

THE POTENTIAL INTEGRATION OF $\text{Ba}_2\text{InSbO}_6$ DOUBLE PEROVSKITE OXIDES FOR ELECTRONIC DEVICES FABRICATION

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Abstract: In the last years double perovskites elements are emerging as possible elements to use for electronic devices fabrication because of their interesting properties. In this study, we explored the ability to develop new electronic devices based on the double perovskite $\text{Ba}_2\text{InSbO}_6$ through a theoretical investigation of optical and electronic properties using density functional theory as implemented in Wein2k code. The compounds show a semiconductor nature with a direct bandgap of 1.62 eV. Furthermore, the calculated optical properties show that the material strongly absorbs and reflects the incident radiation in the visible and ultraviolet range. Therefore, the $\text{Ba}_2\text{InSbO}_6$ compounds can be effectively used in optoelectronic devices.

Keywords: Perovskites, optoelectronic, Wein2k.

1. Introduction

Currently perovskite materials have been established as a family of materials for energy conversion and storage [Szuromi et al (2017); Kumar et al (2016)], especially for light emitting diodes and solar cells technologies in which those materials have become a promising technology due to their high light absorption coefficient, long carrier diffusion length, and solution processibility [Wang et al (2019); Jiang et al (2019); Jung et al (2019); Wu et al (2019); Deng et al (2019); Khandy et al (2017a)]. The double perovskite oxide compounds with the general formula $\text{A}_2\text{BB}'\text{O}_6$, exhibit novel electronic and magnetic properties such as ferromagnetism (e.g. $\text{Sr}_2\text{SnMnO}_6$ [Khandy et al (2017b)], $\text{La}_2\text{CoMnO}_6$ [Joseph et al (2001)]), metallicity (e.g. $\text{Sr}_2\text{CrReO}_6$ [Kato et al (2002)]), half-metallicity (e.g.

$\text{Sr}_2\text{SnFeO}_6$ [Khandy et al (2017b)], colossal magnetoresistance (Sr_2FeWO_6 , $\text{Sr}_2\text{Fe-MoO}_6$ [Chan et al (2005) ; Kobayashi (1998)]), multiferroicity (e.g TbMn_2O_5 , $\text{Ba}_2\text{NiMnO}_6$ [Hur et al (2004); Azuma et al (2005)]), magneto-optic properties (e.g $\text{Sr}_2\text{CrOsO}_6$, $\text{Sr}_2\text{CrReO}_6$ [Das et al (2008)]) and magneto-dielectric properties (e.g $\text{La}_2\text{NiMnO}_6$ [Rogado et al (2005)]). Based on these properties DPO have a huge potential in the technological field, they are used in magnetic memories, storage devices, optical resonators, filters, etc. In general, the A or AA' sites in the general formula of DPO represent alkaline-earth, rare earth, or transition metals (Ba, Sr, Mn, Pb, etc.) and the B and B' sites are transition metals such as Sb, Ta, Sc, Fe, Mo, Co, W, V, Mo, etc. [Kobayashi et al (1998); Bandyopadhyay et al (2016) ; Bonin et al (1995)].

