Thermal conductivity and enhanced thermoelectric performance of SnTe bilayer

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ABSTRACT

Tin chalcogenides (SnS, SnSe, and SnTe) are the emerging key thermoelectric (TE) materials with low toxicity and eco-friendly nature, which could serve as an alternative to the toxic lead-based alloys. The Sn-based alloys are also found to have improved TE properties upon the reduction of their dimensionality. The structural optimization, molecular dynamics and phonon transport calculations predicted the tilted AA + s stacked two-dimensional (2D) SnTe bilayer as the energetically stable structure. The deformation potential theory was used to evaluate the carrier mobility and relaxation time, which are found to be relatively high due to the high 2D elastic modulus, low deformation potential constant and moderate effective masses. The SnTe bilayer shows a high Seebeck coefficient (> 400 μV K⁻¹), high electrical conductivity and significantly low lattice thermal conductivity (< 1.91 W m⁻¹ K⁻¹). The as high TE figure of merit (ZT) values as 3.48 (4.61) along the armchair (zigzag) direction, are predicted for the SnTe bilayer within a practically attainable carrier concentration range of the order 10¹²–10¹³ cm⁻². These ZT values are greatly enhanced as compared to the bulk as well as monolayer SnTe and other 2D compounds.

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**Introduction**

With the rapid growth of global population, the energy demand by human needs is increasing day by day. To date, most of the energy consumption is being fulfilled by the conventional energy sources based on the fossil fuels, which, in addition of being non-renewable, are responsible for the large amounts of carbon dioxide emissions and adverse global environmental changes [1, 2]. These facts have stimulated researchers and governmental authorities to search for the eco-friendly, carbon neutral and renewable energy sources. Solar, wind, hydropower, geothermal and thermoelectric (TE) energy are some of the renewable energy sources. Recently proposed technologies: self-charging perovskite solar capacitors, dye-sensitized solar cells, photo-electrochemical cells, etc. are of a particular interest for promising solar photovoltaics energy applications [2–8]. Likewise, the TE materials are solid-state semiconductors which can be used for harvesting the waste heat produced in thermal power generation and convert it into electricity or vice versa. The ideal energy conversion efficiency ($\eta$) of a TE device can be expressed as [9, 10]:

$$\eta = \frac{T_H - T_C}{T_H} \left[ \frac{1}{\sqrt{1 + ZT_{avg}}} - 1 \right],$$

where $T_H - T_C$ is the Carnot efficiency; $T_H$ and $T_C$ are the temperatures of the hot and cold ends, respectively. $ZT_{avg}$ is the average temperature TE figure of merit ($ZT$) value, which is given as:

$$ZT = \frac{S^2 \sigma T}{(\kappa_l + \kappa_c)}.$$
Here $S$, $\sigma$, $T$, $\kappa_f$, and $\kappa_l$ represent the Seebeck coefficient, electrical conductivity, temperature, lattice and electronic thermal conductivities, respectively. From Eq. (1) it is clear that, optimizing the $ZT$ value is essential to increase the value of $\eta$.

Group IV–VI compounds are widely used in photovoltaics and thermoelectrics as they are earth-abundant, less toxic, chemically stable and environment-friendly [11–14]. Germanium and tin chalcogenides have attracted a great deal of interest due to their large Seebeck coefficient, high power factor and low thermal conductivity [15, 16]. In addition, lead and tin chalcogenides are investigated intensively as TE materials due to their intrinsic lattice anharmonicity and structural anisotropy, which are useful in improving the TE performance [17–20]. The TE materials based on SnTe alloys have been explored with great interests in the industrial and automotive applications as alternatives of PbTe-based alloys due to the toxicity associated with Pb to the environment and human health [21–25]. Although SnTe is nontoxic, earth-abundant and known to be stable in the intermediate temperature range of 400-900 K, it has much smaller Seebeck coefficient and $ZT$ values in its pristine bulk form as compared to that of PbTe. This is related to the inherent Sn vacancies of SnTe, which lead to higher hole concentrations [25].

