Exploring temperature dependent electron–electron interaction of rocksalt SnS and SnSe within Matsubara-time domain

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Abstract
Both experimental and theoretical studies show non-trivial topological behaviour in native rocksalt phase for SnS and SnSe and categorize these materials in topological crystalline insulators. Here, the detailed electronic structures studies of SnS and SnSe in the rocksalt phase are carried out using many-body $GW$ based theory and density functional theory both for ground states and temperature dependent excited states. The estimated values of fundamental direct bandgaps around $L$-point using $G_0W_0$ (mBJ) are $\sim0.27\,(\sim0.13)$ eV and $\sim0.37\,(\sim0.17)$ eV for SnS and SnSe, respectively. The strength of hybridization between Sn $5p$ and S $3p$ (Se $4p$) orbitals for SnS (SnSe) shows strong $k$-dependence. The behaviour of $\bar{W}(\omega)$, which is the averaged value of diagonal matrix elements of fully screened Coulomb interaction, suggests to use full-$GW$ method for exploring the excited states because the correlation effects within these two materials are relatively weak. The temperature dependent electronic structure calculations for SnS and SnSe provide linearly decreasing behaviour of bandgaps with rise in temperatures. The existence of collective excitation of quasiparticles in form of plasmon is predicted for these compounds, where the estimated values of plasmon frequency are $\sim9.5\,eV$ and $\sim9.3\,eV$ for SnS and SnSe, respectively. The imaginary part of self-energy and mass renormalization factor ($Z_k(\omega)$) due to electron–electron interaction (EEI) are also calculated along $W$–$L$–$\Gamma$ direction for both the materials, where the estimated ranges of $Z_k(\omega)$ are 0.70 to 0.79 and 0.71 to 0.78 for SnS and SnSe, respectively, along this $k$-direction. The present comparative study reveals that the behaviour of temperature dependent EEI for SnS and SnSe are the almost same and EEI is important for high temperature transport properties.

Keywords: first-principles calculations, density functional theory, $GW$, electron–electron interaction

(Some figures may appear in colour only in the online journal)

1. Introduction
From past few decades, topological materials (TM) opened a new era of research in the field of condensed matter physics due to the presence of exotic phenomena and multiple application purposes they offer [1, 2]. Topological crystalline insulator (TCI) is one of the important members in the class of TM after mentioned by L Fu [3]. TCI shows finite bandgap in the corresponding bulk band structure, whereas the conducting surface states of this material are usually protected by mirror
In this work, we explored both, ground and excited electronic structures of SnS and SnSe in rocksalt phase using many-body theory based on $G_0W_0$ and full-GW methods. The comparative study between DFT and $G_0W_0$ methods for ground state calculations is also carried out. The importance of SOC for estimating the values of bandgaps are seen for both the materials. The calculated value of bandgap for SnS using $G_0W_0$ (mBJ) is $\sim 0.13$ ($\sim 0.27$) eV, where the fundamental bandgap is not seen from PBEsol functional. In case of SnSe, the computed values of bandgaps is found to be $\sim 0.22$ eV, $\sim 0.17$ eV and $\sim 0.37$ eV for PBEsol, mBJ and $G_0W_0$, respectively. The strength of hybridization between Sn 5$p$ and S 3$p$ (Se 4$p$) orbitals for SnS (SnSe) has shown strong dependency on the $k$-point. The correlation strength of SnS and SnSe is estimated from the $\omega$ dependent $\overline{W}$, which defines the averaged value of diagonal elements of fully screened Coulomb interaction. The different forms of Coulomb interactions are calculated using random phase approximation (RPA) for both the materials. The temperature dependent bandgap along with spectral functions due to EEI are explained. The presence of plasmon excitations are predicted from the full-GW calculations for SnS and SnSe. The detailed studies of many-body EEI for both TCIs are carried out by explaining the temperature and $k$-point (i.e. along $W\rightarrow L\rightarrow \Gamma$ direction) dependent imaginary part of the self-energy ($\text{Im}E(k, \omega)$). The similar amount of EEI is evident from the present study for both materials. Momentum-resolved spectral function is also discussed for SnS and SnSe.

