe_2MnSi_{1-x}Ge_x$ alloys for selected concentrations (x=0.0, 0.25, 0.5, 0.75 and 1.0). The frequency dependent optical properties, namely reflectivity, absorption coefficient, electron energy loss spectra, refractive index, are given. We should emphasize that such kind of alloys may be of particular interest due to large electron–phonon anharmonities [48].

The rest of the paper is organized as follows. The computational details regarding the calculation of structural properties as well as the optical ones are given in Section 2. In Section 3, the results from our calculations are presented and discussed. The most important conclusions drawn from our calculations are given in Section 4.

2. Structural aspects and computational details

The Heusler compounds Fe_2MnSi and Fe_2MnGe crystallize in the $L2_1$ structure which consists of four face centered cubic (FCC) sublattices. The space group is (Fm3m) (No. 225). The $L2_1$ phase is a cubic superstructure of four interpenetrating fcc sublattices, **A**, **B**, **C**, and **D**, centered at (0 0 0), ($\frac{1}{4}\frac{1}{4}\frac{1}{4}$), ($\frac{1}{2}\frac{1}{2}\frac{1}{2}$) and ($\frac{3}{4}\frac{3}{4}\frac{3}{4}$). Each **A** atom is at the center of a cube of four **B** atoms and four **D** atoms. To trace the variation in the composition, we employ supercell approach.

We make use of the Vienna package WIEN2K [49] for all calculations. This is a full potential linear augmented plane wave plus local orbitals method within the density functional theory [50]. The electronic exchange-correlation energy is treated using the generalized gradient approximation parameterized by Perdew–Burke–Ernzerhof (GGA–PBE) [51]. Relativistic effects are taken into account within the scalar approximation. For ***k***-space integration, 1000 points is used in the whole Brillouin zone (BZ) for structural properties. The BZ integrations are performed using the modified tetrahedron interpolation method [52]. We used denser meshes of 200 ***k***-points in the irreducible Brillouin zone (IBZ) for optical properties of binary as well as ternary alloys. We set $K_{max}=9.0/R_{MT}$ and make the expansion up to $l=10$ in the muffin tins. The convergence of the total energy in the self-consistent calculations is taken with respect to the total charge of the system with a tolerance 0.0001 electron charges. The structures are fully relaxed until the forces on the atoms reach values less than 2 mRy/a.u. The total energy dependence on the cell volume is fitted to the Murnaghan equation of state (EOS) [53]. We found that the lattice parameter increases linearly from 5.59 to 5.70 Å with increasing Ge content while the bulk modulus decreases from 234.96 to 216.69 GPa when we move from x=0.0 to 1.0.

3. Results and discussion

3.1. Magnetic moments and electronic structure

The total calculated magnetic moment per formula units is found to be $3\mu_B$/f.u., and it is not affected by the change of Ge content. That is attributed to the fact that both Si and Ge are nonmagnetic and exhibit negligible induced magnetic moments. The most contribution of the magnetic moment comes from Mn atoms, which is related to the half d band filling. In contrast Fe atoms exhibit much smaller local magnetic moments as compared to their bulk values. This behavior is related to the location of these atoms at A and C sites, where the first nearest neighbors of Fe atoms are Si (or/and) Ge atoms. This leads to the hybridization between p bands of Si (Ge) with d bands of Fe atoms that suppresses their magnetic moments. This behavior has been observed earlier from other structures such as Fe_2CrSi [54] and $Fe_{3-x}Mn_xZ$ (Z=Al, Ge, Sb) Heusler alloys [55]. It is also noticed that the local spin magnetic moments of Fe atoms slightly decrease as a function of Ge content. In contrast, the local spin magnetic moments increase for Mn atoms (Mn_1) that have higher number of next nearest neighbors of Ge in the non-stoichiometric alloys. These Mn_1 atoms occupy the perfect octahedral sites with eight nearest neighbors of Fe atoms and six next nearest neighbors of Ge atoms. However, those of other kind of Mn atoms (Mn_2) have lower symmetry due to the presence of two kinds of the next nearest neighbors Si and Ge. These Mn_2 atoms exhibit lower local spin magnetic moments. The *sp* atoms, however, exhibit induced magnetic moments due to the proximity of Fe first nearest neighbors and they couple antiferromagnetically with Fe and Mn local magnetic moments.

From the calculated band structures of $Fe_2MnSi_{1-x}Ge_x$ alloys (Fig. 1a–f), one can see that the majority spin (spin-up) exhibits a metallic nature, as prototype see Fig. 1a, whereas the minority spin shows a semiconducting behavior with energy band gaps of about 0.507, 0.531, 0.539, 0.514 and 0.547 eV for x=0.0, 0.25, 0.5, 0.75 and 1.0, respectively. This is an indication that these compounds are half-metallic alloys. Carefully looking at the details of Fig. 1, we should emphasize that the stoichiometric Heusler alloys exhibit indirect band gaps along the Γ–X symmetry lines, whereas the non-stoichiometric ones possess direct band gaps located at the Γ point. The same behavior was observed with our previous calculations for other alloys such as $Fe_{3-x}Mn_xSi$ [56] and $Fe_{3-x}Cr_xSi$ [57]. The values of the spin-down band gaps are slightly increased with increasing content of Ge. The upper valence bands shift towards Fermi energy (E_F) as a function of Ge content slightly cross E_F in Fe_2MnGe Heusler alloy causing a negative spin flip gap. The spin-flip's values of $Fe_2MnSi_{1-x}Ge_x$ alloys are 0.259, 0.058, 0.005, −0.031, and −0.085 eV for 0.0, 0.25, 0.5, 0.75 and 1.0 of Ge content respectively. While the conduction band minimum shifts E_F rearwards to maintain the value of the energy gap. We should emphasize that this observation is consistent with a previous work [56], in which Hamad et al. has shown that changing the content of Si atom at the expense of Fe in $Fe_{3-y}MnSi_y$ does not affect the band gap [56]. This also goes along with the origin of the band gap that is related to the hybridization between X and Y d bands in X_2YZ full Heusler alloy [58].

3.2. Optical properties

The optical dielectric function helps to obtain further insight into the electronic structure. We investigate the dielectric functions of $Fe_2MnSi_{1-x}Ge_x$ alloys for selected concentrations