In the last years, a lot of works has oriented to the study of optoelectronic and thermoelectric properties of double perovskites because of the potential they provide to incorporate a variety of metal cations [Slavney et al (2016); Tariq (2020)]. The organic based and Pb containing double perovskites (e.g $\text{CH}_3\text{NH}_3\text{Pb}_{1-x}\text{Cu}_x\text{Br}_3$, CsPbX_3 ($X = \text{Cl}$ and Br) have an excellent photoluminescent and low-cost. but the toxic Pb limits their applications [Qiao et al (2019); Liu et al (2020); Liu et al (2022)]. In the other hand, Slavney et al. have studied the optoelectronic properties of $\text{Cs}_2\text{AgBiBr}_6$ double perovskite and they suggested that inorganic and hybrid compositions are potential candidates for optoelectronic and photovoltaic applications due the need to explore them further [Slavney et al (2016)]. In addition, Volonakis et al. performed a theoretical and experimental investigation of $\text{Cs}_2\text{InAgCl}_6$ double perovskite and noticed there a direct bandgap of 3.3 eV and they proposed that the bandgap of these compositions can be reduced by mixing the halides [Volonakis et al (2017)]. In another report, Kangsabanik et al. performed an intuitive study and revealed how the double perovskites are overtaking simple perovskite due to their higher stability and better conversion efficiency [Kangsabanik et al (2018)]. Chakraborty et al. made a comprehensive report regarding how scalability, stability of double perovskites intensifies their use for photovoltaic and solar cell applications [Chakraborty et al (2017)]. On this point, the world highest power conversion efficiency (PCE) of Perovskite solar cells has reached 47.1% achieved by Geisz et al. [Meng et al (2017)] wich is 11.1% higher than the already reported value for thin-film solar cells.

However, the applicability of double perovskites for photovoltaic applications faces different issues, more important the large bandgap value of those materials.

Here in our study, we probed the optical, mechanical, and transport characteristics of X_2InSbO_6 ($X = \text{Sr}$, Ba) double perovskites. A high structural and thermodynamic stability presented their potential for solar cell as well as thermoelectric applications.

So far, there are extremely limited experimental and theoretical reports on these materials. Therefore, for energy systems, we believe that this study would provide a sound basis and appropriate acknowledging to use these compositions.

Thereby, the primary purpose of this work is to provide some information about the physical properties of $\text{Ba}_2\text{InSbO}_6$ double perovskite oxide and provide some additional information of the existing data. In this view, a Density Functional Theory (DFT) calculation has been initiated using Full-Potential Linear Augmented Plane Wave (FP-LAPW) [Bandyopadhyay et al (2016)] as implemented in the WIEN2k code [Bonin et al (1995)]. The present report was structured as follows: We start with computational procedures, and then we analyze and discuss the obtained results about the studied physical properties of $\text{Ba}_2\text{InSbO}_6$ perovskite. Finally, a conclusion of the main results is given in the last section.

2. calculation method

The study of perovskite compound Ba₂InSbO₆ was done by the use of the FP-LAPW method which is implemented in WIEN2k code [Hyberstsen et al (1989)] within the framework of the density functional theory to perform the first principal calculations. In the FP-LAPW method, the solution to the Kohn Sham equations is performed self-consistently and the augmented plane wave plus local orbital basis set is incorporated to represent the system properties for all atoms and their corresponding orbitals. All the calculations were converged with respect to Brillouin zone (BZ) sampling and the size of the basis set. Brillouin zone (BZ) integrations within the self-consistency cycles were performed via a tetrahedron method. the modified Becke-Johnson (mBJ) exchange potential proposed by Tran et al [2009] with the form expressed in the equation (1) have been used to treat the exchange and correlation effects of the Ba₂InSbO₆ double perovskite oxide

$$v_{x,\sigma}^{MBJ}(r) = cv_{x,\sigma}^{BR}(r) + (3c - 2) \frac{1}{\pi} \sqrt{\frac{5}{12}} \sqrt{\frac{2t_{\sigma}(r)}{\rho_{\sigma}(r)}} \quad (1)$$

Where $\rho_{\sigma}(r)$ is the electronic density, t_{σ} is the kinetic-energy density, and $v_{x,\sigma}^{BR}(r)$ is the Becke-Russel (BR) exchange potential.

The separation energy between core and valence electrons is -6,0 Ry. The number of plane waves is limited by $RMT \times K_{max} = 7$. The l_{max} parameter is taken to be 10 and the Fourier expanded charge density is $G_{max} = 12$. The integration of first Brillouin zone is performed with (10 x 10 x 10) k-points grid in reciprocal space. The crystal structure is viewed through XCrySDen program [Kokalj (2003)].

