First-principles study on bilayer SnP₃ as a promising thermoelectric material†

Hongyue Song, a Xuehua Zhang, a Peiling Yuan, a Wencheng Hu a and Zhibin Gao a,b

The bilayer SnP₃ is recently predicted to exfoliate from its bulk phase, and motivated by the transition of the metal-semiconductor when the bulk SnP₃ is converted to the bilayer, we study the thermoelectric performance of the bilayer SnP₃ using first-principles combined with Boltzmann transport theory and deformation potential theory. The results indicate that the bilayer SnP₃ is an indirect band gap semiconductor and possesses high carrier mobility. The high carrier mobility results in a large Seebeck coefficient observed in both n- and p-doped bilayer SnP₃, which is helpful for acquiring a high figure of merit (ZT). Moreover, by analyzing the phonon spectrum, relaxation time, and joint density of states, we found that strong phonon scattering makes the phonon thermal conductivity extremely low (∼0.8 W m⁻¹ K⁻¹ at room temperature). Together with a high power factor and a low phonon thermal conductivity, the maximum ZT value can reach up to 3.8 for p-type doping at a reasonable carrier concentration, which is not only superior to that of the monolayer SnP₃, but also that of the excellent thermoelectric material SnSe. Our results shed light on the fact that bilayer SnP₃ is a promising thermoelectric material with a better performance than its monolayer phase.

1 Introduction

Thermoelectric (TE) materials have attracted more and more attention due to their ability to directly and reversibly convert waste heat and electricity. The conversion efficiency of a TE material is usually defined as the dimensionless figure of merit ZT, where S²σT/k, and σ represent the Seebeck coefficient, electrical conductivity, absolute temperature, and thermal conductivity composed of thermal conductivities of an electron (kₑ) and a phonon (kₚh), respectively. Here, the power factor S²σ is used to balance the properties of the electron. A potential high-performance TE material should have a high ZT value, that is, normally greater than 1 for commercial use. Unfortunately, because of the contradictory relationship between S and σ, together with the Wiedemann–Franz law: κₑ = LσT (L is the Lorenz number and for free electrons, L = 2.45 × 10⁻⁸ W Ω⁻¹ K⁻²), it is complex to enhance the TE conversion efficiency. Band structure engineering⁵ and optimal doping,⁶ for instance, are usually used to optimize the power factor. However, κₚh is a relatively independent factor to improve the ZT value,⁷–⁹ and searching materials with low κₚh is of great significance in the TE field.

For example, the famous layered TE material SnSe reported by Zhao et al.⁰ has an exceptionally low κₚh (0.23 ± 0.03 W m⁻¹ K⁻¹) and an unprecedented ZT of ∼2.6 at 923 K. Zhao et al.⁰ attributed the low κₚh in SnSe to the strong anharmonic and anisotropic bondings between layers. The other similar layered structures of bismuth oxychalcogenides including BiCuOX, Bi₂O₂X, and Bi₂O₂X₂ (X = S, Se, and Te) also possess extremely low κₚh, namely lower than 2 W m⁻¹ K⁻¹ at 300 K, and show a superior performance among thermoelectric materials.¹¹–¹³ Furthermore, much work has been reported to attribute their low κₚh to the anharmonicity of the interlayer coupling resulting from the chemical bonding between Bi and chalcogen atoms.¹³–¹⁵ In fact, for layered structures, the interlayer coupling provides an additional degree of freedom and thus can be used to tune the properties of materials, in particular for κₚh. Recently, Pei et al.¹⁶ have highlighted using stacking order, electric field, intercalation, and pressure of layered materials to control the interlayer coupling, thereby improving their physical properties. Besides, the strategies such as defect engineering,¹⁷ lone-pair electrons,¹⁸ and nanostructuring¹⁹ are also being used to enhance phonon scattering and thus achieving low phonon thermal conductivity.

