Predictive Study of New Chalcopyrite $ZnGeP_2$ for Photovoltaic Cells

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Abstract

In this paper, we have accomplished a predictive study of structural, elastic, electronic and optic properties of chalcopyrite semiconductor $ZnGeP_2$. The calculations have been approvedusing generalized gradient approximation (GGA-PBE) to determine the potential for exchange and correlation via density functional theory (DFT) implemented in CASTEP code. The obtained results predict that the composite $ZnGeP_2$ behaves in, as a somewhat brittle and ductile,manner based on analysis of elastic constants and their derived parameters. Furthermore, our alloy has a direct bandgapof 1.269 eV, which is ideal for the solar light absorber.

Keyword:*DFT; GGA PBE; Chalcopyrite; $ZnGeP_2$; Proprieties.*

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Introduction

Many researchers are rigorously working on how to improve the efficiency of photovoltaic solar cells to generate more electrical energy. Photovoltaics can play an important role in the transition to a system sustainable energy supply for the 21st century and is likely to cover a significant part of the electricity needs of several countries.Algeriawith its geographical location, occupies a privileged position in the exploitation of solar energy with a duration of sunshine which varies from

2650 hours / year in the north to 3500 hours / year in the south, one of the highest in the word.It receives the maximum energy during the summer solstice (June 21 or 22) and the minimum during the winter solstice (December 21or 22).The conversion of light into electricity (photovoltaic conversion) occurs in semiconductor materials.

The choice of semiconductor materials will give a variation in the energy of gap and a variation in the efficiency of the photovoltaic cells.

Since the discovery in 1957 of the photovoltaic properties of silicon hydrogenated amorphous (a-Si:H),crystalline silicon has remained thebest semiconductor known. It has different advantages. It's abundant, can be easily extracted and doped but it is not the greatest adapted to the solar spectrum (between 1.5 and 1.7 eV).

Recently, chalcogenideand chalcopyrite intermetallic compounds with theformula ABX2 have been largely investigated as promisingmaterials in thermoelectric applications[1,2,26].

Generally, an ABX_2 ternary compound is a mixture between two zin-blend AX and BX structures by introducing a change of symmetry group from ($\bar{F}43m$ n° 216) to ($\bar{I}42d$ N°122) and, thus, characterizing a quadratic chalcopyrite structure.[2, 3 Berber]. For these types of crystals, the lattice parameter c should be equal to 2a. But, by the alternation of the cations, the chalcopyrite presents structural modifications. These changes are anionic displacement, which influences only on the distances between the first neighbors, and a quadratic compression c/a. This ration is generally different from the ideal value 2. The difference *(2-(c/2a))*is the measure of the tetragonal distortion [3]

The objective of this work is to achieve a better understanding of the structural, elastic, electronicand optic properties of the $ZnGeP_2$ chalcopyrite compound. This paper is organized as follows: computationaldetails are presented in section 2 after the introduction, and section 3 displays the results and discussions. The conclusions are summarized in section 4. We ended our paper with references that we used to develop our ideas.

2.Computationnel Methods

In this work, we calculated the physical properties and structural geometric optimizationusing the CASTEP code [4, 5]within the pseudopotential methodand in the context of the DFT[6]theory. The generalized gradient approximation (GGA) parameterized by Perdew-Burke-Ernzerh of (PBE) [7] was adopted for the exchange-correlation functional, a Monkhorst-Pack special k points mesh of 15×15×15 was used in the Brillouin zone integrations with a self-consistent field tolerance of 5.10^{-6} eV and a cutoff energy of 250 eV. The following atomic shells have been treated as valence states: $4s^2$ $3d^{10}$ for Zn, $3d^{10}$ $4s^2$ $4p^2$ for Ge, $3s^2$ $3p^3$ for P.

Allinformation, about our calculations, are given in Table 1

Table 1. Lattice parameter, positions for optimizationof ZnGeP2

	Latticeparameter (A°)	Angles		Wyckoff positions		
			Positions	Zn	Ge	P
ZnGeP2	a= 5.44	α=90°	x	1/2	0	¾
	b=5.44	β=90°	y	0	1/2	u= 0.2545
	c=10.73	γ=90°	z	3/4	3/4	7/8

We note that the value of distortion is $2-(c/2a)=1.014$We see that $c\approx 2a$ so we calculate the geometric parameter u[8]

$$u=\frac{1}{2}-\left(\frac{c^2}{32a^2}-\frac{1}{16}\right)^{\frac{1}{2}}=0.2545 \qquad (1)$$

