

Study of Half-Metallicity of Half-Heusler GeLiCa by Using mBJ Potential Method

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Abstract: The spin-polarized calculations of FP-LAPW (full-potential linearized augmented plane-wave) method based on DFT, the mBJ (modified Becke-Johnson) exchange potential by Tran and Blaha was employed with Perdew Burke-Eenzerhof GGA (generalized gradient approximation) and LSDA (local spin density approximation) to analyse the half-metallicity of GeLiCa. We found that in both GGA-mBJ and LSDA-mBJ calculations the GeLiCa alloy is HMF (half-metallic ferromagnet) with a magnetic moment of $1 \mu_B$ per formula unit at equilibrium lattice constant. The mBJ calculations for both GGA and LSDA yielded an increase in the energy gap E_{BG} around the Fermi level and a more accurate magnetic moment of $1 \mu_B$ with respect to the GGA and LSDA results.

Key words: Half-Heusler alloys, half-metals, density functional theory, modified Becke-Johnson exchange potential.

1. Introduction

The rapid development in the field of spintronics and thermoelectric applications has provided strong interest in the search for novel materials such as HMFs (half-metallic ferromagnets). HMFs magnetic materials with high spin polarization were widely used to improve the performance of spintronic devices such as spin filters and spin valves [1]. In 1983, de Groot et al. [2] predicted the first HMF of the half-Heusler alloy NiMnSb.

In the $L2_1$ structure of full-Heusler X_2YZ compounds, X and Y are usually transition metals and Z often is an element from columns III-VI in the Periodic Table, and is regarded as four interpenetrating FCC (face-centered cubic) sub lattices. The X atoms are located at A (0, 0, 0) and C (1/2, 1/2, 1/2) while the Y atom is located at B (1/4, 1/4, 1/4) and Z atom occupies D (3/4, 3/4, 3/4). The $L2_1$ structure becomes the $C1_b$ -type structure of half-Heusler compounds XYZ when the X position (1/2, 1/2, 1/2) is vacant. Compounds which are

maximized for the efficiency of the devices based on spintronics are the so called HM materials, i.e., there a complete spin polarization of the conducting electrons at E_F .

Calculations of the structural, electronic and magnetic properties of the half-Heusler GeLiCa are conducted by using the first-principle FPLAPW (full-potential linearized augmented plane-wave) method [3]. We have adopted the Kohn-Sham [4] (KS method of DFT (density functional theory)) in our calculations [5]. We use the GGA (generalized-gradient approximation) suggested by PBE (Perdew-Burke Ernzerhof) [6], LSDA (local spin density approximation) [7], and mBJ (modified Becke-Johnson) local density approximation functional calculations [8] for the exchange correlation interactions. Even though LSDA and GGA provide the best theory for interpreting experimental data, it has a weakness in the prediction of excited state properties. So to get a better representation of the band gaps we have used the mBJ potential. The mBJ potential is given by

$$V_{mBJ}(r) = cV_x^{BR}(r) + (3c-2) \frac{1}{\pi} \sqrt{\frac{5}{12}} [2t(r)/\rho(r)]^{1/2}$$

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where $v_x^{BR}(r)$ is the Becke-Roussel exchange potential, r is the electron density and $t(r)$ is the Kohn-Sham kinetic energy density.

2. Computational Method

The structural optimization and electronic calculations were made by using a self-consistent FP-LAPW (full-potential linearized augmented plane wave) method, which depends on DFT instigated into the WIEN2K simulation package [9]. We have used a mesh of $12 \times 12 \times 12$, consisting of 72 special k-points that were taken in the Brillouin zone (10,000 k points in the full BZ) of irreducible wedge for the integrations within the modified tetrahedron method. The cut off parameter for the plane wave was set to $K_{\max} \times R_{\text{mt}} = 7.0$, where K_{\max} gives the uttermost value of the reciprocal lattice vector used in plane wave function and R_{mt} represents the smallest muffin tin sphere radii. The G_{\max} value is set to 12, where G_{\max} is the largest vector value in charge density Fourier expansion.

GeLiCa has an atomic arrangement in the $C1_b$ structure of half-Heusler compounds in the space group Fm-43. We used the atomic arrangement where Ge, Li and Ca occupy the (0, 0, 0), (1/4, 1/4, 1/4) and (3/4, 3/4, 3/4) positions for calculations as it gives the most stable structure with the lowest minimized energy. The optimized lattice parameter for GeLiCa at equilibrium lattice constant is 6.94 \AA which is slightly more than 6.84 \AA [10].