Lowering the dimensionality is one of the efficient methods for increasing the TE efficiency due to the increase in the Seebeck coefficient and the electrical conductivity along with the lattice thermal conductivity reduction [9, 26, 27]. Two-dimensional (2D) monolayers (MLs) of black phosphorus and group-IV chalcogenides are found to have much enhanced TE performance than their bulk forms [28–30]. Moreover, ultrathin SnTe layers and SnS thin films are recently synthesized along with their diverse structural, electronic and ferroelectric properties [31–33]. Recent theoretical studies of SnTe MLs predicted promising $ZT$ values above 3.81 in $\beta$ (hexagonal)-phase at 900 K and $\sim$ 1.46 in $\gamma$ (rectangular)-phase at 700 K [30, 34]. The SnTe ML has zigzag and armchair like projections of atoms within the plane. The bilayer structure of a layered compound can be created by stacking two MLs (one at the top of other in a certain pattern). Importantly, the most stable type of stacking (AA, AB, etc.) at room temperature entirely depends on the chemical and structural properties of the corresponding material. The most stable bilayer structure of group IV-VI compounds, such as GeSe and SnSe, was found to be the AA stacked configuration [35, 36]. Although there are intensive studies on the TE properties of SnTe MLs, reports on its bilayer structures are rare, which is the stimulus of the present work.

In this work, the structural, electronic, lattice dynamics and TE properties of AA $+s$ (upper ML shifted by $\frac{1}{6} a_1$) SnTe bilayer are presented. The rest of this paper is organized as follows: section “Computational methods” presents the computational methods implemented in this work, section “Results and discussions” includes the main results and discussion and, finally, the concluding remarks are presented in section “Conclusions”.

### Computational methods

Calculations are based on the density functional theory (DFT) combined with the Boltzmann transport formalism as implemented in VASP package [37]. The exchange and correlation energy contributions were described by the Perdew-Burke-Ernzerhof (PBE) functional [38], and the ionic cores of the atoms were represented within the projector augmented-wave (PAW) formalisms as the pseudopotentials [39].

The van der Waals interaction is included by using the vdW-DF scheme [40] with the optimized cut-off energy of the plane wave expansion set to 500 eV. The Brillouin zone (BZ) is sampled using $18 \times 18 \times 1$ Monkhorst-Pack (MP) $k$-point grids, centered at $\Gamma$-point for the structural optimization. A vacuum region of 15 Å perpendicular to the $z$-direction was employed to avoid the interactions between the periodic images of the unit cell. Total energies and forces were converged up to 0.001 meV per atom and 0.001 eV/Å, respectively. The electronic transport calculations were performed using the semiclassical Boltzmann transport theory as implemented in the BoltzTraP2 package (version—20.2.1) [41]. Sufficiently dense $k$-point grids of $36 \times 36 \times 1$ were used to compute the electronic density of states (DOS) calculations. The deformation potential (DP) theory [42] was applied to evaluate the carrier mobility and the relaxation time as applied in previous studies for the similar 2D and 1D materials [29, 36, 43–46].
The lattice dynamics and vibrational stability were investigated using PHONOPY [47]. A supercell of $4 \times 6 \times 1$ of SnTe bilayer unit cell with the $4 \times 4 \times 1$ $k$-point grids were used for the phonon dispersion and the second-order (harmonic) interatomic force constants (IFCs) calculation with a default finite displacement of 0.01 Å. The third-order (anharmonic) IFCs were calculated using a supercell of $4 \times 4 \times 1$ with the $4 \times 4 \times 1$ $k$-point grids, where the interactions up to the fifth nearest neighbor atoms were considered. The second- and third-order IFCs are evaluated as $\phi_{ij}^{(n)} = \frac{\partial^2 V}{\partial r_i^a \partial r_j^b}$ and $\phi_{ijk}^{(n)} = \frac{\partial^2 V}{\partial r_i^a \partial r_j^b \partial r_k^c}$, respectively, where $V$ is the potential energy of the phonon system, $r$ represents the atomic positions of the corresponding atoms ($i$, $j$, and $k$) and $a$, $b$, $c$ represent the cartesian directions of the finite displacements [48, 49]. The calculated second- and third-order IFCs are used in ShengBTE code [48] to evaluate the lattice thermal conductivity and other thermodynamic parameters. The input file used in this calculation is available in the supplementary information.