3.Results and Discussions

3.1 Structural properties

At first, we have using information given in Table 1, and we have found that our compound $ZnGeP_2$ has a tetragonal structure fig.1. It is structured and crystallized in the tetragonal $I\bar{4}2d$ space group (122). Zn^{2+} is bonded to four equivalent P^{3-} atoms to form ZnP_4 tetrahedra that share corners with four equivalent ZnP_4 tetrahedra and corners with eight equivalent GeP_4 tetrahedra. Ge^{4+} is bonded to four equivalent P^{3-} atoms to form GeP_4 tetrahedra that share corners with four equivalent GeP_4 tetrahedra and corners with eight equivalent ZnP_4 tetrahedra. P^{3-} is bonded to two equivalent Zn^{2+} and two equivalent Ge^{4+} atoms to form corner-sharing PZn_2Ge_2 tetrahedra.The conventional cell contains 16 atoms where possible oxidation states Zn^{2+}, P^{3-}, Ge^{4+}.

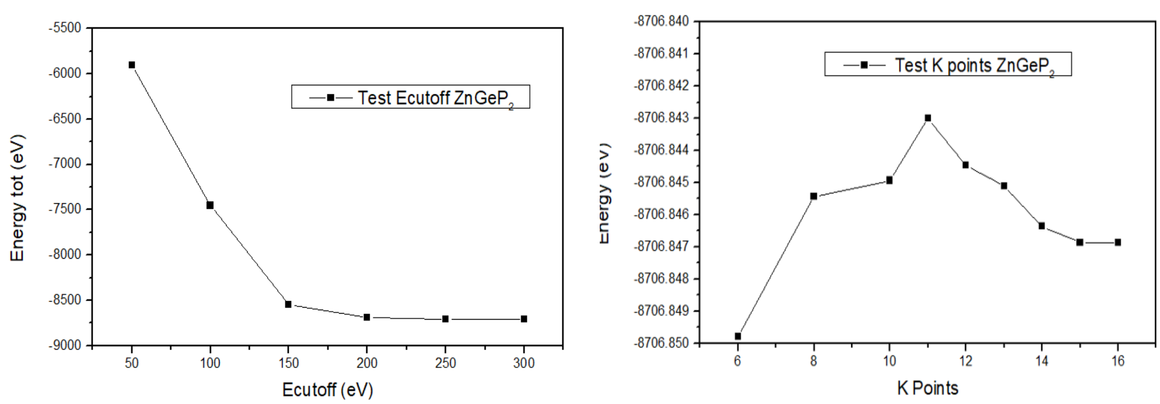

Fig 1 (a) Convergence test within E cutoff (b) Convergence test within K points

We noted that the energy cutoff and the K points for the convergence are 250 eV and $15\times 15\times 15$respectively.

$ZnGeP_2$compound is chalcopyrite structured and crystallizes in the tetragonal$I\bar{4}2d$space group (122)

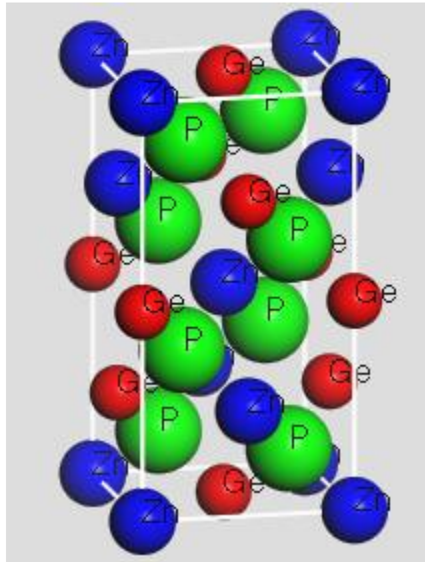

Fig.2. The structure of $ZnGeP_2$ compound.

For structural optimization, we have calculated the energy variation versus the unit cell volume using the Murnaghan equation of state [9, 10,11,25]. This equation depends essentially on two parameters the bulk modulus B_0 and its first derivative pressure B'_0. In general, these two coefficients are determined by adjusting the energy versus the volume; the isothermal compressibility modulus B is related to the curvature of the function E(V) by the following Murnaghan function:

$$E(V) = E_0 + B_0 V_0 + \left[\frac{1}{B'_0(B'_0-1)}\left(\frac{V_0}{V}\right)^{B'_0-1} + \frac{V}{B'_0 V_0} - \frac{1}{B'_0-1}\right] \qquad (2)$$

Where v_0, E_0, B_0 and B'_0 , are respectively the volume, total energy, compressibility modulus and its first pressure derivative at equilibrium. Which are concluded by fitting the curve E(V) within Murnaghan function, as shown in fig.2.