Recently, novel two-dimensional (2D) SnP₃ materials including their monolayer and bilayer phases have been theoretically predicted to be extracted from their bulk counterpart using a mechanical exfoliation method with thermal stability.²⁰–²² The calculated cleavage energies reported by Ghosh et al.²⁰ for

* College of Science, Zhengzhou Key Laboratory of Low-dimensional Quantum Materials and Devices, Zhengzhou University of Technology, Zhengzhou 450007, China
a State Key Laboratory for Mechanical Behavior of Materials, Xi’an Jiaotong University, Xi’an 710049, China. E-mail: zhibin.gao@xjtu.edu.cn
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monolayer and bilayer SnP$_3$ are 0.71 and 0.45 J m$^{-2}$, respectively, which are comparable to that of graphene from the corresponding graphite (0.32 J m$^{-2}$).\textsuperscript{23} Significantly, bulk SnP$_3$ has been synthesized experimentally long since and shows metalllicity.\textsuperscript{24} Nevertheless, both the monolayer and bilayer SnP$_3$ are semiconductors having an indirect band gap.\textsuperscript{25–27} Monolayer and bilayer SnP$_3$ have been reported to possess high carrier mobility and optical absorption coefficient, suggesting their potential for applications in the optoelectronic area, such as anode materials in Na- and Li-ion batteries.\textsuperscript{20–22,25} Moreover, similar layered structures to SnP$_3$, that is, monolayer and bilayer GeP$_3$ are also reported to have a pronounced light absorption coefficient and remarkably high carrier mobility.\textsuperscript{28} Besides, in 2019, Zhu et al.\textsuperscript{29} reported theoretically that the monolayer SnP$_3$ is an excellent p-type thermoelectric material, namely its $ZT$ can reach 3.46 for optimal hole doping at 500 K, which is comparable to that of the well-known TE material SnSe (~2.6 at 923 K) mentioned above.\textsuperscript{10}

Furthermore, Zhu et al.\textsuperscript{29} also attribute the high $ZT$ value of the monolayer SnP$_3$ to its low phonon thermal conductivity (~4.97 W m$^{-1}$ K$^{-1}$ at 300 K) and high Seebeck coefficient (~900 $\mu$V K$^{-1}$ at 300 K). Based on the electronic band structure engineering and the TE transport property tuning,\textsuperscript{30} Wei et al.\textsuperscript{31} also reported the strain modulated thermoelectric performance of the monolayer SnP$_3$, and the results show that although $\kappa_{ph}$ increases, a considerable $ZT$ of 2.01 at 700 K is also obtained for the 6% strained case.

As far as we know, although the thermoelectric transport properties of the monolayer SnP$_3$ are widely studied, the electronic and the phonon transport properties of bilayer SnP$_3$ are still poorly understood. Note that the bilayer SnP$_3$ can be exfoliated from its bulk structure with dynamical stability, offering a new route for interlayer coupling compared with its monolayer phase. Since interlayer coupling plays an important role in the properties of electrons and phonons, we will perform a systematic investigation on the thermoelectric performance of the bilayer SnP$_3$, using first-principles theory accompanied by Boltzmann transport theory and deformation potential theory in this work. Our results indicate that the bilayer SnP$_3$ is a semiconductor with an indirect band gap (~0.9 eV), and it also possesses high carrier mobility. The flat electronic band structure and several sharp peaks in the electronic density of states (DOSs) near the Fermi energy lead to perfect thermoelectric properties. Furthermore, we found that its room-temperature phonon thermal conductivity is below 1 W m$^{-1}$ K$^{-1}$, resulting from the strong three-phonon scattering. Owing to the high power factor and the extremely low phonon thermal conductivity, we surprisingly discovered that a maximum $ZT$ value of 3.8 can be obtained for moderate p-type doping at 300 K. This value is higher than that of the monolayer SnP$_3$.\textsuperscript{29} Our results indicate that the TE performance of the bilayer SnP$_3$ can be improved compared to the monolayer SnP$_3$, and the bilayer SnP$_3$ is an excellent TE material.