The structural vibrational stability of SnTe bilayer was tested by using the Car-Parrinello molecular dynamics (MD) calculations, as implemented in SIESTA package [50], at 700 K for an NPT ensemble. A $4 \times 4 \times 1$ supercell was used up to 17,000 fs with 1.5 fs time resolution for each MD step. Standard DZP basis sets with Troullier-Martins pseudopotentials considering van der Waals correction by Berland and Per Hyldgaard were employed [51–53]. The MD calculations were performed using the identical Parinello-Rahman mass and Nose mass of 3500 Ry and 4.56 Å along the armchair ($x$-) and zigzag ($y$-) directions, respectively. The optimized structure of AA + s stacked SnTe bilayer is shown in Fig. 1, where Fig. 1a represents the top view of the SnTe bilayer structure. Fig. 1b, c are the side views along the armchair and zigzag directions, respectively. It can be clearly noticed that the SnTe bilayer is tilted along the $z$-$x$ plane (with an angle of 77°), as shown in Fig. 1b. The detailed optimized lattice constants along with the atomic positions are presented in the supplementary information.

In addition, the MD calculations at 700 K were performed to confirm the stability of AA + s stacked SnTe bilayer, see Fig. 2. The results here indicate that the total energy of the AA + s stacked structure remains almost invariant after 2 ps (with an average energy of $-21,174$ eV). Therefore, the SnTe bilayer should be thermally stable in the temperature range of 300 K – 700 K, within which the TE properties are calculated.

The electronic band structure of AA + s stacked SnTe bilayer is presented in Fig. 3. The SnTe bilayer is found to have an indirect bandgap of 0.72 eV without spin–orbit coupling (SOC) interaction, which reduces to 0.65 eV upon including the SOC. The conduction band minimum (CBM) is found along $\Gamma - \mathrm{X}$ high-symmetry path, whereas the valence band maximum (VBM) is found along $\Gamma - \mathrm{Y}$ path, as shown in Fig. 3. The electronic bandgap here is smaller than the experimental value $\sim 0.9$ eV [31], which is a typical problem of the Kohn Sham theory to underestimate band gaps up to 50% [54, 55]. The electronic DOS plot shows that p-orbitals of Sn and Te atoms have a significant contribution to the sharper
peaks at the conduction (n-type) and valence (p-type) bands, respectively. As the peaks near the valence and conduction band are nearly equal, AA + s stacked SnTe bilayer can be suitable for both p- or n-type dopings in TE applications.

Carrier mobility and relaxation time

The carrier mobility ($\mu$) for a 2D system is evaluated using the following formula [44, 56]:

