Pressure-induced enhancement in the thermoelectric properties of monolayer and bilayer SnSe$_2$

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The electronic structures of monolayer and bilayer SnSe$_2$ under pressure were investigated by using first-principles calculations including van der Waals interactions. For monolayer SnSe$_2$, the variation of electronic structure under pressure is controlled by pressure-dependent lattice parameters. For bilayer SnSe$_2$, the changes in electronic structure under pressure are dominated by intralayer and interlayer atomic interactions. The $n$-type thermoelectric properties of monolayer and bilayer SnSe$_2$ under pressure were calculated on the basis of the semi-classical Boltzmann transport theory. It was found that the electrical conductivity of monolayer and bilayer SnSe$_2$ can be enhanced under pressure, and such dependence can be attributed to the pressure-induced changes of the Se–Sn antibonding states in conduction band. Finally, the doping dependence of power factors of $n$-type monolayer and bilayer SnSe$_2$ at three different pressures were estimated, and the results unveiled that thermoelectric performance of $n$-type monolayer and bilayer SnSe$_2$ can be improved by applying external pressure. This study benefits to understand the nature of the transport properties for monolayer and bilayer SnSe$_2$ under pressure, and it offers valuable insight for designing high-performance thermoelectric few-layered SnSe$_2$ through strain engineering induced by external pressure.
1. Introduction

Thermoelectric materials, which can directly and reversibly convert heat into electricity, have potential applications in power generation and refrigeration [1]. The conversion efficiency of thermoelectric materials is described by the figure of merit, $ZT$, which is defined as $ZT = S^2 \sigma T / (\kappa_e + \kappa_l)$, where $S$, $\sigma$, $\kappa_e$ and $\kappa_l$ are the Seebeck coefficient, electrical conductivity, electric and lattice thermal conductivities, respectively. A good thermoelectric material must possess high $ZT$ value. The current researches are focused on enhancing the $ZT$ value by increasing the Seebeck coefficient and electrical conductivity values while reducing the value of thermal conductivity. However, this is not easy because the correlation and coupling among Seebeck coefficient $S$, electrical conductivity $\sigma$ and electric thermal conductivity $\kappa_e$ make it extremely difficult to control independently, and the parameter of lattice thermal conductivity $\kappa_l$ is related to crystal structure that is difficult to manipulate as well. As a result, most of the recent efforts in $ZT$ improvement have been concentrated around these compounds that possess intrinsically low lattice thermal conductivity and then optimize electronic properties $S^2 \sigma$ (power factor: PF).

Because of their quantum confinement effect, two-dimensional materials such as metal dichalcogenides have recently gained attention due to anharmonicity leading to low thermal conductivities and high figures of merit [2,3]. For example, the highest dimensionless figure of merit $ZT$ of monolayer WSe$_2$ can reach 0.8 at 1200 K [4]. Moreover, it was reported that a maximum $ZT$ of 1.65 was achieved at 300 K in ZrS$_2$ monolayer due to the $\kappa_l = 3.29$ W K$^{-1}$m$^{-1}$ [5]. Recently, the binary compound SnSe$_2$ is predicted as a promising thermoelectric material based on it possessing comparative $\beta$ value with that of SnSe [6]. Based on the naturally layered structure which can possess very low lattice thermal conductivity, the two-dimensional SnSe$_2$ compound is believed to be a potential candidate for thermoelectric materials. As a member of the metal dichalcogenides, SnSe$_2$ is a hexagonal close-packed CdI$_2$-type structure, and it consists of alternating sandwiched sub-layers bonded by van der Waals (vdW) interaction [7]. Ding et al. [8] studied the thermoelectric properties of $n$-type SnSe$_2$ and the calculated optimal $ZT$ of 2.95 in the $a$ direction at 800 K was observed due to the strong anisotropic feature. Just like the other metal dichalcogenides, SnSe$_2$ can be easy to construct two-dimensional sheets due to the weak interlayer bonding [9]. On the theoretical side, electronic and magnetic properties of SnSe$_2$ nanostructures under strains were calculated using first-principles calculations, and the controllable electronic and magnetic properties were presented [10]. Li et al. [11] use thermoelectric properties of SnSe$_2$ monolayer by using first-principles methods combined with Boltzmann transport theory, showing that SnSe$_2$ monolayer is a promising candidate for thermoelectric applications.

