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# QUASIPARTICLE BAND STRUCTURE STUDY OF TERNARY MIXED CHALCOGENIDE $Bi_2Te_2S$ AS TOPOLOGICAL INSULATOR

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In this paper, ternary mixed chalcogenide $Bi_2Te_2S$ has been investigated using first principle within the density functional theory (DFT) calculation as well as many body perturbation theory within the GW quasiparticle approximations. For the DFT calculation, we have used generalize gradient approximations (GGA) as exchange correlation function and found that, $Bi_2Te_2S$ has a narrow band gap with a band gap of about 0.058eV, while for the GW calculation, we have evaluated both the quarsi bands structure, and absorption spectrum using random phase approximation (RPA). From the results we have observed that the GW corrections to the DFT- Kohn-Sham eigenvalues increase the fundamental band gap to 0.099eV showing the effect of electron-phonon coupling present in the structure of $Bi_2Te_2S$, while, for the absorption spectrum, it exhibits a close value in agreement with the theoretical results of similar compounds reported by (Anjan A. Reijnders, et. al 2014).

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## 1. Introduction

Recent theoretical and experimental studies of structural and thermoelectric properties of materials that display a topological insulator (TI) behaviour reveal a new quantum state that can be explain the effect of non-degenerate surface [1] which form a Dirac cone at a certain point in a bulk band gap of the materials [2]. A mixed chalcogenide group such as $Bi_2Te_3$, $Bi_2Te_2S$, $Bi_2Te_2Se$ and $Bi_2Se_2Te$ are found to be stable TI material due to their narrow gap properties [1, 17]. Among them, binary mixed combination has been investigated in details with experimental confirmations on $Bi_2Te_3$ and $Bi_2Se_3$ [1, 8, 9]. Recently, the behaviour of ternary $Bi_2Te_2Se$ as strong TI material has been confirmed experimentally [3, 12]. To the great surprise, one of the major properties that have not been exploited in details in case of both binary and ternary combinations is the coupling between the electronic and the atomic degrees of freedom (electron-phonon coupling). This property has proved to be a key role in understanding numerous physical phenomena of almost all the insulator materials, as such one of the ternary compound that recently predicted as a good TI material is $Bi_2Te_2S$, this material has been studied within the DFT calculation, but with almost no experimental study as TI. $Bi_2Te_2S$ is observed by replacing the Te central layer of binary $Bi_2Te_3$ with S, which lead to the smaller lattice parameters (see Table 1). It is found that the binding of the site in the central layer to two Bi sites increases hybridization and decreases spin-orbit coupling, something that can be explain clearly in quasiparticle band gap.

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In this paper, the quasiparticle band structure of $Bi_2Te_2S$ has been investigated within the GW approximation as implemented in the Yambo code, to understand more clearly the effect of electron-phonon interaction on the DFT-band gap, the absorption spectrum is evaluated within the Random Phase approximation.

## 2. Computational Method

DFT calculations was performed using Quantum ESPRESSO package [4], with generalize gradient approximation (GGA) parameterized by Perdew– Burkew–Enzerhof (PBE) as exchange correlation functional. The valences as well as the core-valence electrons are clearly described by means of Troullier-Martins scalar relativistic pseudopotentials. The two sets of pseudopotentials obtained from [5] were used in the calculations, one set has five valance electrons, i.e., $5s^2 5p^3$ for Te atom and $6s^2 6p^3$ for Bi are used during DFT calculation, while the other set has additional semi core $4d^{10}$ and $5d^{10}$ electrons for Te and Bi respectively is used during the GW calculation. In each case $3s^2 3p^4$ electrons for S atom are considered. All the calculation was performed on the primitive rhombohedral unit cell with five atoms in the cell (Fig 1a), and a Monk horst-Pack grid of $7 \times 7 \times 1$ k-point mesh was used during the relaxation calculation. The kinetic energy cut-off is 40 Ryd. For the self-consistent calculation $10 \times 10 \times 1$ and $4 \times 4 \times 2$ *k*-point meshes are used for DFT and GW calculations, respectively.