### 2 Computational methods

First-principle calculations are carried out based on the framework of density functional theory (DFT) using the Vienna $ab initio$ Simulation Package (VASP)\textsuperscript{32,33} code. The generalized gradient approximation (GGA) in the form of the Perdew, Burke, and Ernzerhof (PBE)\textsuperscript{34} is adopted. For the structural relaxation, the plane wave energy cutoff is picked to be 500 eV, and a $15 \times 15 \times 1$ Monkhorst–Pack $k$ point mesh is used. The energy and the residual force on the atom are set to be $10^{-4}$ eV and 0.01 eV Å$^{-1}$, respectively, which can ensure the convergence of the structure relaxation. Moreover, the vacuum space between layers is set to 15 Å along the $z$-axis to avoid interlayer interactions. Considering that the PBE approach underestimates the electronic band gap of materials, we adopt the Heyd–Scuseria–Ernzerhof (HSE06)\textsuperscript{35} functional to obtain a more accurate electronic band gap of the bilayer SnP$_3$. According to the band structure obtained by the DFT calculations, we adopt the semiclassical Boltzmann theory within the rigid band method\textsuperscript{36} and constant relaxation time approximation as implemented within the BoltzTraP code\textsuperscript{37} to calculate the electrical transport coefficients such as $S$ and $\sigma/\tau$ except for the carrier relaxation time. In general, the carrier relaxation time $\tau$ is related to many factors such as the temperature, lattice structure and doping content.\textsuperscript{38} In our work, we adopt the deformation potential theory based on the effective mass approximation to calculate $\tau$.\textsuperscript{39} Here, the limitation of the method is that it only considers the dominated scattering contributed by the coupling between free carriers and longitudinal acoustic phonons. Nevertheless, the method has been proven to be valid to predict the mobility of two-dimensional materials, and the results are reliable.\textsuperscript{40–42} Here, based on the Wiedemann–Franz law, $\kappa_e = L_0 T$ where $L$ is the Lorenz number and we use it as $2.45 \times 10^{-8}$ W Ω K$^{-2}$ in our work,\textsuperscript{2,3} and the electrical thermal conductivity $\kappa_e$ can be acquired. In order to obtain accurate transport coefficients, a dense $45 \times 45 \times 1$ $k$-point mesh is used in the Brillouin zone (BZ). The phonon transport properties are obtained by the Boltzmann transport equation as realized in phonopy\textsuperscript{43} and phonon3py\textsuperscript{44} codes. Together with the VASP, the second- and the third-order interaction force constants (IFCs) are calculated by a finite-difference method using $3 \times 3 \times 1$ and $2 \times 2 \times 1$ supercells, respectively, and with a $2 \times 2 \times 1$ mesh $k$-points because of the computing resource. Besides, the energy convergence is set to $10^{-8}$ eV. Here, the carefully tested $k$-points of $21 \times 21 \times 1$ in the BZ are used to calculate the phonon thermal conductivity.

### 3 Results and discussion

The crystal structure of SnP$_3$ in the bulk phase is a widely known layered material and belongs to the trigonal space group $R\bar{3}m$ (166).\textsuperscript{45} From the side and top views, the structures of each layer combined with P and Sn atoms are analogous to those of blue phosphorene with a puckered type and graphene with a honeycomb type, respectively. Owing to the weak van der Waals interactions between the layers, it has been theoretically reported that the monolayer and bilayer SnP$_3$ can be relatively easy to exfoliate from the bulk phase based on DFT calculations.\textsuperscript{20–22} The calculated cleavage energies reported by

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SnP$_3$ (purple: P atoms and gray: Sn atoms). (c) Electronic band structures using the potentials of PBE and HSE06, and the total and partial DOSs of the bilayer SnP$_3$. The high symmetry $k$ points are $I$ (0, 0, 0), $X$ (0.5, 0, 0) and $M$ (0.33, 0.33, 0).

Ghosh et al.\textsuperscript{20} for monolayer and bilayer SnP$_3$ are 0.71 and 0.45 J m$^{-2}$, respectively, which can be comparable to that of graphene from the corresponding graphite (0.32 J m$^{-2}$).\textsuperscript{23} Afterwards, Sun et al.\textsuperscript{21} and Feng et al.\textsuperscript{22} also reported the theoretical cleavage energies of these two novel 2D materials and predicted that monolayer and bilayer SnP$_3$ can be fabricated using a stripping method. The structure of the bilayer SnP$_3$ is shown in Fig. 1(a and b). The optimized lattice parameter is $a = b = 7.49$ Å, which is consistent with the results reported by Ghost et al.\textsuperscript{20} Meanwhile, the value is a little larger than those of the in-plane of bulk SnP$_3$ (7.38 Å)\textsuperscript{46} and monolayer SnP$_3$ (7.37 Å).\textsuperscript{29} More details about lattice parameters and atomic positions of the bilayer SnP$_3$ are given in section A in the ESL.\textsuperscript{†}