Applying variable pressure is proved to be an effective tool to tune the electronic and thermoelectric properties in some thermoelectric semiconductors [12]. The thermoelectric transport properties of these bulk compounds, such as SnSe [13], BiCuOSe [14], Bi$_2$Te$_3$ [15], PbTe [16], were investigated, and remarkable improvements of their thermoelectric properties under pressure were observed. For transition metal dichalcogenides, the electronic properties of single-layer and multilayer MoS$_2$ under pressure were analysed [17] and the effect of high pressure on the electronic and transport properties of 2H-MoS$_2$ was also systematically studied [18]. Additionally, the thermoelectric properties of bulk SeS$_2$ under hydrostatic pressure were investigated and an enhancement about 3.8 times of PF in the $c$ direction at 20 GPa was observed [19]. For SnSe$_2$, the crystal structure of bulk SnSe$_2$ under pressure was studied by experiment [20]. As for layered transition-metal dichalcogenides, the figure of merit of these compounds is limited by the moderate PF due to their intrinsically low electronic conductivity [21], suggesting that a strategy to enhance thermoelectric performance $ZT$ of layered transition-metal dichalcogenides is to increase their electrical transport properties. Recently, it has been found that higher electrical conductivity can be obtained for the monolayer and bilayer transition metal dichalcogenides when compared with bulk materials, and such phenomenon can be explained by the valley degeneracy at the band edge [21]. On the other hand, the thermoelectric properties of $n$-type layered transition-metal dichalcogenides can be enhanced under either normal compressive strain or biaxial compressive strain [22]. Meanwhile, application of hydrostatic pressure can offer an effective path to realize the reduction of out-of-plane and in-plane lattice constants as well; thus, applying external pressure is expected to be a potential method for improving thermoelectric performance of two-dimensional layered transition-metal dichalcogenides. Owing to low-dimensional structure and application of hydrostatic pressure, the thermoelectric properties of few-layered SnSe$_2$ can be enhanced; however, the theoretical predictions of few-layered SnSe$_2$ under pressure are lacking. Therefore, it is necessary to understand how the high pressure affects the electronic structures and transport properties of monolayer and bilayer SnSe$_2$, to evaluate the role of interlayer atomic interactions and to explore the origins of the improvement of the thermoelectric parameters under pressure.
In this paper, the effect of pressure on the electronic and transport properties of monolayer and bilayer SnSe$_2$ was systematically investigated by using first-principle calculations and semi-classic Boltzmann transport theory. The relationships among lattice parameters, electronic structures and thermoelectric properties of monolayer and bilayer SnSe$_2$ under pressure were estimated, and the nature of difference in electronic conductivity of monolayer and bilayer SnSe$_2$ under pressure was analysed. According to the doping dependence of electrical transport properties, we estimated the changes of PF of monolayer and bilayer SnSe$_2$ with the increasing pressures. It is expected that the thermoelectric performance of few-layered SnSe$_2$ can be effectively improved by applying external hydrostatic pressure.