3. Results and discussions

3.1. Structural properties

The double perovskite Ba₂InSbO₆ cubic structure (Fm-3m) is shown in Fig. 1. In the crystal, the Ba²⁺ cations are located at positions (0.25, 0.25, 0.25), whereas the In and Sb cations are located at (0.5, 0, 0) and (0, 0, 0), respectively. Conversely, the O anion is located at (0.257, 0, 0) in the unit cell. The structural properties of the Ba₂InSbO₆ compounds are computed using an energy minimization procedure.

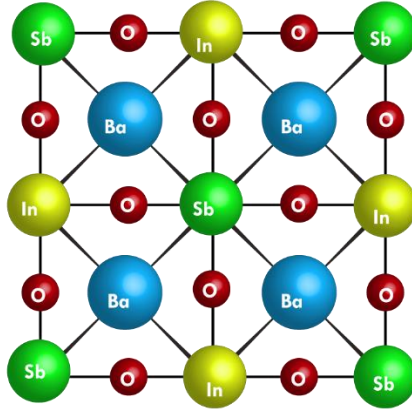


Fig. 1. Illustration of the unit cell structure of $\text{Ba}_2\text{InSbO}_6$.

The optimization plots of the $\text{Ba}_2\text{InSbO}_6$ compounds are presented in Fig. 2 where the local minimum values of the fitted curve are the ground states of the compounds. At the ground state, the structural parameters are computed and these structural parameters are calculated using the unit cell volume at ambient pressure.

The structural parameters, including the lattice constant, $a_0(\text{\AA})$, bulk modulus, $B_0(\text{GPa})$, optimization volume, $V_0(\text{a.u.}^3)$ and the ground state energy, $E_0(\text{Ry})$ are listed in Table 1. The calculated values of lattice constant (a) and bulk modulus (B) of our compound are summarized in table 2. We notice that the obtained results are in good agreement with the theoretical and experimental works [Lalhriatpuia et al (2019); Baba-Kishi et al (2001); Rajavardhan et al (2016)].

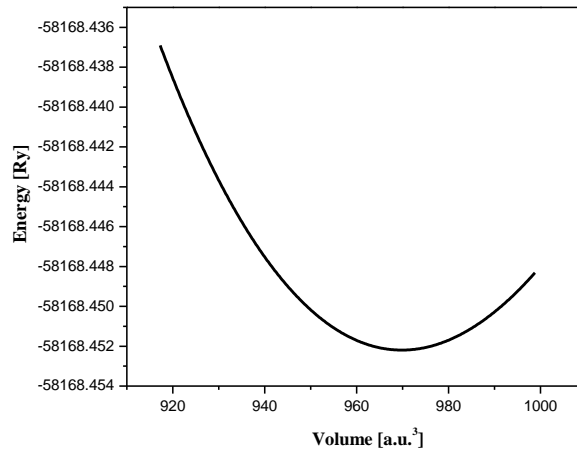


Fig. 2. Volume optimization of $\text{Ba}_2\text{InSbO}_6$.

Table 1. Experimental and the calculated values of the lattice constant, a_0 (Å), bulk modulus, B_0 (GPa), pressure derivative of B_0 (BP), optimization volume, V_0 (a.u.³), and the ground state energy, E_0 (Ry) of Ba₂InSbO₆.

compound	V_0 (a.u. ³)	Lattice constant a_0		Bulk		E_0 (Ry)
		Calculated (a_0)	Experimental (a_0)	B_0 (GPa)	BP	
Ba ₂ InSbO ₆	969.8354	8.3148 Å	8.2938 [25]	140.1448	5.2517	-58168,45

Table 2. Calculated lattice constant (a) and Bulk modulus (B) of Ba₂InSbO₆ compound.