In Fig. 1(c), we present the electronic band structures of the bilayer SnP$_3$ in PBE and HSE06 hybrid functional potentials and the total and partial electronic density of states (DOSs). The results show that the bilayer SnP$_3$ is a semiconductor with a valence band maximum (VBM) and a conduction band minimum (CBM) located at the $M$ and $I$ points, respectively. According to the method of the PBE potential, the calculated band gap is 0.5 eV, which is always underestimated compared to the experimental value. Then, the more accurate band gap obtained by the HSE06 functional is 0.9 eV. Thus, the next calculation of the electronic transport properties is all based on the HSE06 approach. It is worth mentioning that the two-fold degeneracy appears in the conduction band at the M point, which is useful for band engineering to improve the ZT value. For example, Wei et al.\textsuperscript{44} have reported the strain-tuned band structure and the thermoelectric properties of the monolayer SnP$_3$, and it possesses a promising ZT value at a 6% strained case.

Besides, Fig. 1(c) also shows that the valence band is flatter than the conduction band suggesting the higher Seebeck coefficients for the hole-doped SnP$_3$. From the DOS plots, we can see that the top of the valence band is contributed mainly by the P atoms, while the bottom of the conduction band is dominated by the Sn atoms. Further analysis shows that this contribution mainly originates from the p orbitals for both cases.

According to the electronic band structure, the electronic transport coefficients including the Seebeck coefficient, electrical conductivity, and power factor are obtained by using the Boltzmann transport theory (BTE) within the method of constant relaxation approximation (CRTA). The results as a function of the carrier concentration at 300, 500, and 700 K are shown in Fig. 2. We can see that the bilayer SnP$_3$ exhibits a high Seebeck coefficient for both p- and n-type doping, and the value of $S$ of hole doping is slightly higher than that of electron doping, which is ascribed to the flatter valence band than the conduction band displayed in Fig. 1, and, also leading to a higher effective mass of holes than that of the electrons presented in Table 1. For instance, the maximum Seebeck coefficients of 620 and 510 $\mu$V K$^{-1}$ at 300 K are obtained for the p- and n-doped bilayer SnP$_3$, respectively, which facilitated achieving high ZT values.

Moreover, the temperature-dependent Seebeck coefficient shows normal characteristics, that is, with the increasing temperature (both holes and electrons), the Seebeck coefficient gradually increases. Based on the BTE, the results of the electrical conductivity ($\sigma$/$\tau$) are shown in Fig. 2(c) and (d). Therefore, there is an issue that should be pointed out, namely, we need the carrier relaxation time which is hard to acquire with high precision. Here, we use the approach of the
deformation potential theory to evaluate $\tau$, and the detailed calculation process will be discussed in the next section.

Fig. 2(c) and (d) indicate that $\sigma/\tau$ shows an opposite tendency to temperature and the tendency of carrier concentration is found to be that of the Seebeck coefficient. Furthermore, owing to the smaller effective mass for electrons than the hole, a larger $\sigma/\tau$ of n-type doping is obtained than p-type doping for the same carrier concentration. Based on the calculated Seebeck coefficient and the electrical conductivity, the results of the power factor $(S^2/\sigma/\tau)$ are shown in Fig. 2(e) and (f). Looking at the temperature dependence, we found that, at 300 K, the $S^2/\sigma/\tau$ of electron doping is slightly higher than that of hole doping, indicating that, to improve the thermoelectric performance in the bilayer SnP$_3$, the n-type doping is more superior compared with p-type doping, which is contrary to the case of the monolayer SnP$_3$. However, at 500 and 700 K temperatures, both p- and n-type $S^2/\sigma/\tau$ show almost the same value for the same carrier concentration. Besides, $S^2/\sigma/\tau$ also shows the same temperature dependence with the Seebeck coefficient, rather than that of $\sigma/\tau$, which is similar to the previous studies about other materials. In the end, the temperature dependence of the chemical potential is considered by the formula: $dF = -pdV - SdT + \mu dN$, where $F$, $p$, $V$, $S$, $T$, $\mu$ and $N$ are the Helmholtz free energy, pressure, volume, entropy, temperature, chemical potential and number of particles, respectively. Then, we can make the more useful definitions: $\mu = \left(\frac{\partial F}{\partial N}\right)_{V,T}$.