2. Computational methods

All calculations of the structural and electronic properties were carried out by using the projector-augmented wave [23] methods, as implemented in Vienna ab initio Simulation Package (VASP) [24]. The exchange correlation energy has been calculated using generalized gradient approximation (GGA) in the Perdew–Burke–Ernzerhof (PBE) form [25]. The cutoff energy of the plane-wave expansion is chosen to be 400 eV for all the calculations. The convergent criterion was $10^{-6}$ eV for energy and 0.03 eV Å$^{-1}$ for force. The bulk SnSe$_2$ compound is crystallized in the space group P$ar{6}$m with a layered crystal structure, as shown in figure 1a. For the geometric and self-consistent calculations of bulk SnSe$_2$, a well-converged Monkhorst–Pack k-point grid of $7 \times 7 \times 5$ is used to sample the Brillouin zones. The interlayer Se and Se interactions in SnSe$_2$ are dominated by vdW interactions, which are overestimated in the simple GGA method [26]. In order to describe the interlayer interactions well, the vdW corrections using Grimme’s vdW-D2 method are included in our calculations [27]. To apply external (hydrostatic) pressure in bulk SnSe$_2$, we chose to add external stress to stress tensor in VASP code [24,28]. The structural change of bulk SnSe$_2$ at different pressures up to 20 GPa was investigated, and lattice parameters and atomic positions of bulk SnSe$_2$ under pressures are shown in figure 1b,c. As shown in figure 1b, the calculated lattice constants $a$ and $c$ of bulk SnSe$_2$ at zero pressure are 3.835 and 6.153 Å, respectively, which are very close to experimental data ($a = 3.81$ Å and $c = 6.14$ Å) [29], and this is because the method of vdW-D2 can provide an excellent description of the crystal structure of bulk SnSe$_2$ [26,27]. Figure 1b presents the variation of lattice constants of bulk SnSe$_2$ under different pressures. From 0 to 20 GPa, the lattice constants of bulk SnSe$_2$ decrease with the increase of hydrostatic pressures. It can also be found that the in-plane lattice constant $a$ is much more weakly affected by pressure than out-of-plane lattice constant $c$. Meanwhile, the layer thickness $d$ decreases under pressure, reflecting the fact that interlayer van der Waals interactions change with pressure. In order to gain an insight into the change of intralayer atomic positions under pressure, the bond and angle parameters at different pressures are shown in figure 1c. As the pressure increases from 0 to 20 GPa, the length of Se–Sn bond is decreased while the distance of Se–Se increases slightly under pressure, resulting in increase of the band angle under pressure. These structural parameters of bulk SnSe$_2$ under pressure are then used to construct single- and double-layer SnSe$_2$ structures corresponding to different pressures, and this approach has been applied to obtain these structures of single-layer and multilayer MoS$_2$ under pressure [17]. For each monolayer or bilayer SnSe$_2$, a vacuum region of 15 Å is added in the direction normal to the layers to avoid the interaction between the periodic images. The Brillouin zone is sampled using a Monkhorst–Pack k-point mesh ($13 \times 13 \times 1$) for the geometry optimizations and self-consistent calculation.

The BoltzTraP package was employed to calculate transport properties of single-layer and bilayer SnSe$_2$ under different pressures [30]. The principle of this code is to solve transport properties based on semi-classical Boltzmann theory in conjunction with rigid band and constant relaxation time approximations, and it can yield accurate results for various types of thermoelectric materials [14–16,18,19,31,32]. In order to gain well-converged transport quantities, we employ a finer $35 \times 35 \times 1$ k-mesh to calculate the transport properties.

3. Results and discussion

3.1. SnSe$_2$ monolayer under pressure

As shown in figure 2a,b, the model of the SnSe$_2$ monolayer can be viewed as cleaved from the SnSe$_2$ surface, where a Sn atom is sandwiched between two Se atoms that formed a Se–Sn–Se triple layer. To investigate high pressure effect on the band of SnSe$_2$ monolayer, hydrostatic pressures from 0 to 20 GPa were applied. The typical band structures of SnSe$_2$ monolayer along the symmetry lines at
different pressures are shown in figure 2c–e. At zero pressure, the band of SnSe$_2$ monolayer shows an indirect band gap feature as reported [11]: the conduction band minimum (CBM) is located at M, while the valence band maximum (VBM) is located between $\Gamma$ and M points. The calculated electronic band gap of SnSe$_2$ monolayer at 0 GPa is 0.77 eV, which is slightly lower than the previous calculated value 0.85 eV [11], in which vdW corrections have not been considered. Additionally, we can see that an electronic pocket of the conduction band located between $\Gamma$ and M points. The depth of the electronic pocket of the conduction band along the $\Gamma$–M direction near Fermi level can determine the electronic effective mass, and it then can determine the electrical conductivity of $n$-type SnSe$_2$ monolayer, thus, it is a very important parameter (denoted as $g_2$) for describing the band structure characteristic SnSe$_2$ monolayer. With the increase of pressure from 0 to 20 GPa, the band gap of SnSe$_2$ monolayer decreases almost linearly. On the other hand, the conduction band parameter $g_2$ increases as pressure increases, which means the electron effective mass of SnSe$_2$ monolayer becomes smaller with
pressure. As discussed above, the reduction of conduction band effective mass brings out the decrease of the Seebeck coefficient along with the increase of the electrical conductivity of $n$-type thermoelectric materials [33–36]. Therefore, the increase of conduction band parameter $g_2$ under pressure will cause change of the transport properties of $n$-type SnSe$_2$ monolayer.