$$\mu_{2D} = \frac{2e\hbar^3 C^{2D}}{3k_B T m^* m_d E_1^2},$$

where $e$ is the electronic charge, $\hbar$ is the reduced Planck’s constant, $T$ represents the temperature, $C^{2D}$ is the 2D elastic modulus, $E_1$ is the DP constant, $m^*$ is the effective mass and $m_d$ is defined as $m_d = \sqrt{m_x m_y}$. Here $m_x$ and $m_y$ are the effective masses along the armchair ($x$-) and zigzag ($y$-) directions, respectively. The value of $C^{2D}$ can be obtained by fitting the energy-strain curves along the armchair/zigzag directions. The two curves are nearly identical as shown in Fig. S1 (supplementary information), and, respectively, the values of $C^{2D}$ along the two directions are very close (86.65 N m$^{-1}$ along the armchair and 89.84 N m$^{-1}$ along the zigzag direction). The DP constant ($E_1$) can be defined as $E_1 = \frac{\delta E_{edge}}{\delta\delta}$. The shift in CBM by applying the uniaxial strain along both directions is shown in Fig. S2 (supplementary information).
The effective mass \( m^* \) for charge transport can be computed from the band structure. The \( m^* \) value of the electron (hole) at CBM (VBM) along the armchair and zigzag directions can be calculated as \( m^* = \frac{\hbar^2}{2\partial^2 E(k)/\partial k^2} \), where the term \( \partial^2 E(k)/\partial k^2 \) is obtained as the second derivative of the band energy \( E(k) \) with respect to \( k \) vector (along \( \Gamma - X \) and \( \Gamma - Y \) for armchair and zigzag directions, respectively). At 300 K, similar effective masses for electrons (0.15\( m_e \) along \( x \)- and \( y \)-directions) and holes (0.12\( m_e \) along \( x \)- and 0.15\( m_e \) along \( y \)-directions) are predicted, which is attributed to the similar parabolic nature of band dispersions of CBM and VBM (see Fig. 3). Based on the obtained values of \( C^{2D}, E_I \) and \( m^* \), the carrier mobility \( \mu_{2D} \) can be calculated using Eq. (4). Normally, the adopted value of relaxation time \( \tau \) is in the order of \( 10^{-14} \) s within the constant relaxation time approximation [16, 57–59]. However, the \( \tau \) actual value of a specific material may vary depending on its physical properties [29, 34, 36, 44]. Here, the relaxation time \( \tau \) is evaluated using the relation [29, 34, 60]:

\[
\tau = \frac{m^* \mu_{2D}}{e}
\]

The calculated \( C^{2D}, E_I, m^*, \mu_{2D} \) and \( \tau \) for AA + s stacked SnTe bilayer at 300 K and higher temperatures are compiled in Table 1. The SnTe bilayer is found to have relatively high relaxation time and mobility as compared to the other group IV–VI layered compounds as a consequence of its low deformation potential, high elastic modulus and the moderate value of effective mass [29, 34, 36].

**Lattice dynamics and thermal conductivity**

The TE efficiency of a material is contributed by its lattice and electronic transport properties. The phonon dispersion relation is a good measure to test the vibrational stability and lattice dynamics of a system. The positive phonon frequencies, as shown in Fig. 4a, indicate that the AA + s stacked SnTe bilayer is dynamically stable. As the unit cell has eight (N) atoms there are 24 (3N) phonon branches, where the lowest three are the acoustic modes and rest phonon branches are the optical modes. Out of three acoustic modes, two are transverse acoustic (TA) and one is longitudinal acoustic (LA). TA1 and TA2 are clearly noticed to be degenerate along the \( X - S - Y \) path, as shown in Fig. 4a. The phonon DOS is depicted in Fig. 4b, which shows that both Sn and Te atoms occupy the similar frequency range since there is no significant difference in the atomic weight of both atoms. The group velocity \( (v_g) \), which is defined as \( v_g = \partial \omega(k)/\partial k \), is an important factor in determining the lattice thermal conductivity \( (\kappa_l) \) since it is directly proportional to \( v_g \) (see Eq. 6). From Fig. 4c, the minority of the TA2, LA and optical branches are found to contribute to higher group velocity.

In addition to \( v_g \), the Grüneisen parameter \( (\gamma) \) and the specific heat capacity \( (C_v) \) are also important quantities to calculate \( \kappa_l \). The Grüneisen parameter \( (\gamma) \), which measures the anharmonicity of a system, is inversely related to \( \kappa_l \) according to Slack’s theory [61]. Accordingly, the value of \( \gamma \) is found to increase with increasing the temperature and reaches the saturation on increasing the temperature, as shown in Fig. 5a. The temperature dependence of \( C_v \), which is obtained using the second and third-order IFCs, is presented in Fig. 5b. The value of \( C_v \) is found to increase by increasing temperature and starts to saturate near 700 K approaching the classical limit of Dulong and Petit [62]. The total converged scattering rate along the irreducible \( \mathcal{q} \)-points at 300 K is displayed in Fig. 5c, which indicates that the scattering rate is larger for the branches that have a larger value

