**ABSTRACT:** Based on first-principles calculations, we explore the electronic and phonon transport properties of a new-type two-dimensional (2D) hexagonal material XSe (X = Ge, Sn, and Pb), which can be prepared by atomic isovalent substitutions of the recently synthesized crystal Ge₄Se₃Te. Among them, 2D PbSe possesses a large Seebeck coefficient of \( \sim 1150 \) \( \mu V/K \) and an ultralow lattice thermal conductivity of \( \sim 0.50 \) W/mK at room temperature. Theoretical calculations prove that the antiparallel movements of the atoms could lead to the strong optical-acoustic phonon coupling with low values of acoustic group velocities of 0.81–2.03 km/s and large Grünisen parameters of \( \sim 4 \), which accordingly greatly suppresses the heat transport ability. Using our calculated transport parameters, large values of the thermoelectric (TE) figure of merit (ZT) of 1.76, 2.32, and 3.95 can be obtained at an effective temperature range (GeSe and SnSe at 700 K and PbSe at 500 K) under p-type doping for 2D GeSe, SnSe, and PbSe, respectively. Interestingly, after checking several series of 2D materials, we find that their lattice thermal conductivities are almost proportional to their values of the lowest optical phonon frequencies. Our work clearly shows the advantages of these novel 2D group-IV selenides as TE materials and may stimulate further experimental and theoretical studies in this field.

1. **INTRODUCTION**

TE materials have been widely investigated in the scientific community on the basis of their ability to realize the conversion of heat and electricity. However, low conversion efficiency and high production costs have greatly hindered their commercial applications for decades. Usually, the conversion efficiency of the TE material is governed by a dimensionless quantity ZT:

\[
ZT = \frac{\sigma S^2 T}{\kappa_e + \kappa_l}
\]

where \( \sigma S^2 \) is the power factor (PF) including the Seebeck coefficient \( S \) and electric conductivity \( \sigma \), \( T \) is the absolute temperature, and \( \kappa_e \) and \( \kappa_l \) are electron and lattice thermal conductivities, respectively. In order to acquire high ZT values, low thermal conductivity and/or a high PF are essential.\(^{1-6} \)

However, optimizing these transport coefficients independently is relatively difficult because of complex correlations among them. Several effective strategies to enhance TE performance mainly improve the PF by modifying the electronic band structure\(^{7-9} \) and/or reducing the thermal conductivity through phononic crystal patterning\(^{10,11} \) or nanostructuring.\(^{12} \) Additionally, novel concepts have emerged such as band alignment and compositionally alloyed nanostructures, which can simultaneously keep a high PF and decrease thermal conductivity.\(^{13} \) These approaches can help people to effectively regulate the conflicting parameters and further enhance TE performance.

The successful preparation of graphene sheets\(^{14} \) has attracted people’s interests in 2D materials with potential applications in photovoltaic cells, nanodevices, and TE technologies.\(^{15-20} \) Among the well-known TE materials, PbTe\(^{21} \), SnSe\(^{22,23} \), and Bi\(_2\)O\(_2\)Se\(^{24} \) have been studied intensively in theory and/or experiment. Usually, 2D materials composed of group IV–VI elements show unprecedented TE performances, stemming from their low thermal conductivities, strong surface-phonon scattering, and high electronic transport abilities.\(^{25-28} \) For example, the phosphorene-like SnSe monolayer, a typical layered material, possesses an intrinsic low thermal conductivity of 2.02 W/mK at room temperature.\(^{29} \) With outstanding electronic transport properties, a high ZT value can reach up to 3.27 for n-type doping at 700 K. Considering these facts, investigations of the TE performances of novel monolayers in group IV–VI materials still deserve to be explored in experiments and theory.