In order to further understand the electronic structure under pressure, the partial densities of states (PDOS) of monolayer SnSe$_2$ in the energy interval between $-2$ eV and 3 eV at pressure of 0, 10 and 20 GPa are shown in figure 3. From the PDOS at zero pressure, we can see that the CBM of monolayer SnSe$_2$ primarily comes from Se $4p$ and Sn $4d$ orbitals, whereas the VBM is dominated by the Se $4p$ orbital and Sn $5p$ and $4d$ orbitals. The characteristics of PDOS of CBM and VBM in monolayer SnSe$_2$ are similar to bulk SnSe$_2$ [37]. When the high pressure is applied, an obvious shift to the $E_F$ of both the Sn $5s$ orbital and Se $4p$ orbital in the valence band in PDOS can be observed, resulting in the decrease of the band gap. On the other hand, the movement of Sn $5s$ orbital and Se $4p$ orbital in the valence band gradually getting close to the Fermi level under pressure is the reason of the change of conduction band parameter $g_2$, which can determine the transport properties of $n$-type SnSe$_2$ monolayer.

In order to further understand the band engineering induced by hydrostatic pressure, the partial charge densities in the conduction band bottom at M point of monolayer SnSe$_2$ under different pressures were plotted in figure 4. At free states without external pressure, the conduction band bottom at M point is mainly contributed by the Sn $5s$ orbital and Se $4p$ orbital according to the calculated PDOS,
Figure 3. Calculated projected density of states of monolayer SnSe$_2$ at different pressures. The Fermi levels are set to zero.

and there exist obvious antibonding characteristics between Se and Sn atoms. As seen from figure 4, the charges of Sn atom tends to escape from Sn atomic site which means the Sn 5s orbitals become delocalized with the increase of the external pressure; that is, the Se–Sn antibonding states become more hybridized with increasing pressure, which causes the electrical conductivity increase under external pressure. Such pressure-dependent distribution of charge density is also observed in other thermoelectric materials [14]. The charge delocalization of Sn 5s orbitals under pressure is consistent with the analysis from PDOS, and it can be ascribed to the change of Se–Sn bond length under pressure. With the increase of external pressure, the Se–Sn bond length decreases linearly, and then the overlapping of the Se and Sn atomic orbitals will rise, and it will drive the charges of Sn atom to gather close to the Sn atom.