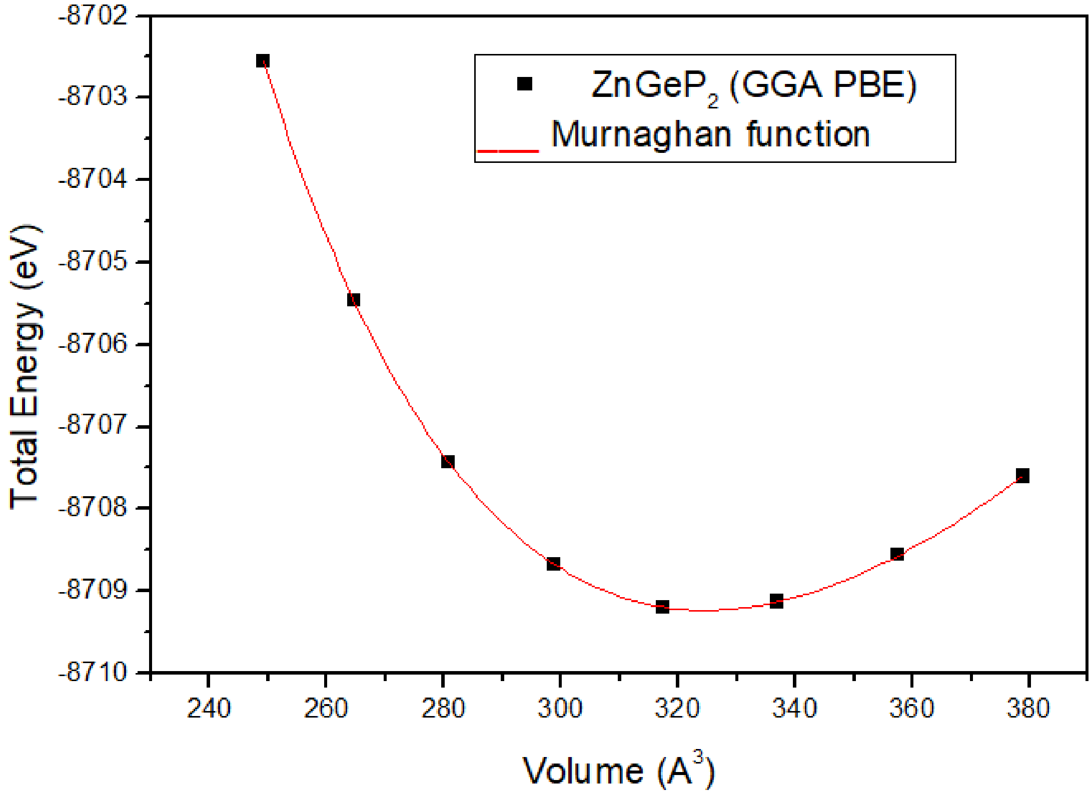

Fig. 3. Total Energy versus volume for $ZnGeP_2$ compound

Table 2. Lattice parameter, Bulk modulus and it's pressure derivative, Energy at equilibriumof ZnGeP2

ZnGeP2	a_0 (A°)	B (GP)	B' (GP)	E_0(eV)
	5.455	74.172	4.207	-8709.23444

3.2. Electronic properties

3.2.1/Band structure

The band structure is a fundamental characteristic to study the electronic properties of solids. The width of energy band gap E_g which is the energy separating the valence band (top) to the conduction band (bottom), is a significant plateful of delivered by the band structure. The variation of E_g versus T in semiconductor materials was proposed by Varchni as [12]

$$E_g(T) = E_g(0) - \frac{\alpha T^2}{T+\beta} \quad (3)$$

This equation becomes in the case of linear behavior of E_g

$$E_g(T) = E_g(0) - \alpha T \quad (4)$$

The band structure of $ZnGeP_2$(Fig.4.) shows that this material has an indirect gap type. The analyses of the obtained curves show that our compound has an E_g = 1.269 eV from G-X high symmetry point. Located between G for valence band maximum (VBM) and X for conduction band minimum (CBM). Therefore, it can captivate a wide range of the solar spectrum. So, chalcopyrite $ZnGeP_2$will be a good candidate to do this role because its gap is equal to 1.269 eV, by comparison with previous works which equal 1.18 eV[13] it is better.