| Direction   | Carrier | \( E_I \) (eV) | \( C^{2D} \) (Nm\(^{-1}\)) | \( m^* \) (\( m_e \)) | \( \mu_{2D} \) (cm\(^2\) V\(^{-1}\) s\(^{-1}\)) | \( \tau \) (10\(^{-14}\) s) |
|-------------|---------|----------------|-----------------|----------------|----------------|----------------|
| Armchair    | e       | -4.09          | 86.65           | 0.15           | 3158.06        | 27.47          | 16.48          | 11.77          |
|             | h       | -4.09          | 86.65           | 0.12           | 4413.52        | 30.71          | 18.43          | 13.16          |
| Zigzag     | e       | 4.68           | 89.84           | 0.15           | 2503.24        | 21.77          | 13.06          | 9.33           |
|             | h       | 4.68           | 89.84           | 0.15           | 2798.70        | 24.34          | 14.61          | 10.43          |

Here \( m_e \) represents the rest-mass of an electron.

**Table 1** Deformation potential constant \( (E_I) \), 2D elastic constant \( (C^{2D}) \), effective mass \( (m^*) \), carrier mobility \( (\mu_{2D}) \) and relaxation time \( (\tau) \) of the SnTe bilayer for electrons \( (e) \) and holes \( (h) \) at different temperatures along the armchair and zigzag directions.
The effect of grain size on the $\kappa_l$ values at different temperatures (300 K, 500 K and 700 K) can be investigated by plotting the cumulative $\kappa_l$ as a function of mean free path (MFP) (see Fig. 5d). As an example, the Grüneisen parameter and specific heat capacity as a function of temperature are shown in Fig. 5a and 5b, respectively. The phonon scattering rate at 300 K for different phonon branches is shown in Fig. 5c.
example, at 300 K an MFP of 0.79 Å is required to change the κ⊥ value by about 50% (from 0.165 W m⁻¹ K⁻¹ to 0.252 W m⁻¹ K⁻¹). Large MFP phonons contribute to a significant amount on the cumulative κ∥ values at a lower temperature (300 K). The cumulative κ∥ values are found to increase with increasing the MFP (for all temperatures) and saturate at a certain threshold.

The lattice thermal conductivity (κ) is calculated using the following equation [48]:

$$\kappa = \frac{1}{k_B T^2 \Omega N} \sum_{\lambda} f_0 \left( f_0 + 1 \right) \left( \frac{\hbar \omega_{\lambda}}{k_B T} \right)^2 v_{\alpha}^2 F_{\beta}^1,$$

where $k_B$, $N$, $T$ and $f_0$ are the Boltzmann constant, the number of q-points, temperature and the phonon (Bose–Einstein) distribution function, respectively; $\omega_{\lambda}$ and $v_{\lambda}$ are the angular frequency and group velocity of phonon mode $\lambda$, respectively. Using the iterative method for phonons, the calculated κ values of AA + s stacked SnTe bilayer along the armchair and zigzag directions are shown in Fig. 6, which indicates an anisotropic behavior. This anisotropy in κ is closely related to the anisotropy of phonon group velocity, as depicted mathematically in Eq. (6) [63, 64].

The calculated value of κ is 1.63 (1.91) W m⁻¹ K⁻¹ at 300 K along armchair (zigzag) direction, which is much lower as compared to the bulk SnTe experimental value (~ 3 W m⁻¹ K⁻¹) [65, 66], other 2D compounds such as graphene, phosphorene, SnS, SnSe MLs [29, 67, 68]; higher as compared to SnSe bilayer [36] and γ-SnTe ML [30]. The value of κ decreases with the increase of temperature due to the anharmonic phonon scattering resulting from the lattice vibration at higher temperatures [69]. The lower κ value contributes to the enhancement of the TE performance of a material by the increase of its figure of merit (ZT).