Recently, a new-type of 2D material, XSe (X = Ge, Sn, and Pb), has been built successfully by equivalent substitutions of Ge and Te atoms in the layered Ge₄Se₃Te crystal.\(^{30} \) Unlike...
convergence of 10 forces on atoms are less than 0.01 eV/Å within a total energy. Crystal structures were completely optimized until the residual employed to sample the reciprocal space of the unit cell. All Kohn–Sham transport coefficients. We found that these monolayers of XSe exhibit very low thermal conductivities and relatively high Seebeck coefficients. These features endow them with high ZT values of 1.76, 2.32, and 3.95 at an effective temperature range for p-type doping of GeSe, SnSe, and PbSe, respectively.

2. COMPUTATIONAL METHODS

Our first-principles calculations were carried out by using the Vienna ab initio simulation package (VASP). We adopted the generalized gradient approximation in the Perdew–Burke–Ernzerhof form for the exchange-correlation functional. The cutoff energy of the plane wave was chosen as 500 eV in all calculations. To eliminate the interaction between adjacent images, the vacuum layer thickness was chosen as 20 Å along the vertical direction. An 11 × 11 × 1 k-mesh was employed to sample the reciprocal space of the unit cell. All crystal structures were completely optimized until the residual forces on atoms are less than 0.01 eV/Å within a total energy convergence of 10−6 eV/Å. The hybrid functional approach of Heyd–Scuseria–Ernzerhof (HSE06) was employed to calculate their electronic structures. The electronic transport properties were obtained by using the Boltzmann transport equation and the rigid band approach, implemented in the BoltzTraP code. On the basis of this approach, the theoretical calculation of many TE materials can be in good agreement with the experimental results. Herein, the electronic relaxation time was employed in the calculation of TE transport coefficients. To accurately Fourier interpolate the Kohn–Sham eigenvalues, a dense 45 × 45 × 1 k-mesh was chosen in the Brillouin zone (BZ).

The harmonic second-order interaction force constants (second IFCs) were obtained by the Phonopy code, employing 3 × 3 × 1 and 4 × 4 × 1 supercells for GeSe (SnSe) and PbSe, respectively, with a 3 × 3 × 1 k-mesh. Combining with the anharmonic third-order IFCs (third IFCs), we calculated the lattice thermal conductivity of XSe monolayers by solving the linearized phonon Boltzmann transport equation in Phono3py. Herein, an interaction range of 10 Å is considered with 1260 supercells, which meant full elements of third IFCs were computed. To accurately compute the lattice thermal conductivity, a very dense 200 × 200 × 1 k-grid was employed, which can reach the convergence criteria of the lattice thermal conductivity.

3. RESULTS AND DISCUSSION

3.1. Crystal Structures and Electrical Transport Properties. The XSe monolayers are narrow-gap semiconductors with a hexagonal crystal structure (space group P63m1, no. 164) and contain four atoms in each unit cell as shown in Figure 1. Similar to many layered 2D group IV–VI materials, it displays a graphene-like honeycomb texture with the insertion of atoms in the center of the hexagon from the top view, indicating that it may possesses stronger structural stability. The relaxed lattice constants for GeSe, SnSe, and PbSe are 3.81, 4.09, and 4.18 Å, respectively. Similar to their lattice constants, the interatomic distances of X-Se also exhibit an increasing trend: from 2.58 to 2.76 and then 2.84 Å. More details are summarized in Table 1.

The electronic structure can reflect the intrinsic characteristics of electrical transport. Herein, the band structures and density of states (DOSs) are presented for XSe sheets in Figure 1c–e. Obviously, these monolayers are indirect band-gap semiconductors with the valence band maximum (VBM) lying along the Γ–K path and the conduction band minimum