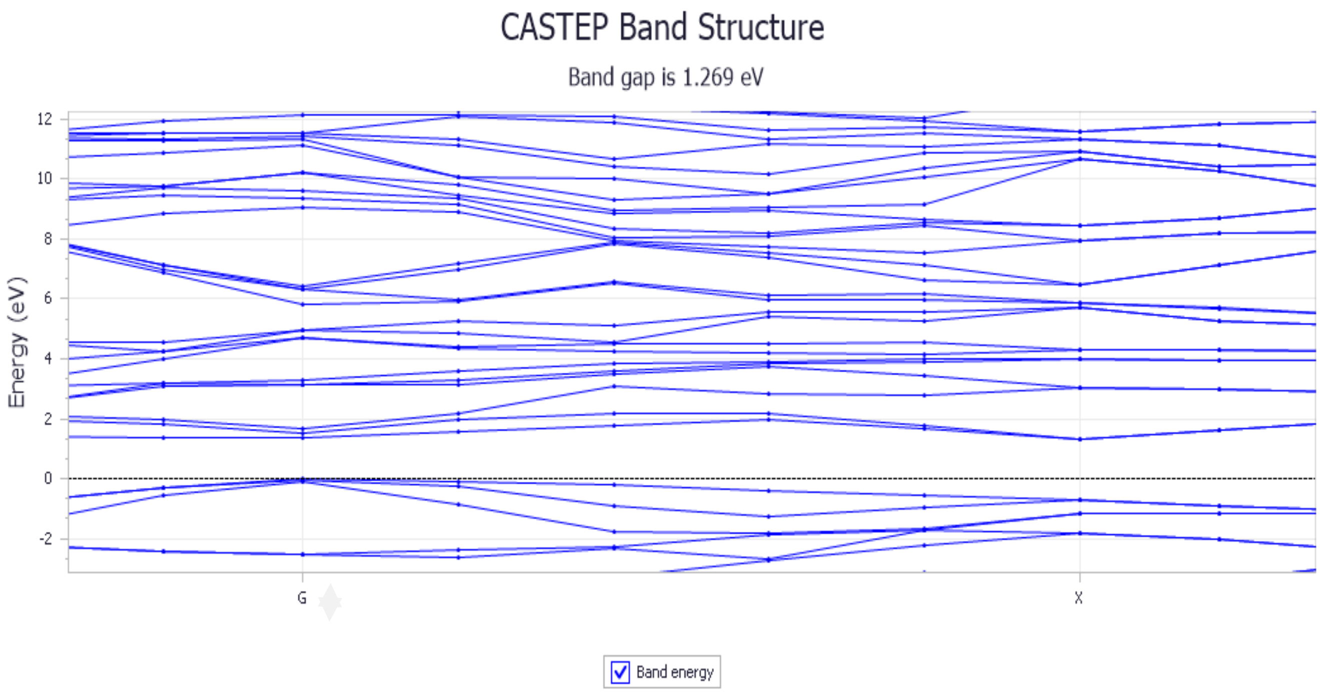

Fig.4.Band structure of $ZnGeP_2$

3.2.2. The density of states

The analyses of the obtained curves show that our compound $ZnGeP_2$ has E_g = 1.269 eV from G–X high symmetry point. Our compound has an indirect energy band gap (E_g) which is closer to the desired value. Otherwise, $ZnGeP_2$ alloys exhibits a semiconductor nature. Hence, it can captivate a wide range of the solar spectrum.

Fig 5 shows the total state density curves of ZnGeP2

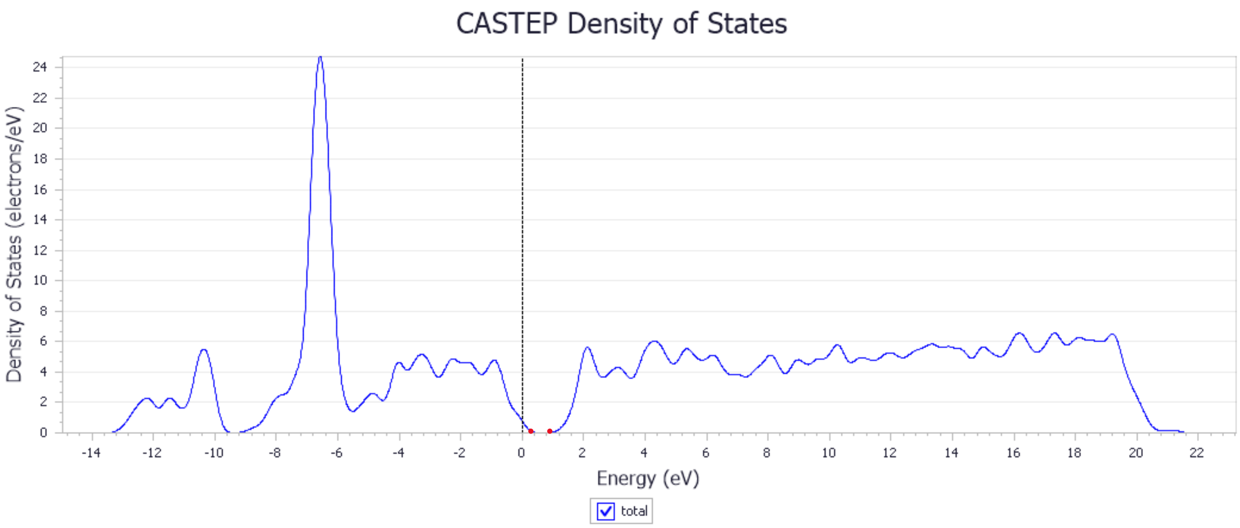

Fig.5.Total DOS

In an attempt to elucidate the nature of the electronic band structure, we have also calculated the total density of states. Most transport properties are determined on the basis of the knowledge of the density of states. It also allows you to know the nature of chemical bonds in a material and therefore the charge transfer between orbitals and atoms.