2. Theoretical methods

2.1. Description of different Coulomb interactions

RPA is used for estimating different Coulomb interactions. In order to calculate these quantities, we need to form the local atomic-like orbitals from the Kohn–Sham orbitals. Maximally localized Wannier function (MLWF) is chosen to build the local orbitals for particular bands which are found in certain energy region. The orbitals of dominating characters around the $E_F$ are usually taken for obtaining the MLWF. In general, the fully screened Coulomb interactions ($W$) and bare Coulomb interaction ($\nu$) are given below [23],

$$W^{\nu,R_1R_2R_3R_4}_{L_1L_2L_3L_4}(\omega) \equiv \langle \phi_{R_1L_1} \phi_{R_2L_2} | W(\omega) | \phi_{R_3L_3} \phi_{R_4L_4} \rangle$$

(1)

$$\nu^{\nu,R_1R_2R_3R_4}_{L_1L_2L_3L_4}(\omega) \equiv \langle \phi_{R_1L_1} \phi_{R_2L_2} | \nu(\omega) | \phi_{R_3L_3} \phi_{R_4L_4} \rangle$$

(2)

where $L = (m, \alpha, \sigma)$, $m$ is an orbital quantum number, $\alpha$ represents the atomic index of a given unit cell centered at $\mathbf{R}$ and spin degree of freedom is denoted by $\sigma$. $\phi_{R\mu}$ represents the MLWF of corresponding Kohn–Sham orbital. The relation between $W(\omega)$ and $\nu$ is found to be $W = [1 - \nu P]^{-1} \nu$, where $P$ represents the total polarization within RPA and $\nu$ is the Coulomb interaction between two bare charges [24]. It is noted that the $W$ is a function of $\omega$, since $P$ depends on $\omega$. The on-site Coulomb interactions are obtained when we fixed $R_1 = R_2 = R_3 = R_4$. Therefore in case of on-site intra-atomic, the notation $L$ only depends on the value of $m$ and the on-site bare exchange Coulomb interaction ($J_{\text{bare}}$) is denoted by,
2.2. Discussion on Matsubara-time domain GW method

Now, it is well understood that preforming temperature dependent GW calculation in Matsubara-time domain is more easy and computationally convenient to implement than the conventional statistical mechanics approach. On other hand, the effect of temperature on the electronic structure properties of any material is also possible to study using the former formalism [25]. This Matsubara-time Green’s function method is normally utilized within DFT + DMFT method form few decades [26–28]. In continuation with this, the information about the many-body effect due to EEI can be obtained from the calculated values of electronic self-energy ($\Sigma$) using this methodology. The $\Sigma(\tau)$ is obtained from this calculation, where $\tau$ denotes the Matsubara-time. Next, it is transferred into $\Sigma(i\omega_n)$ using Fourier transformation, where $\omega_n = (2n + 1)\pi T$ for fermions. Here, $n$ is integer, $\beta = \frac{1}{k_B T}$ and $T$ represents temperature. But, in order to obtain the spectral function in real $\omega$, the analytic continuation is generally performed to transform $\Sigma(i\omega_n)$ to $\Sigma(\omega + i\eta)$, where $\eta$ denotes a positive infinitesimal number.

3. Computational details

Here, WIEN2k [29] code is chosen to carry out the spin-unpolarized electronic structure calculation for SnS and SnSe compounds. This code is based on full-potential linearized-augmented plane-wave. The space group of $Fm-3m$ is used in present study for both materials. The schematic diagram of the crystal structure is shown in figure 1. The PBEsol XC functional [30] and mBJ exchange potential with GGA correlation [31] are utilized for DFT calculation. The estimated values of optimized lattice parameters, which are computed using PBEsol functional for SnS and SnSe, are 5.753 Å and 5.955 Å, respectively. It is noted that these values are showing good agreement with the previously reported theoretical and experimental data of these two materials [6, 9, 10, 32]. The self-consistent DFT calculations are performed on $10 \times 10 \times 10$ k-mesh size. The Wyckoff positions for Sn and S (Se) are fixed at (0.0, 0.0, 0.0) and (0.5, 0.5, 0.5), respectively, where the muffin-tin radius are set to be 2.5 Bohr for Sn and Se along with 2.43 Bohr for S. The convergence criteria is kept to $10^{-8}$ Ry cell$^{-1}$ for calculating the total energy. In order to get the better convergence, $K_{max}$ is considered as 8.5 in whole calculations. GAP2 code based on Wannier basis function is utilized for computing different Coulomb interaction with the help of RPA [33, 34]. GAP2 is also used for carrying out $G_0W_0$ calculations on both the compounds, where it generally corrects the Kohn–Sham states by perturbative manner for obtaining the quasiparticle states of interacting electrons. In addition to this, Elk [35] code is chosen for performing the full-GW calculation, where the Matsubara-time domain is implemented to perform the calculation on imaginary axis. In order to get the spectral function on real axis, Padé approximation [36] is employed to transform the data from imaginary to real axis. The procedure of analytic continuation based on Padé approximation is chosen due to its’ lower computational cost and simple implementation within the full-GW methodology. $4 \times 4 \times 4$ q-mesh size is taken together with $8 \times 8 \times 8$ k-mesh for full-GW calculation. It is known that the $G_0W_0$ calculation depends on using the starting XC functional at DFT level. Therefore, to get rid from this issue, we keep PBEsol XC functional at DFT level for all types of GW based calculations [30]. Here, the ground state electronic structures of SnS and SnSe are studied both in presence and absence of SOC.