<table>
<thead>
<tr>
<th>type</th>
<th>a/b (Å)</th>
<th>lX−Se (Å)</th>
<th>lX−X (Å)</th>
<th>hX−Se (Å)</th>
<th>PBE (eV)</th>
<th>HSE06 (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GeSe</td>
<td>3.81</td>
<td>2.58</td>
<td>2.95</td>
<td>4.67</td>
<td>0.47</td>
<td>0.87</td>
</tr>
<tr>
<td>SnSe</td>
<td>4.09</td>
<td>2.76</td>
<td>3.32</td>
<td>5.19</td>
<td>0.51</td>
<td>0.89</td>
</tr>
<tr>
<td>PbSe</td>
<td>4.18</td>
<td>2.84</td>
<td>3.82</td>
<td>5.93</td>
<td>0.68</td>
<td>0.96</td>
</tr>
</tbody>
</table>
(CBM) located at the Γ point. In general, a good TE material should possess a narrow energy gap with the value being from 0.3 to 1.0 eV.\(^8\) At the PBE level, we can see clearly that XSe monolayers exhibit suitable energy gaps with the following order: GeSe (0.47 eV) < SnSe (0.51 eV) < PbSe (0.68 eV). Refined calculations based on the HSE06 method give enlarged gaps of 0.87, 0.89, and 0.96 eV, which are comparable to those of the polymorph SnSe monolayer (0.91 eV).\(^5\) From the orbital-resolved band structures, we can see clearly that the conduction band (CB) is contributed by the X atoms, while the valence band (VB) is occupied by the Se atoms. Additionally, the sharp peaks of DOSs can be observed at the VB top region. These band features are beneficial to high Seebeck coefficients and electrical conductivity.

The carrier mobility \( \mu \) of the electron and hole can be obtained by using the deformation potential (DP) theory\(^4\) for monolayer XSe, which has been extensively adopted to computing the transport properties of materials. For 2D systems, these parameters can be calculated by the formula as follows:\(^41\)

\[
\mu_{2D} = \frac{2e\hbar^3C_{2D}^2}{3k_B^2T}\tau \tag{2}
\]

\[
\tau = \frac{\mu m^*}{e} \tag{3}
\]

where \( C_{2D} \) is the 2D elastic constant, \( m^* \) is the effective mass, \( k_B \) is the Boltzmann constant, and \( E_i \) is the DP constant. The calculated carrier mobility, effective mass, and relaxation time at room temperature are tabulated in Table 2. Obviously, the carrier mobilities exhibit nearly isotropic characteristics, and the mobility of the electron is slightly larger than its hole part. Noticeably, there appears a decreasing trend of monotony from SnSe (174.21 cm\(^2\)/V s) to PbSe (152.61 cm\(^2\)/V s) for electron mobility, and the carrier mobility of monolayer XSe can be comparable to the monolayer MoS\(_2\) (60–200 cm\(^2\)/V s). For more parameter contents, refer to Table 2.

On the basis of above calculated band structures, the corresponding electrical transport properties can be obtained by using the Boltzmann transport theory with a constant scattering time approximation. Herein, we mainly study their Seebeck coefficient \( S \), electrical conductivity \( \sigma \), and electronic thermal conductivity \( \kappa_e \), which are crucial parameters for evaluating the TE performance of a material. Employing the rigid band approximation, effects of doping on electrical transport properties are studied. These transport coefficients with respect to \( T \) and \( \mu \) can be calculated as follows:\(^46\)

\[
S_{\text{DP}}(T, \mu) = \frac{1}{eTV\sigma_{\text{DP}}(T, \mu)} \int \Sigma_{\text{DP}}(\epsilon)(\epsilon - \mu) \left[ -\frac{\partial f_\mu(T, \epsilon)}{\partial \epsilon} \right] \tag{4}
\]

\[
\sigma_{\text{DP}}(T, \mu) = \frac{1}{V} \int \Sigma_{\text{DP}}(\epsilon) \left[ -\frac{\partial f_\mu(T, \epsilon)}{\partial \epsilon} \right] \tag{5}
\]

\[
\Sigma_{\text{DP}}(\epsilon) = \frac{e^2}{N_0} \sum_k tr_{\mu}(i, \mathbf{k}) \eta_{\mu}(i, \mathbf{k}) \delta(\epsilon - E_k) \tag{6}
\]