We now turn our attention to assess the transport properties of monolayer SnSe$_2$ under pressure. Since experimental works reveal that SnSe$_2$ tends to form n-type semiconductors [33–36], we only considered the n-type doping in the following thermoelectric properties of monolayer or bilayer SnSe$_2$. It is reported that the effect of spin–orbit interaction on the electronic structure of monolayer or bilayer SnSe$_2$ is negligible [11]; therefore, the spin–orbit coupling effect is not included in layer-dependent studies of electronic transport properties in this work. In figure 5, we plot the results of n-type monolayer SnSe$_2$ for the Seebeck coefficient, electrical conductivity and PFs at different pressures as a function of number of electrons per unit cell at room temperature. As can be seen in figure 5a, the Seebeck coefficient reduces slightly when applying external pressure, and such phenomenon is also observed in other thermoelectric systems under pressure [14,18,19]. As shown in figure 5b, the pattern of electrical conductivity with respect to relaxation time ($\sigma/\tau$) change with pressure is just opposite: the $\sigma/\tau$ increases with the increase of pressure. The electrical conductivity $\sigma/\tau$ of n-type monolayer SnSe$_2$ increases with pressure, which agrees with the above analysis of the PDOS and partial charge density of monolayer SnSe$_2$ under pressure. Combining the values of Seebeck coefficient and electrical conductivity, we can obtain the PFs with respect to relaxation time ($S^2\sigma/\tau$). The results of $S^2\sigma/\tau$ of n-type monolayer SnSe$_2$ under pressure as a function of electrons per unit cell are shown in figure 5c. Comparing with the PFs at 0 GPa, the values of $S^2\sigma/\tau$ increase under pressure in the whole range of carrier concentrations, and this is because the reduction of Seebeck coefficient is excessively compensated by the electrical conductivity under pressure. The PF $S^2\sigma/\tau$ of n-type monolayer SnSe$_2$ can be improved under pressure, indicating that the band structure manipulated by external pressure is a potent method to improve the thermoelectric properties of n-type monolayer SnSe$_2$. 
Figure 4. Contour plots of the partial charge density in the conduction band bottom at M point of monolayer SnSe$_2$ on the Se–Sn–Se plane at different pressures. (a) 0 GPa, (b) 10 GPa and (c) 20 GPa. The Fermi levels are set to zero. The unit of charge density is e Å$^{-3}$.

3.2. Bilayer SnSe$_2$ under pressure

Previous sections have only discussed the monolayer SnSe$_2$ for band structure and electronic transport properties under pressure. The effect of pressure on monolayer SnSe$_2$ is primarily from changing the intralayer atomic interactions. In order to further investigate the influences of intralayer and interlayer atomic interactions of the layered SnSe$_2$, the pressure-dependence of band structure and transport properties of bilayer SnSe$_2$ under pressure are also discussed in this work. The top and side views of SnSe$_2$ bilayer are shown in figure 6a–b, respectively. The distance between the layers and minimum total energy of bilayer SnSe$_2$ at different pressures are shown in figure 6c. From 0 to 20 GPa, the distance between the layers decreases with the increase of hydrostatic pressures, which can result in the strengthening of interlayer van der Waals interactions under pressure. Meanwhile, we can see from figure 6c that the total energy of bilayer SnSe$_2$ increases as the increase of pressure. The total energy of
Figure 5. Calculated electronic transport coefficients of n-type monolayer SnSe$_2$ at different pressures as a function of number of electrons per unit cell at 300 K. (a) Seebeck coefficient $S$, (b) electrical conductivity with respect to relaxation time $\sigma/\tau$ and (c) PFs with respect to relaxation time $S^2\sigma/\tau$.

bilayer SnSe$_2$ at zero pressure is the minimum, which means the structure under the free state is the most stable. With the pressure increasing, the minimum total energy of bilayer SnSe$_2$ increases, suggesting that the stable structure of bilayer SnSe$_2$ at higher total energy can be developed under proper geometrical constraint.

Figure 7a–c presents the band structures of bilayer SnSe$_2$ for 0, 10 and 20 GPa. From the band structure at 0 GPa, an indirect gap of 0.65 eV can be obtained, which is slightly less than the one (0.77 eV) of the monolayer SnSe$_2$. The layer-dependent band gap is observed in SnSe$_2$ and other two-dimensional transition metal dichalcogenide materials [22,26]. As in the case of bilayer SnSe$_2$, the CBM is located at M point while the VBM is located between $\Gamma$ and M points, and there are pockets in the valence band. Figure 7a–c, displays the calculated band structure of bilayer SnSe$_2$ under different pressures. It is found
Figure 6. (a,b) Top and side view of bilayer SnSe₂. (c) The distance between the layers and the total energy of bilayer SnSe₂ as a function of pressure.

Figure 7. (a–c) Band structures of bilayer SnSe₂ under pressure for 0, 10 and 20 GPa, respectively. (d) Partial band structure of bilayer SnSe₂ at 0 GPa. (e) Band gaps and change of band parameters of bilayer SnSe₂ as a function of pressure.
that the band gap decreases under pressure and the pockets of the valence band change with pressure. Meanwhile, the conduction band structure of bilayer SnSe$_2$ contains two pairs of parabolic bands which are different with monolayer SnSe$_2$. The important band parameters of bilayer SnSe$_2$ are indicated in figure 7 and then summarized in figure 7. Similar to the monolayer SnSe$_2$, the pocket parameter of conduction band ($\Delta_2$) increases almost linearly. Additionally, there is a parameter $\Delta_1$ which represents the width of valence band splitting, and it grows linearly as the pressure increases.