3.3. Optical properties

we present the evolution of the real ε1(ω) and imaginary ε2(ω) part of the dielectric function ε(ω), as a function of the photon energy in the energy range [0–36 eV] for $ZnGeP_2$compounds. The imaginary part of the optical function's dispersion represents the optical absorption in the crystal were calculated from the momentum matrix elements between occupied and unoccupied electronic states over the Brillouin zone BZ.

Fig. 6. shows the dielectric function

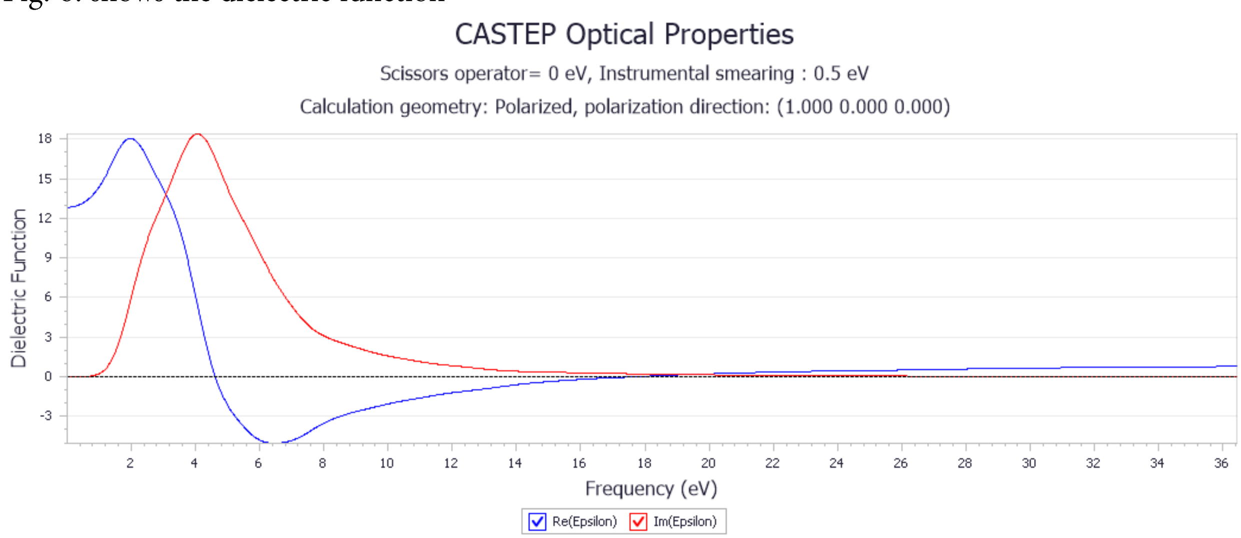

Fig.6. Dielectric function of ZnGeP2

The imaginary part show that there exists an optical gap for our compound, which supports our previous observation that this compound shows a semiconducting character. It is clear, that there are several peaks appears in the energy range of (0–18) eV. From the analysis, the density of states, the main peak in the imaginary part corresponds to the transitions between s/p-(transition metals) states between the valence and conduction bands, where the energy range in this negative value of ε1(ω) interprets that the totally incident photons are reflected. The ε1(ω) become constant from 26eV.

Fig.7.shows the absorption coefficients α(ω) of ZnGeP2 compounds. It is observed that the absorption edge starts from energy 0 eV of, and there is a wide band absorption up to ~30.8 eV for our compound. The maximum values of α(ω) are 270 10^3 cm^{-1}. Our results suggest that the alloy $ZnGeP_2$ is beneficial in optoelectronic applications between 0 eV and 6 eV.