Compound	a(Å)	B_m (GPa)	Methods
Ba ₂ InSbO ₆	8.2938	140,144	Our work
	8,20		Exp.
	8.1866- 8.1967	147,709	Theory

3.2. *Electronical properties*

3.2.1. *Band structure*

The band structure calculation for Ba₂InSbO₆ compound is depicted in Fig. 3. The results show that the material have a direct energy bandgap of 1.62 eV, where the valence band (VB) maxima and the conduction band (CB) minima are at the Γ point in the Brillouin zone. Importantly, materials with such bandgap value work well in visible light device applications [R.F. Berger and J.B. Neaton (2014); Wittmann et al (1981)].

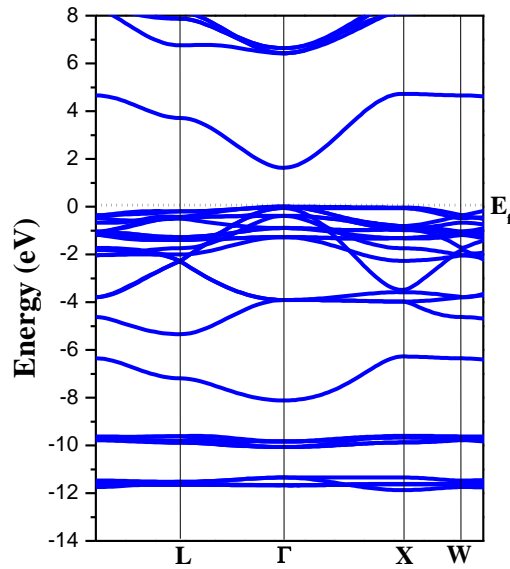


Fig. 3. Band structure plot.

3.2.2. Density of states

The partial density of states (PDOS) of the $\text{Ba}_2\text{InSbO}_6$ compound is presented in Fig. 4. It was observed that the various bands appear in the energy range of -8 eV to 0 eV due to the main contribution of the O-p state with a minor contribution of the Sb-p, Sb-s and In-d states. On the other hand, from 0 eV to 5 eV, the major contributor states are Sb-s with a little contribution from the O-p states.

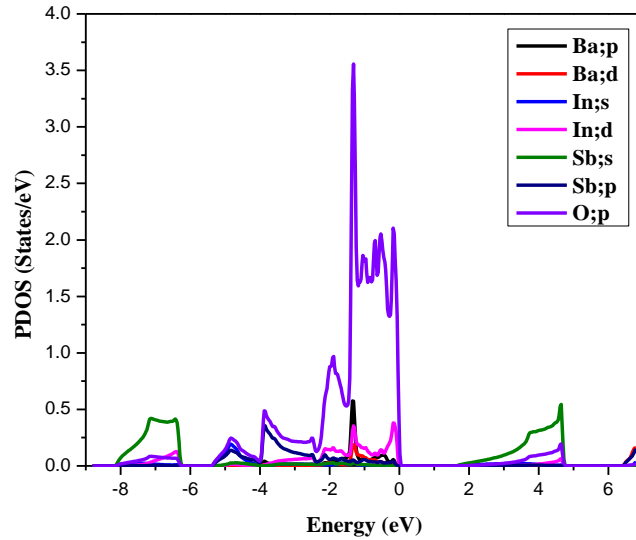


Fig. 4. Partial density of state plot

3.3. Optical properties

It is known that the complex dielectric function $\varepsilon(\omega)$ describe the optical behavior of a material. $\varepsilon(\omega)$ is an addition of a real part ($\varepsilon_1(\omega)$) and imaginary part ($\varepsilon_2(\omega)$) as expressed below [Berger et al (2014)].

$$\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega) \quad (2)$$

From the electronic band structure of the material, the imaginary part $\varepsilon_2(\omega)$ of the complex function $\varepsilon(\omega)$ in cubic symmetry can be calculated as follows:

$$\varepsilon_2(\omega) = \left(\frac{\hbar^2 e^2}{\pi m^2 \omega^2} \right) \sum_{c,v}^n \int d^3k \langle c_k | p^\alpha | v_k \rangle \langle v_k | p^\beta | c_k \rangle \chi \delta(\varepsilon_{c_k} - \varepsilon_{v_k} - \omega) \quad (3)$$