where \( \alpha \) and \( \beta \) are Cartesian indices and \( \Sigma_{\text{DP}}(\epsilon) \) is the transport distribution function. It is worth noting that XSe monolayers show large values of \( S \) with similar behaviors under n- and p-types of doping, as shown in Figure 2a–c. Especially for the 2D PbSe sheet, a large value of 1150 \( \mu V/K \) at room temperature is shown, which is significantly larger than that of GeSe (955 \( \mu V/K \)) and SnSe (899 \( \mu V/K \)). Compared with some state-of-the-art TE materials,\(^21,43\) XSe monolayers exhibit comparatively high Seebeck coefficients. Besides, we can see an obvious temperature-dependent behavior in that \( S \) decreases upon heating, which is a typical phenomenon for many TE materials. In the vicinity of low chemical potentials, we can find that the \( S \) of 2D XSe shows peaks because of the energy-dependent DOSs, that is, large \( dn(\epsilon)/d\epsilon \) in the vicinity of the Fermi level (see Figure S1). For the doped system, \( S \) can be defined by the Mott relation:\(^44\)

\[
S = \frac{\pi^2 k_B^2 T}{3e} \left( \frac{1}{n} \frac{dn(\epsilon)}{d\epsilon} + \frac{1}{\mu} \frac{dn(\epsilon)}{d\epsilon} \right) \tag{7}
\]

\( S \) can be further improved by two strategies: localized DOS or optimizing the carrier concentration. Through enhancing the energy-dependent DOSs, an improvement of \( dn(\epsilon)/d\epsilon \) can be realized. As shown in Figure 1, the increasing rates of the DOSs near the Fermi level for GeSe, SnSe, and PbSe are similar but different, following the order of GeSe < SnSe < PbSe, which is consistent with the order of the Seebeck coefficients. Thus, one can modulate the electronic properties through atomic replacement, in our case, replacing Ge by Sn or Pb.

The electrical conductivity (\( \sigma \)) as a function of chemical potential at 300 to 700 K is shown in Figure 2d–f. Unlike the Seebeck coefficient, \( \sigma \) exhibits obvious different behaviors for n- and p-types of doping. For XSe sheets, it can be seen that the \( \sigma \) of the p-type is evidently larger than that of the n-type in the low-chemical potential region (ranging from −0.4 to 0.4 eV) due to their different slopes. It should be noted that 2D GeSe and SnSe have a larger \( \sigma \) than that of 2D PbSe, stemming from the competitive relation between the Seebeck coefficient and electrical conductivity. Since conduction electrons obey Fermi–Dirac statistics, the slopes of the \( \sigma \) are gradually flat with temperature increasing near the low-chemical potential region. \( \kappa_e \) is essential to accurately predict the material’s TE performance. Herein, we further study the \( \kappa_e \) of XSe monolayers by solving the Wiedemann–Franz law:

\[
\kappa_e = L\sigma T \tag{8}
\]
where $L$ is the Lorenz number by $L = 1.5 + \exp[-8.5T]$ (see Figure S2). Similar to the electrical conductivity, $\kappa_e$ exhibits analogous curves as well, as shown in Figure 2g–i. Additionally, we can find that $\kappa_e$ presents a declining trend from GeSe to PbSe. Based on these analyses of $S$ and $\sigma$, we can conclude that the XSe monolayers have relatively good electrical transport abilities, which are beneficial for TE applications.

### 3.2. Photon Spectrum and Vibration Mode

The phonon spectrum is of great significance in the study of heat transport. It can reflect some intrinsic thermal properties, such as phonon velocities and Grüneisen parameters. The phonon spectrum can be obtained by using the second IFCs. As shown in Figure 3, no negative frequencies are observed in phonon spectra for our studied systems, indicating their dynamical stabilities at ambient pressure. Herein, we highlight three acoustic branches and the lowest optical branch with different colors, while remanent optical branches are black. Two transverse (ZA/TA) and one longitudinal acoustic (LA) modes get entangled with each other, which could strengthen the phonon scattering mechanism and thus lower $\kappa_l$. Compared with the GeSe and SnSe monolayers, 2D PbSe exhibits evident low and flat phonon spectra curves. One can see that their highest optical modes decrease successively, while their large optic–optic phonon gaps increase from GeSe (22.31 cm$^{-1}$) to SnSe (29.59 cm$^{-1}$) and then PbSe (65.47 cm$^{-1}$). Their phonon modes are becoming flat. Along with the increase of the phonon gap, the three low optical branches are lowered down to strongly mix with the acoustic branches, leading to strong phonon scattering and lowering the heat transfer. Specifically, 2D PbSe exhibits a very low optical mode of 20.87 cm$^{-1}$ at the $\Gamma$ point, which is much lower than that of GeSe (64.17 cm$^{-1}$) and SnSe (52.06 cm$^{-1}$). Figure 3 shows atomic motions of the acoustic phonon modes and the lowest optical mode near the $\Gamma$ point. For the lowest optical mode, one XSe layer moves in an opposite direction with respect to another XSe layer, vastly hindering the phonon transport in 2D XSe.