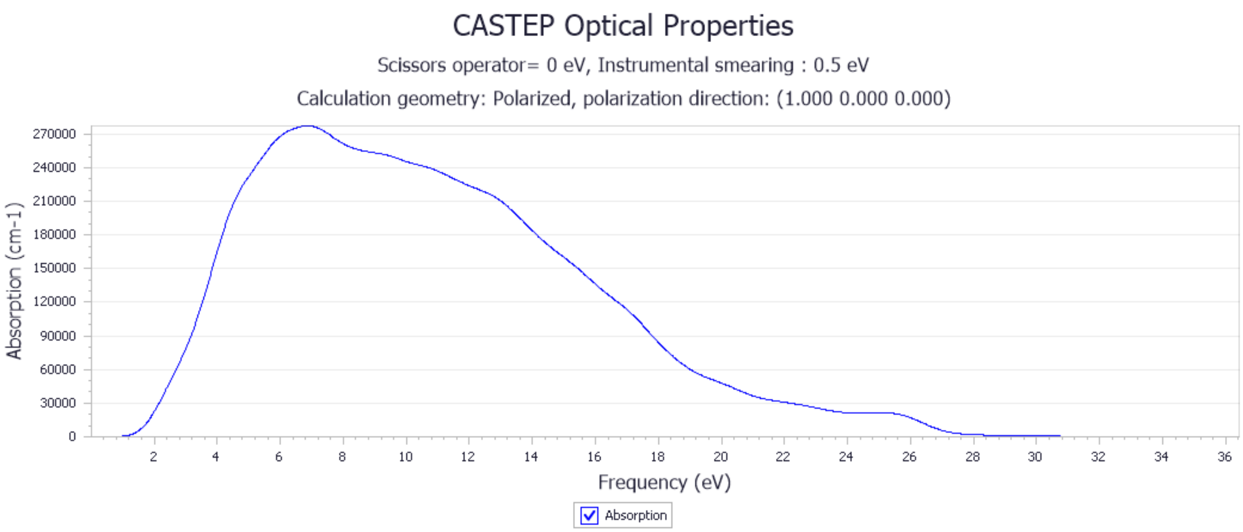

Fig.7.Absorption of ZnGeP2

In Fig. 8, presented the reflectivity R(ω) versus photon energy of ZnGeP2 compound. The reflectivity R(ω) is 34% at zero value, confirming the semiconducting nature of this compound that are seen it before in Fig. 3 . We can also observe, the maximum values of R(ω) at 58% forour compound. We can be seen, there aresix peaks located at$< 20\ eV$.Our result indicates that our compound behaves like semiconductor.

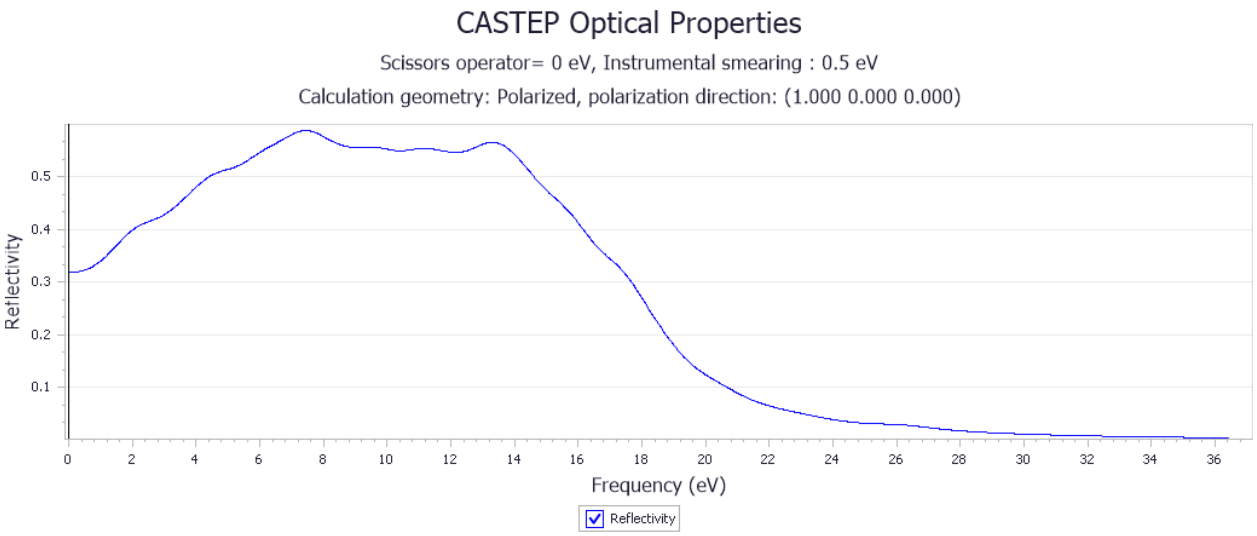

Fig.8. Reflectivity *of ZnGeP2*

The reflectivity spectrum is shown in figure 8. This material is reflective in the energy range (0-26) eV. From this energy, we notice that the reflectivity becomes almost zero. The maximum values of the reflectivity obtained for this compound is approximately equal to 58%.