We compute the quasiparticle correction to the DFT band gap of the bulk $Bi_2Te_2S$ within many- body perturbation theory using GW method as implemented in the YAMBO code [6]. The self-consistent GW energy is evaluated within the first-short $G_0W_0$ approximation as $\Sigma = iG_0W_0$, where $G_0$ represented the electron Green's function defined by the Kohn-Sham eigenstates $\Psi_{mk}(r)$ and eigenvalues $\Phi_{mk}$ corresponding to the band index $m$ and the wave vector $k$, and $W_0$ represents the screened Coulomb interaction calculated in the random phase approximation (RPA). The quasiparticle energies $E_{mk}$ are obtained as

$$E_{mk} = \Phi_{mk} + \chi_{mk} h\Psi_{mk} \left| \Sigma\left(p\nu\Phi_{mk}\right) - V_{XC} \right| \Psi_{mk} i, \quad (1)$$

where $E_{mk}$ is the quasiparticle energy, $\chi_{mk}$ the associated quasiparticle renormalization, and $V_{XC}$ is the exchange and correlation potential.

*Table 1. Lattice parameters for both rhombohedral and hexagonal unit cells used for this calculation obtained from reference [10-15], the hexagonal structure is stacked along the c-axis.*

| $Bi_2T_2S$ | Rhombohedral unit cell | Hexagonal unit cell |
|---|---|---|
| Space group | $D^5_{3d}(R\bar{3}m)$ | $D^5_{3d}(R\bar{3}m)$ |
| Lattice parameters (Å) | $a = b = c = 10.255$ | $a = b = 4.346,$ $c = 32.269$ |
| Lattice angles(°) | $\alpha = \beta = \gamma = 22.695$ | $\alpha = \beta = 90, \gamma = 120$ |

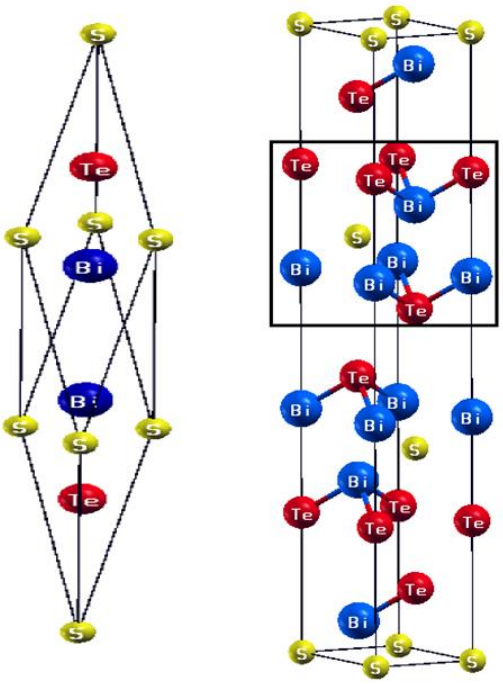


*Fig. 1(a) rhombohedral unit cell with five atoms in the cell. (b) The equivalent bulk hexagonal conventional lattice cell with fifteen atoms stacking with quintuple layers indicated by the black box.*

## 3. Results

### A. DFT-GGA Band gap structure

Fig 2 shows our calculated band gap structure of the bulk $Bi_2Te_2S$ within the DFT-GGA methods. For simplicity we have only shown how the top valence band is mostly observed from *p* orbitals of Bi as well as *p* orbitals of both Te and S character (Fig. 2a), while the bottom of the conduction band comprises of the *s* orbital of Bi, Te and S (Fig. 2b), this result is in consistently with previous calculations more especially for the binary mixed chalcogenide compound like $Bi_2Te_3$.

We have also observed the presence of several extrema in proximity of the fundamental gap, making the direct and indirect transitions almost degenerate. The separation of energy between direct and indirect gaps is found to be around 0.05 eV. This energy differences show the presence of Dirac point (DP) at the Gamma point as well as at $E_F$ indicating that $Bi_2Te_2S$ can also be a good TI material, it is also suggests that the direct transition will probably lead more absorption than the indirect one as observed in [19] for similar compound. Similar result has been observed using Angle Resolved Photo-Emission Spectroscopy (ARPES) on $Bi_2Te_3$. This can also confirm the presence of weak absorption just below the threshold for the direct absorption in bulk of this material. Therefore for most of the practical application, in particular for electro-optic devices applications, ternary $Bi_2Te_2S$ can also be considered as good material.