4. Results and discussion

4.1. Ground state electronic structure and correlation effect

To understand the basic electronic structure of any non-trivial TCI, the detailed discussion on ground state properties with proper estimation of fundamental bandgap is known to be important. Therefore, the systematic theoretical studies on both SnS and SnSe are carried out in present work with different functionals at DFT level together with different methods (i.e. DFT, $G_0W_0$). Here, it is noted that zero energy in the dispersion plot represents the Fermi energy ($E_F$). The $E_F$ is fixed at middle of the bandgap when a clear bandgap is seen from any calculated band structure. Figures 2(a) and (b) show the dispersion curves for SnS and SnSe along the high symmetric $k$-direction, which are calculated using PBEsol, mBJ and $G_0W_0$ without considering SOC. In case of conduction band (CB), the bands of PBEsol and $G_0W_0$ are showing almost similar behaviour along the observed $k$-direction for both samples. For SnS in CB region, the shifting of mBJ bands toward the lower energy with respect to PBEsol bands has been clearly seen, where the maximum shifting is found around the $L$-point. But for SnSe, the mBJ bands nicely followed the PBEsol bands for CB region. Similarly for valence band (VB) region, the bands obtained from $G_0W_0$ move towards to lower energy than PBEsol for both materials, whereas the further shifting of the bands to lower energy is seen for mBJ bands with respect to $G_0W_0$. The energy difference between mBJ ($G_0W_0$) bands and
Figure 2. Electronic band structures of SnS and SnSe (a) and (b) without and (c) and (d) with including SOC using PBEsol (black solid line), mBJ (red dashed line) and $G_0W_0$ (blue dotted dash line).

Figure 3. Projected band structure along $W$–$L$–$\Gamma$ direction using PBEsol functional of (a) SnS and (b) SnSe.

PBEsol bands is estimated to be maximum for $W$–$L$–$\Gamma$ direction, where the calculated differences are 0.6–0.9 (0.4–0.5) eV and 0.4–0.53 (0.2–0.22) eV for SnS and SnSe, respectively. Here, it is noted that the energy shifting is more prominent for SnS than SnSe, which may be due to the increase of lattice parameters from SnS to SnSe. In addition to this for SnS, $G_0W_0$ shows direct bandgap of amount $\sim 0.23$ eV near the $L$-point, whereas PBEsol and mBJ provide metallic behaviour. In case of SnSe, the calculated values of bandgap using PBEsol and $G_0W_0$ are found to be $\sim 0.15$ eV and $\sim 0.32$ eV, respectively and the band structure of mBJ shows metallic nature. Moreover, due to the presence of heavier element Sn in both the compounds, SOC is expected to play important role for predicting the better electronic ground state. Therefore, we have performed the further ground state calculation including SOC. Here, the SOC correction at DFT level is considered in $G_0W_0$ method by perturbative manner. The calculated dispersion curves using PBEsol, mBJ and $G_0W_0$ including SOC are plotted in figures 2(c) and (d) for SnS and SnSe, respectively. The band features are almost similar with the
Table 1. Averaged values of diagonal and off-diagonal matrix elements of bare Coulomb interaction ($\tau$) for Sn ($S$) $5p$ ($3p$) orbitals & matrix elements of bare ($\tau_{Sn-S}$) and screened ($\overline{W}_{Sn-S}$) inter-atomic Coulomb interactions. The on-site bare exchange interaction $J_{bare}$ and the averaged values of diagonal matrix elements of on-site fully screened Coulomb interaction ($\overline{W}$) at $\omega = 0.0$ eV for Sn $5p$ and S $3p$ orbitals. All numbers are in unit of eV.

|          | Bare Coulomb interaction ($\tau$) | Inter-atomic Coulomb interactions | On-site $J_{bare}$ | $\overline{W}$ at $\omega = 0.0$ eV |
|----------|----------------------------------|----------------------------------|-------------------|----------------------------------|
|          | Sn $5p$                          | S $3p$                           |                   | Sn $5p$                          | S $3p$ |
| Diagonal | 4.37                             | 3.11                             | 3.95              | 3.04                             |       |
| Off-diagonal | 3.11                           |                                  |                   |                                  |       |