3. Results and Discussion

3.1 DOS (Density of State) and Band Structures

We discuss the electronic and magnetic properties of GeLiCa HH compounds in this section. The spin polarized total density of states and partial density of configuration in the Ferromagnetic state in the equilibrium lattice keep constant. The energy gaps in the majority spin channel are about 1.23 eV, 1.73 eV, 1.14 eV and 1.72 eV respectively. These values are larger than those of NiMnSb (0.5 eV) and CoMnSb (1

eV) [11]. The half-metallic gap E_{HM} is 0.12 eV for GGA, 0.42 eV for GGA+mBJ, 0.06 eV for LSDA, and 0.42 eV for LSDA+mBJ.

Comparing the total and partial DOS (density of state) with the band structures (Fig. 1) it can be seen that the atomic spin moment mainly originates from the Ge atom. We can also see that in both spin up and down channels, the three lowest bands mainly originate from the hybridized Ge- p states and Ca- d states. The half-metallicity is mainly from the spin polarization of the Ge- p states and the hybridization between the Ge- p states and the Ca- d states.

The different values of energy band gap E_{BG} , half-metallic gap E_{HM} and magnetic moments for GGA-mBJ and LSDA-mBJ are given in Table 1 along with a previous calculation for comparisons.

Fig. 2 shows the respective band structure plots of GeLiCa in GGA and LSDA along with the mBJ plots in both spin channels along with total DOS. The valence region of the spin up and down channels shows a number of bands due to the Ge- p states on GeLiCa. In both cases for GeLiCa with the GGA and LSDA with mBJ calculation the width of the energy gap E_{G} , which is the difference in energies of the maximum occupied band at the symmetry point Γ in the valence band and the minimum unoccupied band in the conduction region at a symmetry point X, is an indirect gap. GeLiCa-GGA has an indirect band gap of 1.23 eV obtained between Γ and X while in GeLiCa-mBJ the indirect gap between Γ and X is 1.73 eV. Similarly, for GeLiCa-LSDA an indirect band gap obtained between L and X shows 0.19 eV and for GeLiCa-mBJ, an indirect gap between Γ and X gives 0.39 eV. With the employment of mBJ potential calculations for both GGA and LSDA, it is observed that the valence band shifts downwards and the conduction band moves up resulting in a wider energy gap around E_{F} . However it is observed that GGA+mBJ calculations reveal a slightly larger energy gap than LSDA+mBJ calculations.