In order to obtain the carrier relaxation time $\tau$, the deformation potential theory which has been proved feasible in many two-dimensional materials is adopted. The detailed calculation method is as follows:\textsuperscript{51,52}

$$\tau = \frac{\mu m^*}{e}$$

(1)

$$\mu = \frac{e\hbar^3 C_{2D}}{k_B T m^* m_0 E_l}$$

(2)

where $\mu$ and $m^*$ are the carrier mobility and effective mass, respectively. $C_{2D}$ (see Fig. S2(b), ESL\textsuperscript{†}) named the effective elastic modulus can be calculated by $C_{2D} = \frac{1}{S_0} \frac{\partial^2 E}{\partial (\Delta a/a_0)^2}$ where $E$, $S_0$, and $\Delta a/a_0$ are the total energy of the strained cases, the area of the unit cells, and the biaxial strain. $E_l$ (see Fig. S2(a), ESL\textsuperscript{†}) is the deformation potential constant calculated by the slope of the maximum energy at the top of the valence or the minimum value at the bottom of the conduction band as a function of the strained cases. $m_0$ is the average effective mass computed by $m_0 = \sqrt{m_{t-X}^* m_{X-K}^*}$. All the obtained parameters of the bilayer SnP$_3$ are shown in Table 1. Clearly, the effective masses are nearly equal along the directions of $I-X$ and $X-K$ for both electrons and holes. Due to the low effective mass and the deformation potential constant, large carrier mobilities are obtained in the bilayer SnP$_3$, namely 1833 and 1477 cm$^2$ V$^{-1}$ s$^{-1}$ for electrons and holes, respectively. Thus, the calculated relaxation times in the bilayer SnP$_3$ are 0.24 ps for electrons and 0.46 ps for holes, and more computational details are shown in Section C in the ESL\textsuperscript{†}.

Table 1  Effective mass $m^*$, elastic modulus $C_{2D}$, deformation potential constant $E_l$, carrier mobility $\mu$, and relaxation time $\tau$ for electrons and holes of the bilayer SnP$_3$ at 300 K

<table>
<thead>
<tr>
<th>Carriers $m^<em>_t-X(m_e)$ $m^</em>_X-K(m_h)$ ($\text{m}^2 \text{e}^{-1}$)</th>
<th>$E_l$(eV)</th>
<th>$\mu$(cm$^2$ V$^{-1}$ s$^{-1}$)</th>
<th>$\tau$(10$^{-13}$ s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>e</td>
<td>0.23</td>
<td>98.77</td>
<td>-4.48</td>
</tr>
<tr>
<td>h</td>
<td>-0.66</td>
<td>98.77</td>
<td>1.96</td>
</tr>
</tbody>
</table>

With the above discussion of the electronic transport properties of the bilayer SnP$_3$, the phonon dispersion and phonon thermal conductivity are now briefly discussed. Fig. 3(a) shows the calculated phonon dispersion and the phonon thermal conductivity as a function of the temperature ranging from 100 K to 900 K for bilayer SnP$_3$. The presented value decreases with the increasing $k_B T$. The calculated phonon thermal conductivity is almost equal to that of other two-dimensional materials.\textsuperscript{29,53,54}

![Fig. 3](image-url)

**Fig. 3** Phonon dispersion and phonon total and partial DOSs (a) and the phonon group velocity (b) of the bilayer SnP$_3$.  

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by Ouyang et al. and twice that of the monolayer GeP \(_3\) (~0.43 W m\(^{-1}\) K\(^{-1}\) reported by Wang et al.), which can be expected to achieve a large \(ZT\) value. In order to further understand the low \(\kappa_{\text{ph}}\), we calculate the phonon relaxation lifetime and the joint density of states (JDOS) shown in Fig. 4 to analyze the three-phonon scattering.

Fig. 4(b) shows that the phonon relaxation lifetime of the acoustic modes is longer than that of the optical modes, which is consistent with the fact that the acoustic modes usually contribute most of the phonon thermal conductivity. Besides, the shorter phonon relaxation lifetime compared with the monolayer phase signifies a stronger anharmonic phonon scattering process, and thus leading to the low phonon thermal conductivity for bilayer SnP\(_3\). Furthermore, the number of anharmonic phonon scattering named the JDOS is expressed as follows:

\[
D(\omega) = \frac{1}{N} \sum_{l, l'} \left[ \delta(\omega + \omega_{l1} - \omega_{l2}) + \delta(\omega - \omega_{l1} + \omega_{l2}) \right] + \frac{1}{N} \sum_{l, l'} \delta(\omega - \omega_{l1} - \omega_{l2}).
\]

(3)

Here, \(\omega_{l1}\) and \(\omega_{l2}\) are the frequency of the three-phonon scattering. From Fig. 4(c), we found that the large JDOS distributed in the low frequency range grows, while in the high frequency range the JDOS does not almost change. These results tend to increase the phase space for acoustics modes and anharmonic scattering, which results in the low \(\kappa_{\text{ph}}\) for bilayer SnP\(_3\).