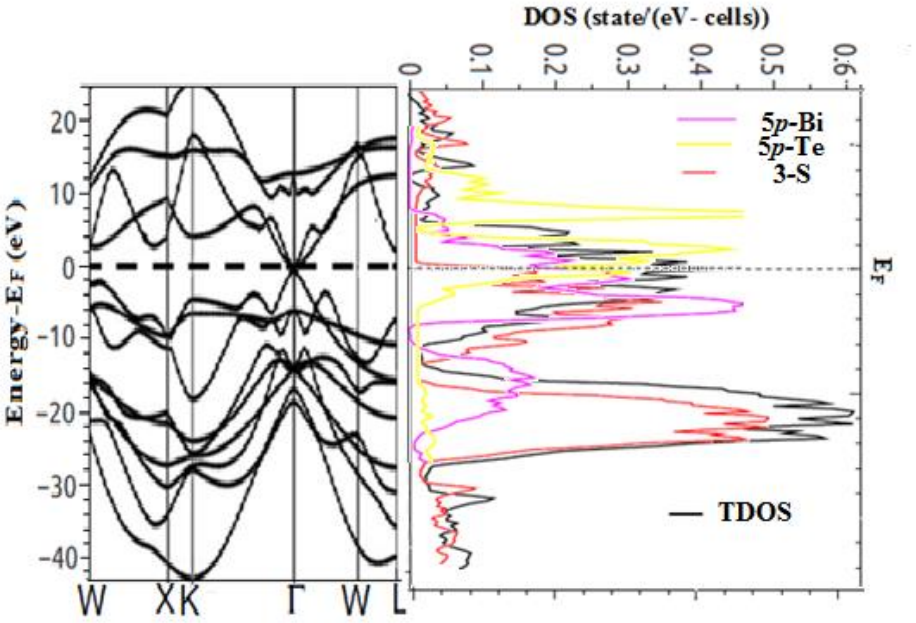


*Fig. 2(a): Band structures of ternary $Bi_2Te_2S$ calculated using DFT-GGA, using the experimental lattice parameters, without spin-orbit interaction, as well as corresponding total density of states (DOS, black lines). The valence band contributions to the DOS from the p states of Bi, Te and S are indicated by the pink, yellow and red colors respectively.*

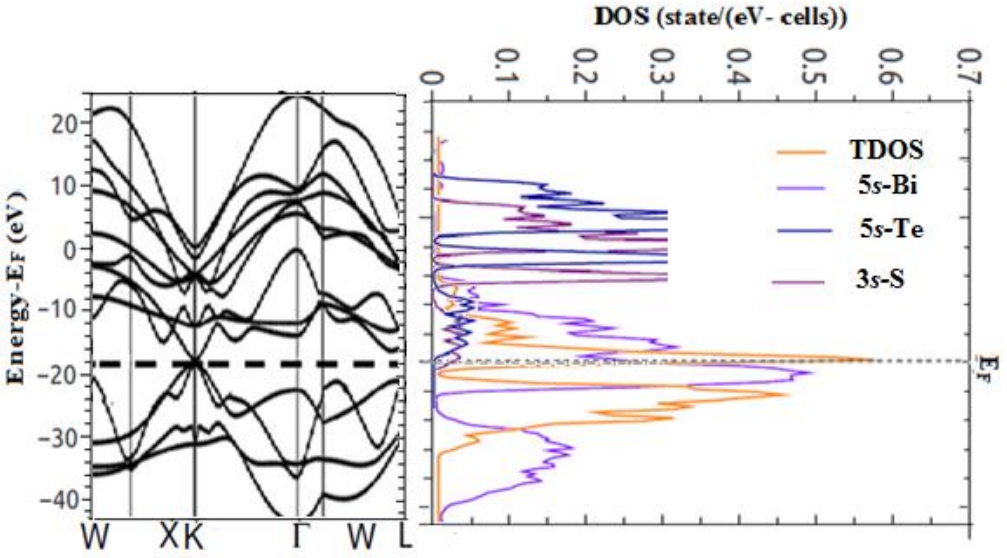


*Fig. 2(b): Band structures of ternary $Bi_2Te_2S$ calculated using DFT-GGA, using the experimental lattice parameters, without spin-orbit interaction, as well as corresponding total density of states (DOS, orange lines). The conduction band contributions to the DOS from the s states of Bi, Te and S are indicated by the pink, blue and light pink colors respectively.*

**B. GW quasiparticle band structure**

We have obtained the quasiparticle band structure of ternary $Bi_2Te_2S$ taking the GW corrections into account to the GGA band structure. Firstly, it is significant to emphasize that in our GW calculations we have considered the two extrema at the DFT-GGA band structure (Fig. 3).

These extrema clearly show the ability of forming a direct band gap that can increases the fundamental gap of $Bi_2Te_2S$ in the case of GGA+GW calculation. Our calculation has carried out on many-body corrections within the one-shot GW approach. We have observed that the one-shot GW results at fixed atomic positions of bulk ternary $Bi_2Te_2S$ depend on the DFT-GGA point of reference band structure. We have noticed a strongly dependency as reported in the case of $Bi_2Se_3$. We have also found that the enlargement of the fundamental band gap due to the one-shot GW corrections has brought its value close agreement with experiment in the case of similar ternary compound $Bi_2Te_2Se$.