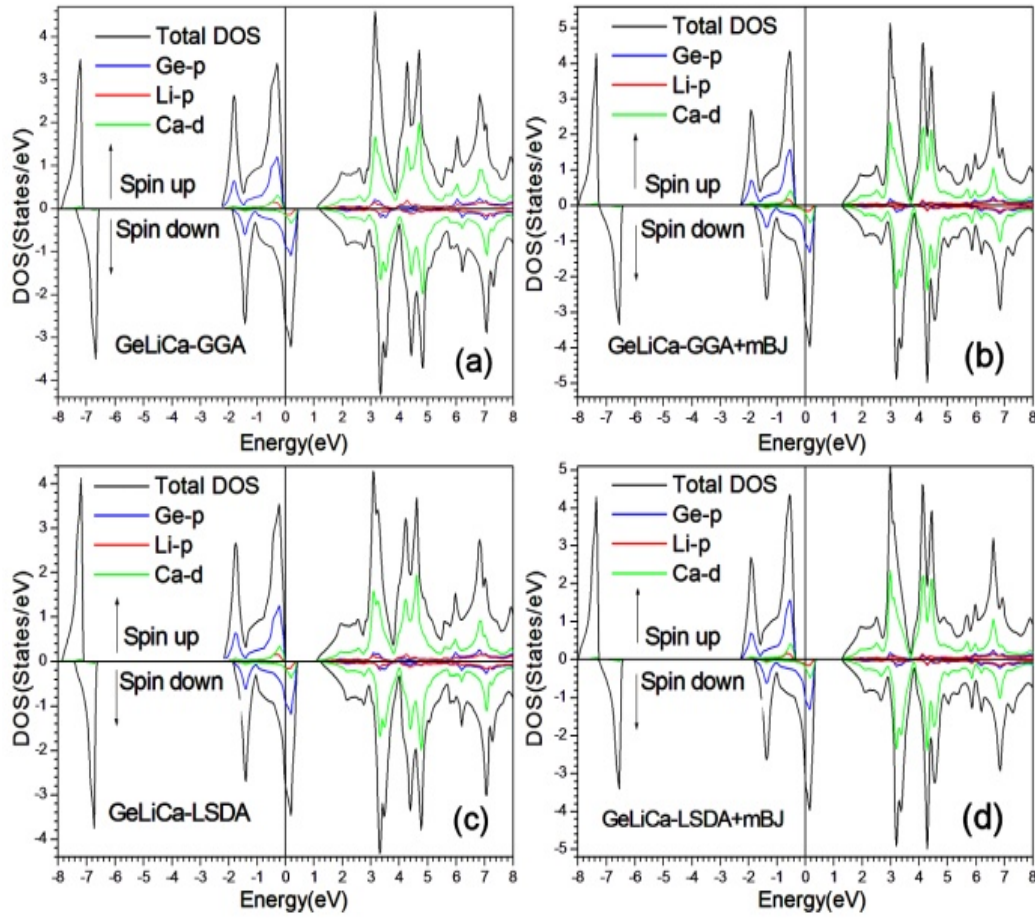


Fig. 1 The spin-projected total and partial density of states of GeLiCa in (a) GGA, (b) GGA+mBJ, (c) LSDA and (d) LSDA+mBJ at equilibrium lattice constant.

Table 1 Different physical properties of GeLiCa at equilibrium lattice constant. E_{BG} is the energy band gap, E_{HM} is the half-metallic gap, MM is the total magnetic moment and * gives previous calculation [10].

Material	V_{XC} (eV)	E_{BG} (eV)	E_{HM} (eV)	E_{HM}^* (eV)	MM (μ_B)	MM* (μ_B)
GeLiCa	GGA	1.23	0.12	0.03	0.99	1.00
	mBJ	1.73	0.42	-	1.00	-
	LSDA	1.14	0.06	-	0.99	-
	mBJ	1.72	0.42	-	1.00	-

3.2 Magnetic Properties

The magnetic moments of Ge, Li and Ca are $0.431 \mu_B$, $0.025 \mu_B$ and $0.089 \mu_B$ respectively for GGA-mBJ calculations with an effective magnetic moment of $1.00 \mu_B$ with the maximum contribution from the Ge atom. The LSDA-mBJ calculations also give an effective magnetic moment of $1.00 \mu_B$ while the GGA and LSDA magnetic moment works out to be $0.991 \mu_B$. So the mBJ calculations give a better result for the

half-metallicity with regards to magnetic moments.

Since the lattice distortion is very crucial in the synthesis of spintronic materials we focused on the robustness of the ferromagnetism with respect to lattice distortion. Fig. 3 shows the calculated total magnetic moment per formula unit as function of lattice constant for GeLiCa. The magnetic moment is still an integer of Bohr magneton, $1.00 \mu_B$, when the lattice constant is contracted to 5.5 \AA and maintaining half-metallicity up to 20% contraction of the lattice