### Thermoelectric properties

The Seebeck coefficient (S) and electrical conductivity (σ) are calculated using the expressions [41]:

$$S = \frac{\int_{-\infty}^{\infty} g(E, T) (E - \mu) \left( -\frac{\partial f(E, \mu, T)}{\partial E} \right) dE}{q T \int_{-\infty}^{\infty} g(E, T) \left( -\frac{\partial f(E, \mu, T)}{\partial E} \right) dE},$$

(7)

$$\sigma = q^2 \int_{-\infty}^{\infty} g(E, T) (E - \mu) \left( -\frac{\partial f(E, \mu, T)}{\partial E} \right) dE,$$

(8)

where $g(E)$, $f(E, \mu, T)$, $\mu$, $q$ and $T$ correspond to the carrier energy, transport function, Fermi–Dirac function, chemical potential, carrier charge and temperature, respectively. The transport function is given as:

$$g(E, T) = N(E) v^2(E) \tau(E),$$

(9)

where $N(E)$, $v(E)$ and $\tau(E)$ are the density of states, Fermi velocity and scattering time, respectively.

A similar carrier concentration range (of the order $10^{12}$–$10^{13}$ cm⁻²) is adopted for a better comparison with the similar layered compounds [29, 34, 36]. The Seebeck coefficient (S) as a function of the carrier concentration ($n$) for the SnTe bilayer along the armchair and zigzag directions at 300 K, 500 K and 700 K is presented in Fig. 7a. The negative values of $n$ represent the electron carrier concentration ($n$-type), whereas the positive values represent the hole carrier concentration ($p$-type). Figure 7a shows an abrupt increase in the values of $|S|$ near the Fermi level due to the Dirac delta function behavior of $\frac{\partial f}{\partial E}$ term, where $|S|$ values are found to decrease by increasing the carrier concentration with a discontinuity at $n = 0$. Furthermore, the value of $S$ is found to increase by increasing the temperature. This implies (i) the occurrence of more frequent carrier scattering events at higher temperatures and (ii) no visible effect of the bipolar conduction at higher temperatures for the SnTe bilayer, in agreement with the experimental results of bulk SnTe [66, 70]. Interestingly, the calculated Seebeck coefficients of bulk SnTe using the Boltzmann transport theory were found to agree well with the experimental results [66, 70, 71], which highlights the potential reproducibility of the
present work in experiments. The maximum value of $S$ for SnTe bilayer here ($>400 \ \mu V\ K^{-1}$) is significantly larger than that of bulk SnTe ($\sim 90 \ \mu V\ K^{-1}$) [66], but smaller as compared to the SnTe ML ($\sim 600 \ \mu V\ K^{-1}$) [30, 34]. This behavior is ascribed to the fact that the bipolar conduction is inversely proportional to the electronic bandgap [25, 72].

The electrical conductivity ($\sigma$) and electronic thermal conductivity ($\kappa_e$) are obtained within the constant relaxation time approximation as $\sigma/\tau$ and $\kappa_e/\tau$ using BoltzTraP2 [41]. The computed values of $\tau$ (see Table 1) are used to obtain $\sigma$ and $\kappa_e$ at different temperatures. The calculated $\sigma$ as a function of carrier concentration ($n$) at 300 K, 500 K and 700 K along the armchair and zigzag directions is shown in Fig. 7b. The value of $\sigma$ increases with the increase of $n$ as they are directly proportional to each other (Eq. (8)). The electrical conductivity ($\sigma$) is found to decrease by increasing temperature—this is related to the intrinsic electrons scattering mechanism at higher temperatures. The value of $\sigma$ for SnTe bilayer here is lower as compared to its bulk form due to the increase in the bandgap. Interestingly, $\sigma$ values are slightly higher than those of SnTe ML [30, 34, 73], indicating a possible enhancement in the TE performance of the bilayer structure. The electronic thermal conductivity ($\kappa_e$) results from the heat transport by electrons and holes in a system. The calculated $\kappa_e$ is found to increase as a function of $n$ at different temperatures (see Fig. 7c). The $\kappa_e$ value is found to be relatively larger for the $p$-type than that of the $n$-type material. The increase in the $\kappa_e$ value by increasing temperature is related to the excitation of more electrons at higher temperatures.