To gain a deeper insight of the pressure-dependent electronic structure of bilayer SnSe$_2$, the projected density of states of bilayer SnSe$_2$ at different pressures are plotted in figure 8. It reveals that the valence band of bilayer SnSe$_2$ consists of Se 4$p$ and Sn 5$s$ orbitals, and the energy states near Fermi level are also affected by external pressure: the peaks of Se 4$p$ and Sn 5$s$ orbitals move toward the Fermi level with the increase of pressure. This tendency is in accordance with the PDOS of monolayer under pressure. The variation of energy states near Fermi level can determine the transport properties of bilayer SnSe$_2$ under pressure.

In order to further explore the subtle differences of electronic states between bilayer and monolayer SnSe$_2$, the partial charge densities corresponding to CBM of bilayer SnSe$_2$ are shown in figure 9. At zero pressure, the charge densities around Se and Sn atoms of bilayer SnSe$_2$ are less than the monolayer one, which means the coupling of the Se–Sn antibonding states in bilayer SnSe$_2$ becomes weaker than monolayer SnSe$_2$, resulting in bilayer SnSe$_2$ possessing less electrical conductivity than monolayer. In view of the construction methods of few-layered SnSe$_2$, the intralayer atomic positions of bilayer and monolayer SnSe$_2$ are the same, and the intralayer atoms have the same impact on the charge density distribution. Thus, the decrease of charge densities in bilayer SnSe$_2$ compared to monolayer can be ascribed to the presence of interlayer atomic interactions: the charge densities around Sn and Se tend to gather in the direction of adjacent layer in bilayer SnSe$_2$ due to the coupling of interlayer introduced by vdW interactions. Meanwhile, another obvious feature is that the charge densities distribution is asymmetric in upper and lower parts, as shown in figure 9a, and this is because the upper two atoms are close to the vacuum region while the lower two atoms are close to the adjacent layer, resulting in the charge redistribution induced by layers coupling. With the increase of pressure, the charge densities around Se atoms become denser (figure 9), suggesting the Se–Sn antibonding states become more hybridized with increasing pressure, and it will enhance the electrical conductivity of bilayer SnSe$_2$ under pressure.

As a further support, the band-decomposed charge density at M point of CBM in bilayer SnSe$_2$ is shown in figure 10. It is found that there emerges an increasing charge density between the layers as
Figure 9. Contour plots of the partial charge density in the CBM at M point of monolayer SnSe$_2$ on the Se–Sn–Se plane. (a) 0 GPa, (b) 10 GPa and (c) 20 GPa. The Fermi levels are set to zero. The unit of charge density is eÅ$^{-3}$.

As pressure increases, and it can be ascribed to shortening of the layer thickness $d$ under pressure (figure 1b), reflecting the fact that the layers’ coupling strengthens, induced by interlayer van der Waals interactions under pressure.

Based on the electronic structure of bilayer SnSe$_2$ under pressure, we now discuss the effect of pressure on the transport properties in bilayer SnSe$_2$ based on semi-classical Boltzmann theory. In figure 11, we show the doping-dependent $S$, $\sigma/\tau$ and $S^2\sigma/\tau$ of n-type bilayer SnSe$_2$ under different pressures. From figure 11a, we can see that the Seebeck coefficient of bilayer SnSe$_2$ has a slight decrease under pressure in the whole carrier concentration, and this tendency is consistent with monolayer SnSe$_2$ under pressure. In figure 11b, we show the calculated electrical conductivity with respect to relaxation time $\sigma/\tau$ under pressure. We can see the value $\sigma/\tau$ of bilayer SnSe$_2$ at zero pressure is $0.434 \times 10^{19}$ $\Omega^{-1}$ m$^{-1}$ s$^{-1}$ at 3% $n$ doping, which is slightly lower than monolayer SnSe$_2$ ($0.498 \times 10^{19}$ $\Omega^{-1}$ m$^{-1}$ s$^{-1}$), and this is mainly caused by interlayer interactions in bilayer SnSe$_2$, as we discussed above. As shown in figure 11b,
Figure 10. Isosurfaces of band-decomposed charge density at M point of the conduction band bottom of bilayer SnSe$_2$ at different pressures. (a) 0 GPa, (b) 10 GPa and (c) 20 GPa. The red and blue balls represent Sn and Se atoms, respectively.