Table 2. Averaged values of diagonal and off-diagonal matrix elements of bare Coulomb interaction ($\tau$) for Sn ($Se$) $5p$ ($4p$) orbitals & matrix elements of bare ($\tau_{Sn-Se}$) and screened ($\overline{W}_{Sn-Se}$) inter-atomic Coulomb interactions. The on-site bare exchange interaction ($J_{bare}$) and the averaged values of diagonal matrix elements of on-site fully screened Coulomb interaction ($\overline{W}$) at $\omega = 0.0$ eV for Sn $5p$ and Se $4p$ orbitals. All numbers are in unit of eV.

|          | Bare Coulomb interaction ($\tau$) | Inter-atomic Coulomb interactions | On-site $J_{bare}$ | $\overline{W}$ at $\omega = 0.0$ eV |
|----------|----------------------------------|----------------------------------|-------------------|----------------------------------|
|          | Sn $5p$                          | Se $4p$                          |                   | Sn $5p$                          | Se $4p$ |
| Diagonal | 8.64                             | 7.89                             | 11.01             | 9.84                             |       |
| Off-diagonal | 7.89                           |                                  |                   |                                  |       |

previous discussion (i.e. without including SOC) except some band degeneracy lifting along the high-symmetric $k$-direction due to SOC. This behaviour is usually observed after including SOC. The PBEsol does not show any bandgap after including SOC for SnS, whereas mBJ ($G_0W_0$) creates bandgap of $\sim0.13$ ($\sim0.27$) eV around $L$-point. Similarly for SnSe after inclusion of SOC, it is seen that PBEsol, mBJ and $G_0W_0$ provide the bandgap of $\sim0.22$ eV, $\sim0.17$ eV and $\sim0.37$ eV, respectively. The fundamental bandgap of many-body electronic system is usually defined as the difference in calculated energy between adding an electron and removing an electron from the sample system [24]. Therefore, the bandgap obtained from PBEsol is not expected to provide correct value as compared to experimental data because the orbital dependency for computing the band energy at CB minima and VB maxima is missing in Kohn–Sham implementation along with the
problem of derivative discontinuity. But at least, it is desired that PBEsol will show semiconducting behaviour for both SnS and SnSe due to the presence of $p$-like electrons at the lower energy region around $E_F$. However, the dispersion plot of PBEsol of SnS shows metallic behaviour, but the semiconducting nature is seen for SnSe. Since $3p$ orbitals are more localized than $4p$ orbitals, therefore the correlation between the electrons of $S$ $3p$ orbitals might be improperly estimated for SnS compound. Thus, it leads to the metallic behaviour of SnS even after including SOC. Now, in case of mBJ, it shows the semiconducting behaviour for both the materials by correcting the kinetic energy density of these compounds. But, the mBJ method also comes under the Kohn–Sham formalism, where the electronic screening is not considered within the methodology. This screening is important for finding the bandgap of any material. Hence, it is also not expected from mBJ to give proper fundamental bandgap as compared to experimental bandgap [14, 39]. At this point, it is important to note that the hybridization strength of these materials show dependency on the choice of $k$-point within first Brillouin zone. The $5p$ and $3p$ ($4p$) orbitals of Sn and S (Se) are mainly responsible for making the energy bands within the energy window of $−8.5$ eV ($−6.0$ eV) to $8.0$ eV ($7.0$ eV) for SnS (SnSe) material. The projected bandstructures of SnS and SnSe materials are shown in figures 3(a) and (b), respectively. The presence of Sn $5p$ and $S$ $3p$ ($Se$ $4p$) orbitals of SnS (SnSe) is clearly seen around the $E_F$ from the figures 3(a) and (b). These energy bands around the $E_F$ are usually expected to participate for showing the transport and topological behaviours. Presence of electronic correlation always brings new exotic behaviour along with these mentioned properties. Therefore, investigation of different Coulomb interactions by considering these orbitals will provide insightful information about the correlation effects of both compounds.