The power factor ($S^2\sigma$) values of SnTe bilayer along the armchair and zigzag directions are presented in Fig. 8a, b, respectively, for electrons and holes at 300 K, 500 K and 700 K. There is no clear monotonic change in the value of $S^2\sigma$ with the temperature as a result of a trade-off between the $S$ and $\sigma$ values. The $S^2\sigma$ value remains as high as 40 up to 96.65 mW K$^{-2}$ m$^{-1}$ within a wide range of temperatures in a carrier concentration range of the order $10^{12}$–$10^{13}$ cm$^{-2}$, where the peak is relatively higher for holes than that for electrons due to the higher electrical conductivity and Seebeck coefficients of holes.

The TE figure of merit ($ZT$) values obtained using the calculated TE parameters along the armchair and zigzag directions, as a function of carrier concentration at different temperatures, are shown in Fig. 8c, d, respectively. The $ZT$ value of SnTe bilayer is found to be higher along the armchair direction in the case of $p$-type, whereas the $n$-type shows a higher $ZT$ value along the zigzag direction. Overall, SnTe bilayer shows better TE performance along the zigzag than the armchair direction, which is mainly due to the higher Seebeck coefficients along the zigzag direction. The $ZT$ peaks are predicted to be 3.48 (4.61), 2.27 (3.41) and 1.48 (3.86) along the armchair (zigzag) direction at 700 K, 500 K and 300 K, respectively. These values are significantly higher than those of bulk SnTe [66], other 2D group IV MLs (Sn, Pb and Ge chalcogenides) [29, 30, 34, 57] and SnSe bilayer [36], as compiled in Table 2. This behavior is attributed to the high Seebeck coefficient, high electrical conductivity and low lattice thermal conductivity of SnTe bilayer. The high values of $ZT$ suggest AA + s stacked SnTe bilayer as a promising material for TE device applications and fabrications.
Conclusions

Two-dimensional Sn-chalcogenides are recognized as potential candidates towards green energy applications due to their significantly low thermal conductivity and high \( ZT \) values. Motivated by these facts, first-principles calculations combined with the Boltzmann transport theory were performed to investigate the structural, electronic, phonon transport and thermoelectric properties of 2D SnTe bilayer. The structural optimization, molecular dynamics and phonon transport calculations predicted AA + s stacked SnTe bilayer as the energetically most stable configuration among several stackings. Being a narrow bandgap semiconductor, SnTe bilayer is found to have a high Seebeck coefficient, high electrical conductivity and low lattice thermal conductivity. These properties lead to significantly high \( ZT \) values of 3.48 and 4.61 along the armchair and zigzag directions, respectively. This study not only presents the detailed promising TE properties of SnTe bilayer, but also stimulates further experimental and theoretical studies with suitable dopants on few layers of group IV chalcogenides regarding TE efficiency enhancement and device applications in emerging technologies.

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Author contributions

A. Pandit performed all the calculations and wrote the manuscript. B. Hamad supervised the work and

Table 2 Maximum values of figure merit for SnTe bilayer and other phases/compound

| Compound          | Maximum \( ZT \) |
|-------------------|-----------------|
|                   | \( h \) (p-type) | \( e \) (n-type) |
| SnTe bilayer      | 4.61            | 4.11           |
| Previous study    |                 |                |
| SnTe bulk [66]    | \(~ 0.3\)       | \(\)           |
| SnTe ML [34, 73]  | \(~ 2.5, 2.2\)  | 3.81, 2.9      |
| SnSe bilayer [36] | \(~ 0.38\)      | 0.78           |

Figure 8 Power factors \((S^2\sigma)\) and TE figure of merit \((ZT)\) of AA + s stacked SnTe bilayer as a function of the carrier concentration along the armchair (a, c) and zigzag (b, d) directions.
edited the manuscript. R. Haleoot assisted A. Pandit in the computational tasks of this work.

**Compliance with ethical standards**

**Conflict of interest** The authors declare no conflicts of interest.

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