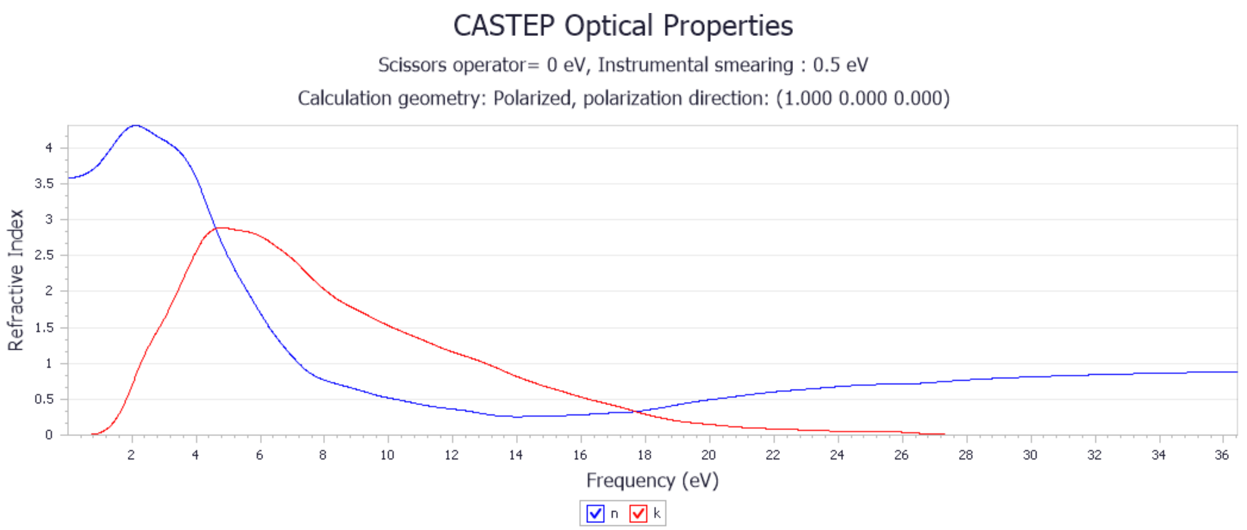

Fig.9. Refractive index of ZnGeP2

The variation of the refractive index $n(\omega)$ is given in Fig. 9 for ZnGeP2 compound. We also evaluated the n (0), this value is 3.8. This value be good enough the relation n (0)= (ε(0))1/2. On the other hand, the refractive index $n(\omega)$ takes the maximum values at energies 2 eV for our compound, beyond this value the $n(\omega)$ decreases with increasing energy.

3.4. Elastic properties

The linear response of an elastic crystal of arbitrary symmetry to a deformation infinitely small is determined by the elastic constants [14,15,16,23]. These constants form a rank four tensor, which can be reduced to 6×6 matrix. Taking into account the symmetry of the directions of the deformation as well as using Voigt's notation [14,15,24], and according to Neumann's principle, the particular symmetry of a crystal cell can impose additional restrictions on the elements of the elastic matrix. Since our chalcopyrite belong to the $\bar{I}42d$ (122) type symmetry space group, with the Laue class 42m; the tensor of elastic constants takes the following form [17].

$$\begin{bmatrix} C11 & C12 & C13 & 0 & 0 & 0 \\ C12 & C22 & C23 & 0 & 0 & 0 \\ C31 & C32 & C33 & 0 & 0 & 0 \\ 0 & 0 & 0 & C44 & 0 & 0 \\ 0 & 0 & 0 & 0 & C55 & 0 \\ 0 & 0 & 0 & 0 & 0 & C66 \end{bmatrix}$$

In this section, the elastic constants calculation is carried out using the theory implemented in CASTEP code. The tetragonal structure requires six elastic constants: C_{11}= C_{22}, C_{12}=C_{21}, C_{13}=C_{31}=C_{23}= C_{32}, C_{33}, C_{44}=C_{55} andC_{66} so we need six distortions in volume conserving.

Table3. Constant elastic of ZnGeP2

Compound	C11	C12	C13	C33	C44	C66
ZnGeP2	121	51	53	119	63	62

So, the tensor becomes

$$\begin{bmatrix} 121 & 51 & 53 & 0 & 0 & 0 \\ 51 & 121 & 53 & 0 & 0 & 0 \\ 51 & 53 & 119 & 0 & 0 & 0 \\ 0 & 0 & 0 & 63 & 0 & 0 \\ 0 & 0 & 0 & 0 & 63 & 0 \\ 0 & 0 & 0 & 0 & 0 & 62 \end{bmatrix}$$

The mechanical stability conditions, recognized as Born-Huang criteria for the tetragonal structure, are expressed as [18]:

$C_{11}, C_{33}, C_{44}, C_{66} > 0$, $C_{11} > |C_{12}|$, $(C_{11}+C_{12})C_{33} > 2C_{13}^2$, $C_{44} > 0$, $C_{66} > 0$