In present work for computing different Coulomb interactions, MLWF are formed using Sn $5p$ and S $3p$ ($Se$ $4p$) orbitals and $\sim 0.98$ eV for SnS and SnSe, respectively. The fundamental bandgap as computed using $G_0W_0$ method for SnS is showing smaller value than SnSe around the $L$-point, but the direct bandgap at $L$-point for former compound provides higher value than the other one. Thus, it is evident for $L$-point (the $k$-point where the fundamental bandgap is observed) that the hybridization strength between Sn $5p$ and $S$ $3p$ orbitals for SnS is stronger (weaker) than Sn $5p$ and Se $4p$ orbitals for SnSe. At this point, it is important to note that the hybridization strength of these materials show dependency on the choice of $k$-point within first Brillouin zone. The $5p$ and $3p$ ($4p$) orbitals of Sn and S (Se) are mainly responsible for making the energy bands within the energy window of $−8.5$ eV ($−6.0$ eV) to $8.0$ eV ($7.0$ eV) for SnS (SnSe) material. The projected bandstructures of SnS and SnSe materials are shown in figures 3(a) and (b), respectively. The presence of Sn $5p$ and S $3p$ ($Se$ $4p$) orbitals of SnS (SnSe) is clearly seen around the $E_F$ from the figures 3(a) and (b). These energy bands around the $E_F$ are usually expected to participate for showing the transport and topological behaviours. Presence of electronic correlation always brings new exotic behaviour along with these mentioned properties. Therefore, investigation of different Coulomb interactions by considering these orbitals will provide insightful information about the correlation effects of both compounds.

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**Figure 4.** Coulomb interactions as a function of $\omega$ for SnS and SnSe.

**Figure 5.** Spectral functions of $L$-point from full-$GW$ at temperature 400 K (black solid line), 1000 K (red dashed line) and 1500 K (blue dotted dash line) for (a) SnS and (b) SnSe.
Figure 6. Calculated (black dot) and extrapolated (red sold line) temperature dependent bandgap at L-point for (a) SnS and (b) SnSe.

Figure 7. Calculated (symbol) and extrapolated (line with symbol) $|\text{Im}\Sigma(\omega)|$ as function of temperatures for 1st and 2nd peak of (a) SnS and (b) SnSe.

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strongly (weakly) correlated materials with changing \( \omega \) values [40–43]. In present case, weakly \( \omega \) dependent behaviour is seen from \( \mathcal{W}(\omega) \) curves for all studied orbitals of both materials. Therefore, the existence of weak correlation within these two samples are evident from the above discussion. In light of this to study the many-body effects of SnS and SnSe materials, the full-GW method is expected to provide the appropriate descriptions of excited electronic states.

4.2. Finite temperature electronic structure

It is known that when one calculates the electronic structure of any material using the many-body perturbation theory then it provides more closer predictive result to experimental observation as compared to single particle theory. In this scenario, the temperature dependent Green function based full-GW methodology is one of the advanced method to tackle the many-body problem. Here, we have performed full-GW calculation at different temperatures for SnS and SnSe. The spectral function \( (A_{jk}(\omega)) \) is defined as [24],

\[
A_{jk}(\omega) = \frac{1}{\pi} \text{Im}[G_{jk}(\omega)]
\]  

(4)