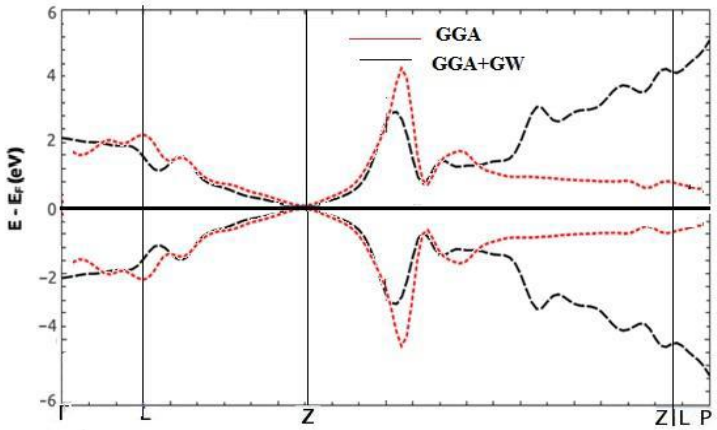


*Fig. 3: The red curve indicate valence and conduction bands as found within the GGA calculation and the black curve indicate the valance and conduction bands in GGA+GW calculation for the relaxed parameters of ternary $Bi_2Te_2S$, the GGA+GW results obtained with different numbers of unoccupied bands and k-point grids.*

We have also evaluated the electron-phonon induced by renormalization of the bulk band-gap of the $Bi_2Te_2S$. We investigate the electronic line-widths induced by the electron-phonon coupling and used it to obtain the finite temperature absorption spectrum of bulk $Bi_2Te_2S$ within the Bethe-Saltpeter equation (BSE) approach as implemented in YAMBO code (Fig.4). From the result, we have observed that the Fermi level lies inside the fundamental band gap which is about 120 meV above the DP, confirming similar TI property of $Bi_2Te_2Se$ as reported in [3].

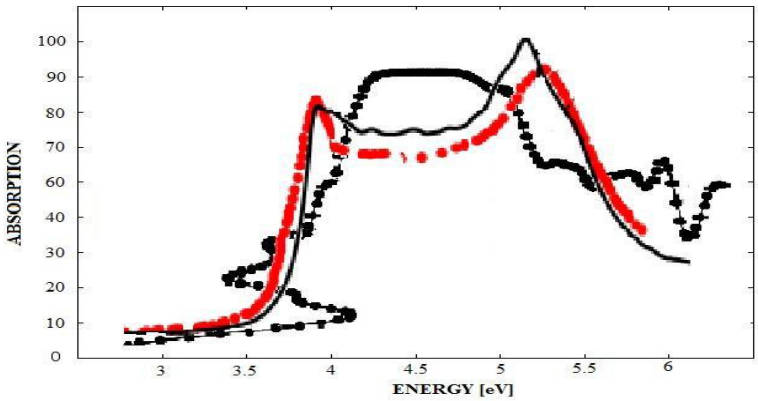


*Fig. 4. Absorption spectra of bulk ternary $Bi_2Te_2S$ topological insulator calculated from Bethe-Saltpeter equation). The red dashes curve shows the experimental spectra for bulk $Bi_2Te_2Se$, the black dash line shows the spectra of $Bi_2Te_2S$ without (RIM), while the black line indicate the spectra with (RIM).*

## 4. Concussion

In summary, we have perform a DFT-GGA as well as GW approximation studies on the bulk ternary $Bi_2Te_2S$ compound, from our DFT-GGA results, a similar properties with some binary and ternary mixed chalcogenide compound as TI material has been observed. We have also validated and then used quasiparticle band structures within the GW approximation, to understand the enlargement properties in the fundamental band gap of this material.

We want to emphasize that the effect of exciton on the optimized structural parameters used for GW calculation are found to be small in our relaxed data, making our results for the absorption spectrum to be in good agreement with experiment in case of similar ternary compound $Bi_2Te_2Se$.It will be also interesting to say that the observed spectrum shows that, the effect of electron-phonon coupling is important in order to achieve the experimental agreement for the Dirac Fermions, Furthermore, our associated absorption spectra are well compare with previous *ab-initio* results, thus establishing the exactness of this approach on ternary $Bi_2Te_2Se$.

For the future, it will be very motivating to treat this absorption spectrum temperature dependent. In this case the most important contribution is expected from the poles of the non-interacting electron-hole Green's function, properties that can be used to confirm the TI of this ternary $Bi_2Te_2S$.

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