Here, p is the momentum matrix element between states of band α and β within the crystal momentum k . c_k and v_k are the crystal wave functions corresponding to the conduction

and valence bands with crystal wave vector k . $\epsilon_1(\omega)$ can be calculated from $\epsilon_2(\omega)$ using Kramers-Kronig relationship [Berger et al (2014)] expressed as follows:

$$\epsilon_1(\omega) = 1 + \frac{\pi}{2} p \int_0^{\infty} \frac{\omega' \epsilon_2(\omega')}{(\omega')^2 - \omega^2} d\omega' \quad (4)$$

Where p denotes the principal value of the integral. $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$ allow us to gain the knowledge to obtain the primary importance of optical functions like the refractive index $n(\omega)$, extinction coefficient $k(\omega)$, absorption coefficient $\alpha(\omega)$ and reflectivity $R(\omega)$ are used given as follows :

$$n(\omega) = \frac{1}{\sqrt{2}} \left[\sqrt{\epsilon_1^2(\omega) + \epsilon_2^2(\omega)} + \epsilon_1(\omega) \right]^{\frac{1}{2}} \quad (5)$$

$$k(\omega) = \frac{1}{\sqrt{2}} \left[\sqrt{\epsilon_1^2(\omega) + \epsilon_2^2(\omega)} - \epsilon_1(\omega) \right]^{\frac{1}{2}} \quad (6)$$

$$\alpha(\omega) = \sqrt{2}\omega \left[\sqrt{\epsilon_1^2(\omega) + \epsilon_2^2(\omega)} - \epsilon_1(\omega) \right]^{\frac{1}{2}} \quad (7)$$

$$R(\omega) = \left| \frac{\sqrt{\epsilon(\omega)} - 1}{\sqrt{\epsilon(\omega)} + 1} \right|^2 \quad (8)$$

Optical properties, especially the absorption spectrum and the optical reflectivity are shown in Fig. 5. The optical absorption describes the ability of a compound to absorb light [Wittmann et al (1981)]. From the absorption spectrum it's clear that the Ba₂InSbO₆ presents a strong absorption behavior in the visible area.

The optical reflectivity is a one of the most fundamental parameters identifying the optical properties of a sample. It describes the ability of a samples to reflect radiation. In fig. 5 (b), plots of the obtained reflectivity of Ba₂InSbO₆ are presented, we can clearly see that the reflectivity is small in the low energy area (up to 5 eV), which reflects that interbond transition can't happen in the far infrared spectrum due to the band behavior of this semiconductor. although, more reflection occurs in the UV region. Between 11 and 14 eV. The reflectivity values also demonstrate that this compound has possible applications in anti-reflection coatings and in the deep ultraviolet region.