where, \( j \) denotes band index at point \( k \) and \( G_{jk}(\omega) \) is the Green function of many-body system. The calculated \( A_{jk}(\omega) \) at \( L \)-point for temperatures 400 K, 1000 K and 1500 K are shown in figures 5(a) and (b) for SnS and SnSe, respectively. The band inversion, which is usually responsible for the presence of topological behaviour within the sample, are seen around the high-symmetric \( L \)-point for both compounds. Therefore in present case, the \( L \)-point is chosen for carrying out the detailed study of \( A_{jk}(\omega) \) at different temperatures. The peaks’ height (broadening) of \( A_{jk}(\omega) \) is decreasing (increasing) with rise in temperature from 400–1500 K. In this case, the \( E_F \) is set at the middle of 1st peak’s and 2nd peak’s center, which are marked in figures. The figures 5(a) and (b) illustrate that if one moves from SnS to SnSe then the peaks’ center of VB (CB) shifted from lower (higher) to higher (lower) energy with respect to \( E_F \). It is known that the atomic energy of S \( 3p \) states is lower than the Se \( 4p \) states. Therefore in case of VB, the resultant quasiparticle energy due to overlap of orbitals between Sn \( 5p \) and S \( 3p \) is lower than the overlap between Sn \( 5p \) and Se \( 4p \) states. The similar argument is also applicable for CB, where opposite behaviour is seen. Moreover, the hybridization energy of Sn \( 5p–3p \), \( 3p \) seems to be stronger than Sn \( 5p–4p \) because the value of bandgap decreases from SnS to SnSe at this \( k \)-point. Here, the energy difference between the center of 1st and 2nd peaks is considered as calculated bandgap at particular temperature. The calculated bandgaps at different temperatures are plotted in figures 6(a) and (b) of SnS and SnSe, where monotonically decreasing behaviour with increasing temperature is seen for both compounds due to EEI. The estimated values of bandgaps for SnS (SnSe) are found to be \( \sim 0.976 (\sim 0.873) \) eV, \( \sim 0.973 (\sim 0.87) \) eV, \( \sim 0.971 (\sim 0.868) \) eV and \( \sim 0.967 (\sim 0.859) \) eV at 400 K, 700 K, 1000 K and 1500 K, respectively. The bandgaps of SnS for the studied temperature range are showing higher values than SnSe. This behaviour suggests that the bonding and anti-bonding of Sn \( 5p–3p \) orbitals are stronger than Sn \( 5p–4p \) orbitals at \( L \)-point. Moreover, performing the full-GW calculation at lower temperature is not possible due to huge computational cost of this methodology and the limitation of present computational resources. Therefore, to compare the calculated bandgap of full-GW method with the estimated value of ground state electronic structure calculations, we extrapolated the values of bandgap till 0 K with linear fitting of these calculated data. The extrapolated value of direct bandgap at 0 K is found to be
The computed values of this quantity are \( \sim 0.79 \) (\( \sim 0.879 \)) eV for SnS (SnSe). The difference between these values with the calculated values of \( G_0W_0 \) at L-point are estimated to be \( \sim 0.1 \) eV for both materials. \( G_0W_0 \) provides larger bandgap value than extrapolated values at L-point for 0 K. The estimated value of bandgap at 300 K from the extrapolated data is \( \sim 0.977 \) (\( \sim 0.875 \)) eV for SnS (SnSe). All full-GW calculations are performed without including SOC because full-GW calculation along with SOC is highly expensive for lower temperature. Moreover, the insight of many-body effect can be explored by studying the self-energy (\( \Sigma(k, \omega) \)) of these two compounds.

The real part of \( \Sigma(k, \omega) \) (\( \text{Re}\Sigma(k, \omega) \)) provides the information of coherent weight of the spectrum, where the relation between \( \text{Re}\Sigma(k, \omega) \) and the mass renormalization factor (\( Z_k(\omega) \)) is given by,

\[
Z_k(\omega) = \left[ 1 - \frac{\partial \text{Re}\Sigma(k, \omega)}{\partial (\omega)} \right]^{-1}
\]

(5)

The presence of incoherent weight of the spectrum can be quantified by \( (1 - Z_k(\omega)) \). In present scenario, the values of \( Z_k(\omega) \) are found to be \( \sim 0.78 \) (\( \sim 0.75 \)), \( \sim 0.775 \) (\( \sim 0.77 \)) and \( \sim 0.79 \) (\( \sim 0.77 \)) for 1st (2nd) peak of SnS at 400 K, 700 K and 1500 K, respectively. Similarly in case of SnSe, the estimated values of this quantity are \( \sim 0.77 \) (\( \sim 0.758 \)), \( \sim 0.794 \) (\( \sim 0.76 \)) and \( \sim 0.79 \) (\( \sim 0.78 \)) for 1st (2nd) peak at 400 K, 700 K and 1500 K, respectively. The computed values of \( Z_k(\omega) \) for both these compounds are showing almost same amount for all studied temperatures. The calculated values of \( Z_k(\omega) \) at L-point for 1st (2nd) peak, which are calculated using the ground state electronic structure calculation as performed by \( G_0W_0 \) method, are \( \sim 0.72 \) (\( \sim 0.73 \)) and \( \sim 0.72 \) (\( \sim 0.725 \)) for SnS and SnSe, respectively. Therefore, it is seen that the estimated incoherent weights of SnS (SnSe) are found to be 0.218 (0.215) and 0.236 (0.234) for 1st and 2nd peak, respectively. The presence of incoherent weight is the evidence of many-body interactions, which typically generates satellite peaks in corresponding spectrum. In order to predict the energy positions of the satellite peaks for SnS, two peaks are plotted in the inset of figure 5(a) along with DFT peaks (orange straight line). The peak center at \( \sim 8.8 \) (\( \sim 13.4 \)) eV is marked as \( S_1 \) (\( S_2 \)) in the inset of figure 5(a). The broadening of \( S_1 \) and \( S_2 \) peaks is showing much higher values than the 1st and 2nd peaks of this compound, where it is estimated that the \( S_1 \) (\( S_2 \)) peaks is \( \sim 8.8 \) (\( \sim 9.7 \)) times more broadened than 1st peak. These behaviour suggest the existence of incoherent peaks around these energy positions along with the corresponding coherent peaks. The energy difference between \( S_1 \) (\( S_2 \)) and the middle of \( 0.7 \) (\( -3.4 \)) eV to 1.5 (\( -2.7 \)) eV is measured to be \( \sim 9.3 \) (\( \sim 9.7 \)) eV. Therefore, it is predicted that \( S_1 \) and \( S_2 \) are formed due to the plasmon excitations within the sample, where the plasmon frequency is estimated to be \( \sim 9.5 \) eV for SnS. In analogues with the previous description, two peaks at \( \sim 8.7 \) eV and \( \sim 13.7 \) eV, which are corresponding to SnSe, are shown in the inset of figure 5(b) together with the DFT peaks (orange straight line). In the inset of figure 5(b), the peak at \( \sim 13.3 \) eV is pointed by \( S_1' \), whereas a kink at \( \sim 8.84 \) eV is marked by \( S_2' \). The \( S_2' \) is estimated to be \( \sim 8.6 \) times more broadened than