Figure 11. (a) Seebeck coefficient $S$, (b) electrical conductivity with respect to relaxation time $\sigma/\tau$, (c) PFs with respect to relaxation time $S^2\sigma/\tau$ of $n$-type bilayer SnSe$_2$ under pressure for 0, 10 and 20 GPa.
electrical conductivity with respect to relaxation time $\sigma/\tau$ of bilayer SnSe$_2$ increases with increasing pressure, and this is consistent with the previous analysis of the band structures and charge density distribution of bilayer SnSe$_2$ under pressure. Figure 11c shows the calculated $S^2\sigma/\tau$ of n-type bilayer SnSe$_2$ under pressure. It can be observed that although the Seebeck coefficient decreases with pressure, the PF of n-type bilayer SnSe$_2$ still increases because the electrical conductivity of the system is increasing [19]. The largest value of PF in n-type bilayer SnSe$_2$ at all pressures is found at 20 GPa which is computed to be $0.60 \times 10^{11}$ W K$^{-2}$ m$^{-1}$ s$^{-1}$ at the optimal carrier concentration, while the maximum value at 0 GPa is only $0.47 \times 10^{11}$ W K$^{-2}$ m$^{-1}$ s$^{-1}$. Therefore, the PF of n-type bilayer SnSe$_2$ increases by 27% by increasing the pressure to 20 GPa at 300 K. It is reported that the average PF of bulk SnS$_2$ can increase by 33% by increasing the pressure to 20 GPa at 800 K [19]. Bhattacharyya et al. [22] investigated the effect of strain on thermoelectric properties of few-layered MoS$_2$, and the PF of n-type layered MoS$_2$ at 900 K can be improved by 28% and 35% in the case of -8% normal compressive strain and biaxial compressive strain, respectively. Compared with these layered materials, it can be found that the enhancing of the pressure-dependent PF for n-type bilayer SnSe$_2$ is significant, suggesting that it can provide a useful guide for improving the thermoelectric performance of few-layered SnSe$_2$ through manipulating lattice strain induced by pressure.

4. Conclusion

In summary, we carried out first-principles calculations to investigate the effect of high pressure on the electronic structure and thermoelectric performance of monolayer and bilayer SnSe$_2$. Results show that the n-doping electrical conductivity of monolayer and bilayer SnSe$_2$ can be improved under pressure, and it is attributed to pressure-induced changes of the Se–Sn antibonding states in conduction band. Meanwhile, the discrepancy of transport properties in monolayer and bilayer SnSe$_2$ was estimated, and monolayer SnSe$_2$ is observed to possess higher electrical conductivity, and the electrical conductivity of monolayer SnSe$_2$ is only controlled by intralayer atomic interactions while that of the bilayer SnSe$_2$ is dominated by intralayer and interlayer atomic interactions. Our results clearly demonstrate that the PF of both monolayer and bilayer SnSe$_2$ can be improved by applying external pressure. This research can be useful to understand the origins of the transport properties for monolayer and bilayer SnSe$_2$ under pressure, and it offers valuable insight of the lattice strain induced by pressure as an efficient route for improving the thermoelectric performance of few-layered SnSe$_2$.

Data accessibility. All data to reproduce figures are available on Dryad (http://dx.doi.org/10.5061/dryad.v6d9n) [38]. Authors’ contributions. D.Z. and Y.L. designed the study; Y.O. and Y.G. performed calculations; D.Z. and C.Y. co-wrote the article. All authors discussed the results and commented on the manuscript. Competing interests. We declare we have no competing interests.

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