The thermal stability of monolayer XSe has been tested by performing ab initio molecular dynamics (AIMD) simulations (see Figure S3). In order to reduce the restraint of periodic boundary conditions, a $3 \times 3 \times 1$ supercell was employed in this calculation. The simulations were implemented at temperatures of 500 and 700 K. Results manifest that the monolayers GeSe and SnSe can be maintained for up to 10 ps.
at 700 K, while PbSe can only be maintained at 500 K. The above research shows that the thermal stability of PbSe is weaker than that of the GeSe and SnSe monolayers.

### 3.3. Lattice Thermal Conductivity

The lattice thermal conductivity is an extremely vital physical quantity for assessing the heat transport ability of materials. Based upon a self-consistent iterative approach,\(^1\)\(^2\) \(\kappa_l\) can be obtained by the following formula:

\[
\kappa_l = \frac{1}{V} \sum_i C_{i,q} v_{i,q}^2 \tau_{i,q}
\]

where \(C_{i,q}\) is the mode specific heat capacity, \(v_{i,q}\) is the phonon velocity, and \(\tau_{i,q}\) is the relaxation time. To obtain a more authentic \(\kappa_l\), we have corrected the correlation of the vacuum thickness and \(\kappa_l\). As shown in Figure 1b, the lengths between the topmost Se and bottom Se (\(h_{Se-Se}\) of XSe are 4.67–5.93 Å, and the van der Waals radii of Se are approximately 2.18 Å\(^5\)\(^3\)\(^4\) therefore, the effective thickness of monolayer XSe \(h_{eff} = 2.18 \times 2 + h_{Se-Se}\) is selected to precisely compute for \(\kappa_l\). Here, we can see clearly that \(\kappa_l\) will decrease gradually with rising temperature in Figure 4a because of the enhancement in the phonon–phonon scattering. This phenomenon has been testified to exist in many materials.\(^5\)\(^4\)–\(^5\)\(^6\)

In Figure 4a, an ultralow \(\kappa_l\) of \(\sim 0.50\) W/mK can be observed for 2D PbSe at room temperature, which is much lower than that of GeSe (5.81 W/mK) and SnSe (3.64 W/mK). Our computational values are comparable to those of excellent TE materials, such as single-layered SnSe sheets (\(\sim 2.5\) W/mK),\(^5\)\(^7\) PbTe (\(\sim 2.0\) W/mK),\(^5\)\(^8\) and monolayer Bi\(_2\)O\(_2\)Se (\(\sim 0.7\) W/mK).\(^5\)\(^9\) Such a low \(\kappa_l\) is an important prerequisite for achieving high TE performance. Additionally, the lattice thermal conductivity as a function of the inverse temperature is fitted in the inset of Figure 4a. \(\kappa_l\) is linearly proportional to \(1/T\) and approaches the minimum value at high temperature, implying that the Umklapp scattering plays an important role in this temperature range.

Figure 4b shows theoretical lattice thermal conductivities versus the corresponding values of the lowest optical modes (\(\omega_{o,min}\)) at the \(\Gamma\) point for group IV–VI isomers. Amazingly, we find that these compounds exhibit nearly similar trends, which is a very interesting phenomenon in that the lattice thermal conductivities are proportional to \(\omega_{o,min}\). Consistent with the previous report,\(^5\) these results adequately illustrate that a low \(\omega_{o,min}\) correlates with a low thermal conductivity. To study more heat transport properties, we compute for the correlative parameters and study them in the next section.