Mechanical constants

We summarized our result in the table 4

Table 4 Mechanic constant of ZnGeP2

Bulk Modulus, Voigt	75GPa
Bulk Modulus, Reuss	75GPa
Bulk Modulus, Voigt-Reuss-Hill	75GPa
Shear Modulus, Voigt	51GPa
ShearModulus, Reuss	47 GPa

ShearModulus, Voigt-Reuss-Hill	49 GPa
Poisson's Ratio	0.23
Universal Anisotropy	0.45

$$G = \frac{G_V+G_R}{2} A = \frac{C_{33}-G_{13}}{C_{11}+C_{12}-C_{33}} E = \frac{9BG}{3B+G} \upsilon = \frac{1}{2}(1-\frac{E}{3B}) \quad (5)$$

$$G_v = \frac{1}{30}(M+3C_{11}-3C_{12}+12C_{44}+6C_{66}) B_v = \frac{1}{30}\{2(C_{11}-C_{12})+C_{33}+4C_{13}\} \quad (6)$$

$$G_R = 15\left\{\frac{18B_V}{C^2}+\frac{6}{(C_{11}-C_{12})}+\frac{6}{C_{44}}+\frac{3}{C_{66}}\right\}^{-1} \quad (7)$$

Where $M = C_{11} + C_{12} + 2C_{33} - 4C_{13}$ and $C^2 = (C_{11} + C_{12})C_{33} - 2C_{13}^2$ (8)

It is noted that the value of Bulk modulus calculated from the elastic constants at zero pressure (B= 75GPa), has almost the same value as obtained from the smoothing points E_{tot} (V) using Murnaghan's equation of state B= 74.172 GPa. As a result, we can qualify as a good estimate of accuracy and conformity of the elastic constants of our compound ZnGeP2.

The calculated value of the anisotropic parameter of Zener at zero pressure is equal to 0.45 diverges from 1 which indicates that our alloy presents an anisotropic elasticity.

We notice that at zero pressure our alloy presents a covalent contribution since the value of the Poisson's ration is 0.23. We know that the typical value of Poisson's ratio for covalent materials is around 0.1, while for ionic materials is around 0.25 [9,19].

Our compound behaves as a somewhat brittle and ductile material according to the results obtained during the calculation of the B/G ratio which is equal to 1.53 and which is near to 1.75 according to Pugh's empirical formula [9,20]

Once we have calculated the young's modulus E, the bulk modulus B and the shear modulus G, we can obtain the Debye temperature θ_D[9], which is a parameter of fundamental importance closely linked to several physical properties such as calorific heat and melting temperature. At the low temperature, the vibratory excitations result only from acoustic vibrations. A standard method for calculate the Debye temperature from the elastic constants. And relation between the average wave velocity V_m and θ_D given by the following relation:

$$\theta_D = \frac{h}{K_b}\left(\frac{3n}{4\pi V_a}\right)^{\frac{1}{3}} \upsilon_m \quad (9)$$

where$h = 6,62607004 \times 10^{-34}$ $m^2 kg\, s^{-1}$Plank's constant,

$k_b = 1,38064852 \times 10^{-23} m^2 kg\, s^{-2} k^{-1}$ Boltzmann's constantand V_a volume atequilibrium state.

$$\upsilon_m = \frac{1}{3}\left(\frac{2}{v^3{}_t}+\frac{1}{v_l{}^3}\right)^{\frac{-1}{3}} \quad (10)$$

$$v_t = \left(\frac{G}{\rho}\right)^{\frac{1}{2}} \quad (11)$$

$$v_l = \left(\frac{3B+4G}{3\rho}\right)^{\frac{1}{2}} \quad (12)$$

With v_m,v_T and$v_l$represent the average velocity of sound, the transversal velocity and the longitudinal velocity respectively[9, 21, 22]

The results are summarized in table 5

Table 5 Velocity of sound and the Debye temperature of ZnGeP2

Compound	$\rho(g/cm^3)$	$V_0(A°)^3$	Nberof atoms	v_t(m/s)	v_l(m/s)	v_m (m/s)	θ_D(k)
$ZnGeP_2$	4.16	324.69	4	108530 .39	183668.11	27789.04	148.10

4.Conclusion

We have studied the structural, elastic, and electronic properties ofthe chalcopyrite ZnGeP2 using the CASTEP code based on DFT within GGA-PBEapproximation. Concerning the structural properties as latticeparameter, bulk modulus, and its pressure derivative, theresult obtained is compatible with those found in the literature.

For elastic properties, our component is mechanicallystable and it is classified in a somewhat brittle and ductile manner.We gave a predictiveresult of shear modulus,Young modulus, Poisson'sratio, and anisotropy parameter.

Using the quasi-harmonicDebye model, we have estimated the sound velocities v_l,v_t,$v_m$and Debye temperature at zeropressure. Theband structure shows that our component has a semiconductor behavior at zero pressure.

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