Figure 9. Calculated values of \( |\text{Im}\Sigma(\omega)| \) along \( W-L-\Gamma \) direction at 700 K for the top most VB of (a) SnS and (b) SnSe.

Figure 10. Computed values of \( Z_k(\omega) \) along \( W-L-\Gamma \) direction at 700 K for the top most VB of (a) SnS and (b) SnSe.
the corresponding 1st peak of SnSe. Therefore, the presence of kink at $\sim -8.84$ eV and the broadened peak of $S_3$ represent the coexistence of coherent and incoherent peaks around these mentioned energy. With the similar argument of the above discussion, the energy difference is found to be $\sim 8.75$ ($\sim 9.9$) eV from $S'_1$ ($S'_2$) to the middle of energy window $\sim -0.8$ ($\sim -3.0$) eV to $\sim -1.5$ ($\sim -2.17$) eV. The energy difference is almost same for both the cases, where the presence of satellite peaks are predicted. Therefore, the averaged value of $\sim 9.3$ eV is estimated as the plasmon frequency for SnSe. It is evident from this study that the value of plasmon frequency reduces from SnS to SnSe, which may be due to the increment of atomic radii of S to Se and the decrement of localization of S 3p to Se 4p orbitals. These estimated values of plasmon frequency for both materials are in good agreement with the predicted values from corresponding $\omega$ dependent Coulomb interactions plots.

Experimental studies of different transport properties suggest to carry out detailed analysis of lifetime $\tau$ of any material for better understanding of many-body interaction within the sample. Topological non-trivial materials can be possibly distinguished from topologically trivial materials with the help of experimental evidence of transport properties. It is also noted that imaginary part of the self-energy $\text{Im}\Sigma(k, \omega)$ is inversely related with $\tau$. Therefore, the temperature dependent $\text{Im}\Sigma(\omega)$ of SnS and SnSe at L-point are plotted in figures 7(a) and (b) for further discussion on EEI. Here, the values of $\text{Im}\Sigma(\omega)$ for 1st and 2nd peaks of both materials are only focused. The monotonically increasing value of $\text{Im}\Sigma(\omega)$ with rise in temperature is observed from the figures for both materials. Therefore, the decreasing trends in $\tau$ is expected with increasing temperature for SnS and SnSe due to EEI. The calculated values of $\text{Im}\Sigma(\omega)$ are found to be $\sim 26.8$ ($\sim 26.5$) meV, $\sim 46.8$ ($\sim 47.6$) meV, $\sim 66.9$ ($\sim 68.4$) meV and $\sim 101.4$ ($\sim 101.0$) meV for 1st peak of SnS (SnSe), whereas for the 2nd peak the values are $\sim 25.6$ ($\sim 26.0$) meV, $\sim 46.1$ ($\sim 45.8$) meV, $\sim 64.1$ ($\sim 66.1$) meV and $\sim 98.8$ ($\sim 99.6$) meV at 400 K, 700 K, 1000 K and 1500 K, respectively. It is seen that the values of $\text{Im}\Sigma(\omega)$ are almost same for both SnS and SnSe for particular temperature. Therefore, it is expected to observe similar amount of $\tau$ value for SnS and SnSe due to both the peaks. Moreover, it is known that the value of $\tau$ decreases if the quasiparticle energy increases. The availability of phase space for scattering increases with rise in quasiparticle energy, which is one of the possible reason for decreasing the value of $\tau$. In present scenario, the same value of $\text{Im}\Sigma(\omega)$ at certain temperature indicates the presence of almost equal phase space for scattering due to EEI. This behaviour may be seen for these two compounds because the electronegativity of S and Se are almost same in Pauling’s scale, which are 2.58 and 2.55 for S and Se, respectively [44]. Therefore, it reflects that the number of quasiparticle states (coherent) and incoherent states are almost identical for SnS and SnSe to perform the electron–electron scattering. Moreover, it is noted that such high values of $\text{Im}\Sigma(\omega)$ of both materials for the studied temperatures range suggest the importance of EEI on the transport behaviour for high temperature. In order to visualize the presence of coherent and incoherent states of both compounds, the momentum-resolved spectral function needs to be computed.