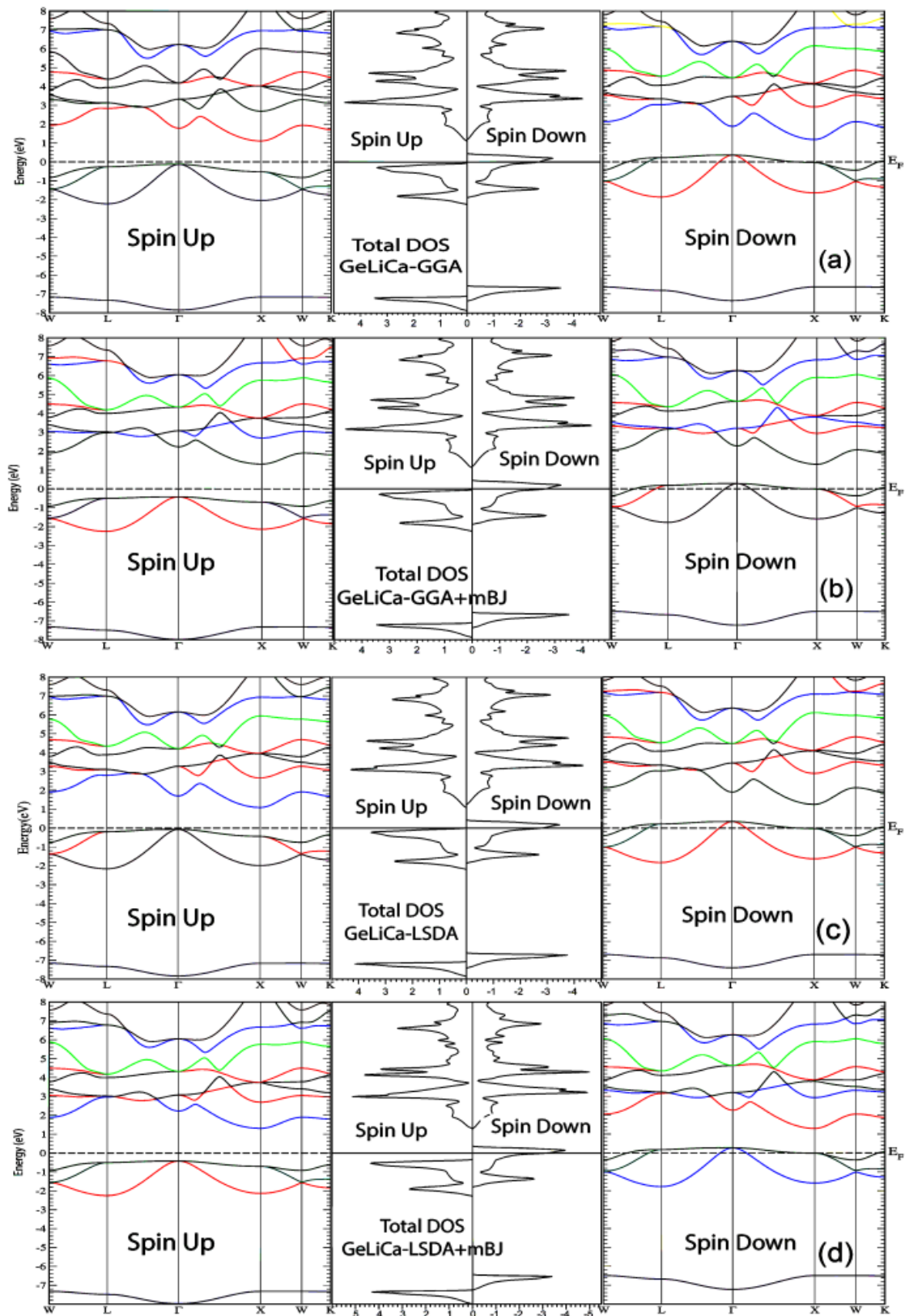


Fig. 2 The spin-projected total density of states of GeLiCa with the corresponding band structures.

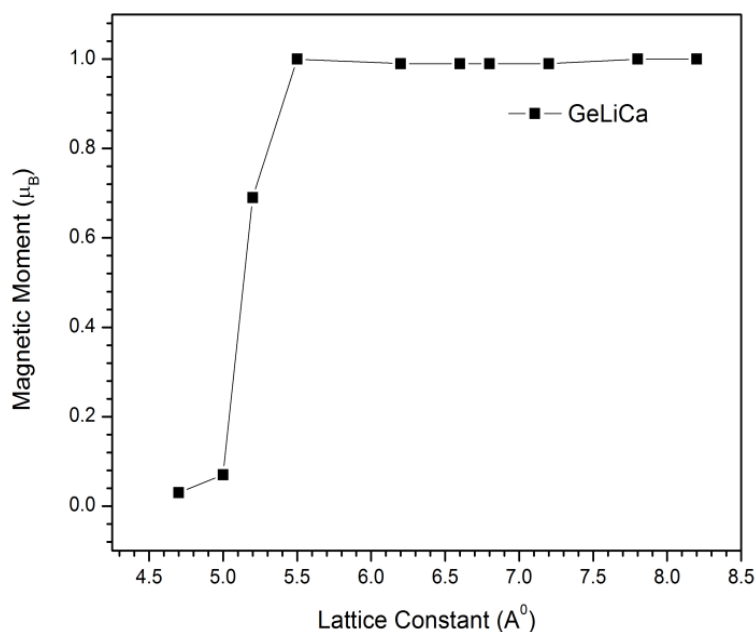


Fig. 3 Magnetic moment as a function of lattice constant.

constant, with respect to equilibrium state which is much more than the lattice contraction of 2% of CoMnSb [11].

4. Conclusions

In summary, we investigated structural, electronic, and HM properties of the hypothetical HH compounds GeLiCa using first principle DFT calculations with GGA, LSDA and mBJ parameterization to calculate the exchange correlation energy and potential. Half-metallicity has been found in the compound GeLiCa at its optimized lattice constant. The half-metallic gap E_{HM} increases from 0.12 eV (GGA) to 0.42 eV (GGA+mBJ) and 0.06 eV (GGA) to 0.42 eV (LSDA+mBJ). The use of mBJ potential shows that the energy gap E_{BG} increases from 1.23 eV (GGA) to 1.73 eV (GGA+mBJ) and 1.14 eV (GGA) to 1.72 eV (LSDA+mBJ). The increase in both the energy gap E_{BG} and half-metallic gap E_{HM} is much more prominent in the mBJ calculations than both the GGA and LSDA values. The hybridization between p - d orbitals of Ge and Ca atoms causes the semi-conducting gap in the majority-spin state for GeLiCa compound, which is similar like in GeKMg

[12].

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