With the electron and phonon transport coefficients calculated above, the \(ZT\) value of bilayer SnP\(_3\) can be estimated. The calculated \(ZT\) values with the change of the carrier concentration at 300, 500, and 700 K are shown in Fig. 5. The results indicate that the \(ZT\) value for hole doping is obviously higher than that for electron doping, which mainly results from the large power factor caused by the high carrier relaxation time for hole doping. Moreover, the hole doping case needs a higher carrier concentration than electron doping to obtain the maximum \(ZT\) value. The maximum \(ZT\) at 300 K is 3.8 for the hole doping bilayer SnP\(_3\) at an appropriate carrier concentration, which is not only higher than some famous thermoelectric materials such as SnSe (2.6), SnSe\(_2\) (2.95), black-P (0.22), phosphorene (0.3), and Bi\(_2\)Te\(_3\) (1.3), but also can be comparable to its similar layered structures of the monolayer GeP\(_3\) (3.3) and monolayer SnP\(_3\) (2.5) as shown in Fig. 6. In our work, only the scattering between electrons and longitudinal acoustic phonons are considered; in fact, other phonon modes, defects, and grain boundaries can also scatter electrons, leading to reducing the carrier mobility, and thus reducing the \(ZT\) value accordingly. Therefore, the current theoretical calculation will sometimes underestimate scattering and thus overestimate \(ZT\). Furthermore, it should also be pointed out that with the increasing temperature, the \(ZT\) values are improved for both hole doping and electron doping. In the end, due to the low \(\kappa_{\text{ph}}\),
we also calculate the electronic contribution to the figure of merit \(\text{ZT}_e\); here, \(\text{ZT}_e = S^2\sigma T/\kappa_e\), and the results are presented in Section D in the ESL. It shows a large difference to the \(\text{ZT}\); therefore, the contribution of lattice vibrations to the TE performance cannot be ignored for the SnP₃.

### 4 Conclusions

In summary, we have studied the thermoelectric transport properties of bilayer SnP₃ using DFT together with Boltzmann transport theory. Our results indicate that it is a semiconductor with an indirect band gap of 0.9 eV calculated using the HSE06 hybrid functional potential. Based on the flat valence and conduction band structures near the Fermi level, high Seebeck coefficients for both n- and p-type doping are observed. Moreover, the bilayer SnP₃ possesses high carrier mobility resulting from the low effective mass and the deformation potential constant, which is beneficial to electrical transport. Owing to the strong scattering phase space between the acoustic and optical phonon modes resulting in a short phonon lifetime, the bilayer SnP₃ exhibits an extremely low phonon thermal conductivity, namely 0.8 W m⁻¹ K⁻¹ at 300 K, and these results are useful to obtain a high thermoelectric performance. Together with the high power factor and the low phonon thermal conductivity, the maximum \(\text{ZT}\) value at 300 K can reach 3.8 with an appropriate carrier concentration for hole doping. Here, we should point out that only the dominated scattering mechanism contributed by the coupling between free carriers and longitudinal acoustic phonons is considered in our calculation methods, resulting in the calculated \(\text{ZT}\) values sometimes overestimated. As a matter of fact, the electron scattering by other phonon modes, defects, grain boundaries, etc. cannot be neglected in nanoscale materials, which will suppress the \(\text{ZT}\) value, resulting in overestimation of the calculated \(\text{ZT}\) value. A more accurate method to calculate the electronic transport properties is based on the maximally localized Wannier functions which can be calculated by the EPW code. Therefore, our results indicate that the bilayer SnP₃ is a promising thermoelectric material, and we hope that an experimentally high \(\text{ZT}\) value of the p-doped bilayer SnP₃ can be achieved in the future.

### Conflicts of interest

There are no conflicts to declare.

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