Figures 8(a) and (b) illustrate the momentum-resolved spectral function of SnS and SnSe, respectively, at temperature 700 K. These are computed using full-GW method. However, performing the calculation on lower temperature is not possible due to huge computational cost. It is verified that the spectral properties of $A \cdot k$($\omega$) for these two compounds are not much dependent on inclusion of SOC in calculation. In these figures, the coherent (incoherent) part of the spectrum are denoted by yellowish (violetish blue) colour. In order to compare with these figures, the DFT dispersion curve of corresponding compounds are also plotted in figures 8(a) and (b), which are calculated using PBEsol XC functional. It is clearly seen for SnS and SnSe that the behaviour of coherent and incoherent part of the spectrum are showing almost similar nature for the $W-L-\Gamma$ direction, which is expected from both these figures. Furthermore, for more detailed explanation on EEI of SnS and SnSe, the estimated values of $\text{Im}\Sigma(\omega)$ are plotted in figures 9(a) and (b) for the top most VB along $W-L-\Gamma$ k-direction. The value of $\text{Im}\Sigma(\omega)$ at W-point is found to be $\sim 44.6$ ($\sim 46.0$) meV for SnS (SnSe). Afterwards along W–L direction, the value of $\text{Im}\Sigma(\omega)$ is first showing the decreasing nature and then increases till L-point for both these compounds. The maximum value of $\text{Im}\Sigma(\omega)$ is estimated to be $\sim 51.7$ ($\sim 51.8$) meV near to $\Gamma$-point for SnS (SnSe) material. These computed values are useful to compare with the experimental data of angle-resolved photoemission spectroscopy (ARPES) along this particular k-direction. To get more insight of the many-body effect through the spectral weight transfer, $Z_k(\omega)$ for the top most VB of SnS (SnSe) in direction of W–L–\Gamma is plotted in figures 10(a) and (b). Almost similar behaviour is seen from figures 10(a) and (b) for $Z_k(\omega)$ along this direction for both compounds. The computed value of $Z_k(\omega)$ is found to be in range of $0.7$ to $0.79$ (0.71 to 0.78) for SnS (SnSe) along W–L–\Gamma direction. Therefore, the range of incoherent weight for the top most VB along this mentioned k-direction is estimated to be 0.3 to 0.21 (0.29 to 0.22) for SnS (SnSe), which is usually responsible for making the satellite peaks in the corresponding spectrum.

5. Conclusions

In present study, the famous TCIIs SnS and SnSe are focused to explore the ground and excited states using advanced GW based methodology along with DFT. The electronic structure calculation using mBJ and $G_0W_0$ methods are shown semiconducting behaviour for both materials. But, the PBEsol provides metallic (semiconducting) ground state for SnS (SnSe) due to the improper (proper) estimation of correlation within S 3p (Se 4p) states. The fundamental bandgaps around L-point using $G_0W_0$ (mBJ) are found to be $\sim 0.27$ ($\sim 0.13$) eV and $\sim 0.37$ ($\sim 0.17$) eV for SnS and SnSe, respectively. The behaviour of hybridization’s strength between Sn 5p and S 3p (Se 4p) orbitals for SnS (SnSe) is seen to be k-point dependent. The nature of frequency ($\omega$) dependent Coulomb interactions of both materials reveal the overall strength of correlation effects within the sample and suggest full-GW method for
studying the excited state calculations. The electronic structure calculations of excited states for SnS and SnSe are carried out using temperature dependent full-GW method in Matsubara-time domain, which have provided the linearly decreasing behaviour of temperature dependent bandgap for both materials. The presence of plasmon excitations within the spectrum is also evident from the full-GW calculations for these two compounds. It is evident from both the temperature and k-point dependent imaginary part of self-energy and mass renormalization factor that the scattering mechanism due to EEL are found to be almost same for SnS and SnSe. All these results of many-body interactions due to EEL can be possibly verified using photoemission spectroscopic experiment.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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