First principles investigation of structural, elastic, electronic and optical properties of Barium seleno-germanate, Ba_2GeSe_4

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Abstract. Ternary and quaternary chalco-germanates and stannates have a rich structural chemistry. Experimental studies of their non-linear optical properties have been reported, but there are few published computational studies on their structural, elastic, electronic and optical properties. In this work, we investigate the structural, elastic, electronic and optical properties of Ba_2GeSe_4 using Density Functional Theory (DFT) and post-DFT many body perturbation theory. The ground state energy and properties, including equilibrium lattice parameters, bulk modulus and band gap were calculated at the DFT level of approximation. The fundamental gap was determined at the post-DFT G_0W_0 level of approximation while optical absorption was determined with the Bether-Salpeter Equation approximation. The ground state energy and mechanical results show that Ba_2GeSe_4 is a stable compound while the caclulated optical absorption results and estimated optical band gap show that it is a wide band gap material that is well-situated for photon absorption in the high energy visible range with potential application in multi-junction solar cells.

1. Introduction

The ternary and quaternary compounds containing metal chalcogenides, chalco-germanates and thiostannates have received considerable attention due to their structural chemistry [1, 2], important physical and chemical properties for potential applications in non-linear optics (NLO) [3, 4], thin film for solar cell [5] and visible light response photocatalysts [6, 7]. Among the family of chalco-germanates is seleno-germanate earth alkali metal barium seleno-germanate Ba_2GeSe_4. The experimental synthesis of Ba_2GeSe_4 has been reported and the compound is found to crystallize in a monoclinic structure with space group p_12/m [8]. The thio/seleno-germanate and stannate compounds exhibit interesting physical properties due their isolated tetrahedral ortho-anion $[MQ_4]^{4-}$ corner shearing, were [M = Ge, Sn; Q = S, Se]. The compounds are well known to have a structure of type A_2MQ_4 [4]. Little has been reported on the structural, elastic, electronic and optical properties of Ba_2GeSe_4. We are only aware of the study of Assoud and Soheilnia [8], were the authors reported experimental data on the structure and lattice constants and theoretically investigated the electronic properties using density functional theory with the local density approximation (LDA). However, the authors did not measure the optical band

3.5. Conclusion

Using first principle calculations, we investigate the structural, elastic, electronic and optical properties of the barium seleno-germanate Ba_2GeSe_4 compound. The calculated lattice parameters are in reasonable agreement with available experimental data. The electronic band structure calculations from MBJ which provide accurate band structure for most large band gap semiconductors shows an 2.26 eV direct band gap at $\Gamma - \Gamma$ points other energy gap calculation from HSE and G_0W_0 are within the predicted value. The significant hybridization of Ba(d), Ge(s) and Se(p) was observed from the PDOS analysis of conduction band indicating covalent character in addition to ionic character. The calculated optical properties shows anisotropy. The optical gap are in energetic order of yy<xx<zz, the onset of yy is at 2.05 eV, xx at 2.38 eV and zz at 2.58 eV, which agreed well with predicted value of 2.6-3.0 eV [8]. Our result shows that Ba_2GeSe_4 potential as wide band gap material [32] with potential applications in multi-junction solar cells. [33].

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