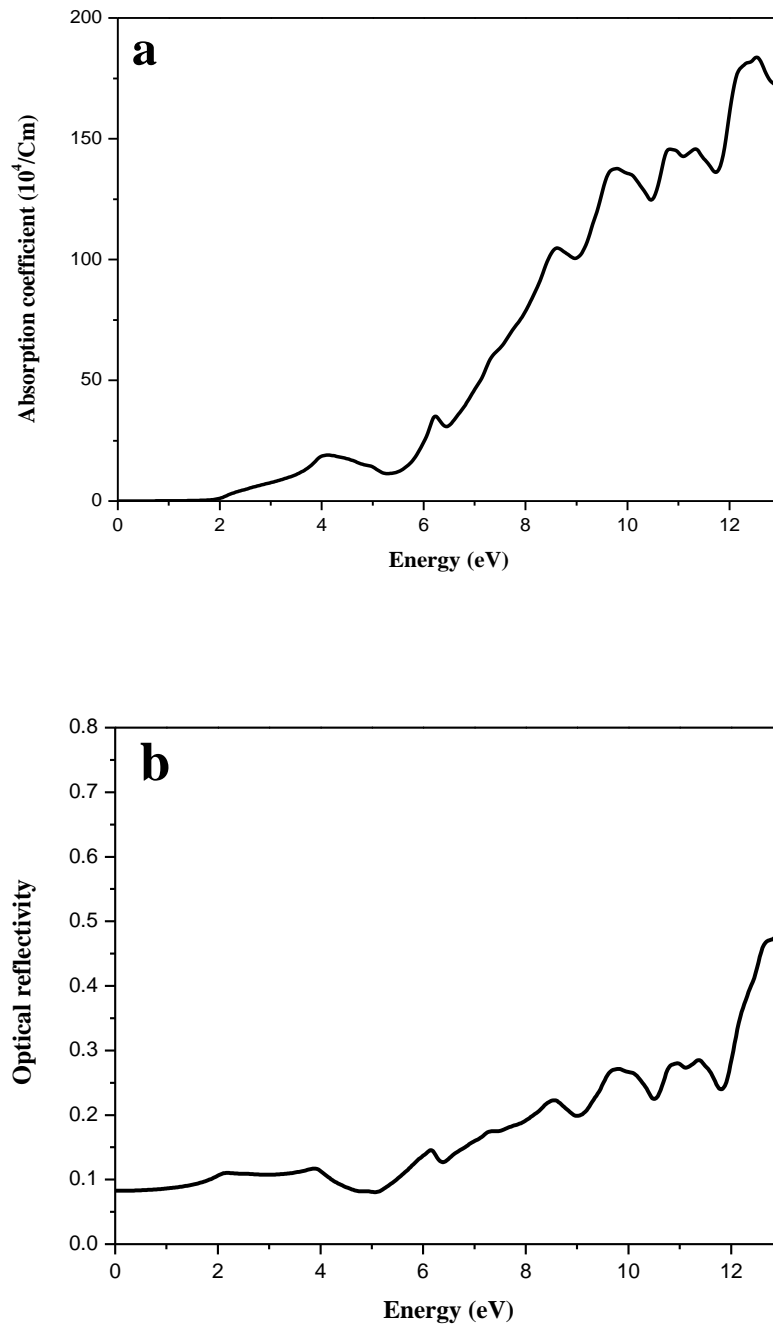


Fig. 5. Plots of (a) absorption coefficient and (b) Optical reflectivity.

Based on the reported data, the double perovskite compounds $\text{Ba}_2\text{InSbO}_6$ show a cubic structure and a direct bandgap of 1.62 eV. Makes them a best candidate for applications that require the emission of photons via the radiative recombination of electrons and holes, making them suitable for uses in light-emitting diodes, lasers, and optical sources

[Bandyopadhyay et al (2016)]. The absorption spectra prevue the potential of the studied materials for visible light device applications. for photovoltaics applications, materials with such behavior are proposed to be used for the development of tandem solar cell. It is also important to note that materials with such band gap value have been used in some experimental studies for photocatalytic water splitting [Kato et al (1999); Nakanishi et al (2017); Kato et al (2003)].

4. Conclusion

In this paper, we have comprehensively investigated a density functional theory investigation on optic and electronic properties of Ba₂InSbO₆ Double perovskite oxide. The structural optimization was carried out using generalized gradient approximation (PBEsol-GGA). Moreover, the new Becke-Johnson (new-mBJ) was utilized in the framework of PBEsol-GGA to precisely evaluate the electronic bandgap. the material demonstrates a direct bandgap of 1.62 eV and a strong absorption behavior in the visible area. Based on the bandgap value and the optic properties, the studied material illustrates strong light absorption characteristics in visible and ultraviolet regions, signifying its importance for optical and optoelectronics applications.

5. References

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