### 3.4. Phonon Velocity and Grüneisen Parameters

The phonon group velocity is an important physical parameter in heat transport. From eq 7, one can find that the phonon velocity is proportional to \(\kappa_l\). It can be computed by using phonon dispersion curves

\[
v_i(q) = \frac{\partial \omega_i(q)}{\partial q}
\]

where \(\omega_i(q)\) is the phonon frequency. In Figure 5a, we present our calculated phonon velocities. It is obvious that the phonon velocities exhibit different distribution curves, mainly due to different shapes of the corresponding phonon modes. In addition, some large phonon velocities can be found located near the \(\Gamma\) point, which are clearly different from those of monolayers with quadratic-trend phonon scattering curves.\(^6\)\(^0\)

The average value of the acoustic velocities of PbSe (1.35 km/s) is much smaller than that of 2D GeSe (3.24 km/s) and SnSe (2.70 km/s). This can be attributed to the low and flat acoustic branches for PbSe.

The Grüneisen parameter is also an essential physical quantity for studying \(\kappa_l\), which can quantize the anharmonicity of materials. It can be defined by

\[
\gamma_i(q) = \frac{\sum_{j} C_{ij} \omega_{j}^2 v_{j}^2}{\sum_{j} C_{ij} \omega_{j}^4}
\]
Herein, we calculate the Grüneisen parameters of 2D XSe and present them in Figure 5b. In general, a large $\gamma$ usually possesses a strong anharmonic scattering progress, which can greatly restrain the phonon transportation and give rise to a relatively low $\kappa_l$. For 2D PbSe, an unexpected large Grüneisen parameter ($\sim 4$) can be observed near the $\Gamma$ point, implying that it possesses strong anharmonic interactions. Additionally, we further compute for the average values of the acoustic Grüneisen parameters for 2D XSe. For the PbSe monolayer, the calculated value is 2.05, which is much larger than that of GeSe (0.71) and SnSe (0.96). The value for PbSe is slightly larger than that of PbTe (1.45) but smaller than that of layered SnSe (2.83). These inherent properties, such as the small phonon velocities and large Grüneisen parameters, can lead to an ultralow $\kappa_l$ for the PbSe monolayer.

### 3.5. Thermoelectric Figure of Merit

Using all our calculated lattice and electronic transport parameters above, the ZT values of XSe monolayers under different temperatures (300, 500, and 700 K) can be obtained (see Figure 6). In order to accurately evaluate TE properties of monolayer XSe, we employ the calculated $\tau$, which can be much larger than that of GeSe (0.71) and SnSe (0.96). The value for PbSe is slightly larger than that of PbTe (1.45) but smaller than that of layered SnSe (2.83). These inherent properties, such as the small phonon velocities and large Grüneisen parameters, can lead to an ultralow $\kappa_l$ for the PbSe monolayer.

The ZT values as a function of chemical potential for XSe monolayers are shown in Figure 6a–c. In general, the ZT values increase with increasing chemical potential $\mu$. The maximum ZT values at the effective temperature range are 1.76, 2.32, and 3.95 for GeSe, SnSe, and PbSe monolayers, respectively. Such high ZT values have greatly exceeded the standard for applicable TE materials (ZT $> 1.0$). Especially, for the PbSe monolayer, a surprising ultrahigh ZT value is obtained, which is much higher than that of the well-known TE material SnSe ($2.6 \pm 0.3$ at $T = 935$ K). In addition, the ZT values as a function of the carrier concentration are plotted in Figure 6d–f. The corresponding electron and hole concentrations to the maximum ZT values at 500 K for PbSe are approximately $5 \times 10^{19}$ cm$^{-3}$ and $2 \times 10^{19}$ cm$^{-3}$, respectively. These low concentrations have a weak effect on their phonon and electronic structure properties. Anyway, such high ZT values indicate that the XSe monolayers are promising TE materials.

### 4. CONCLUSIONS

In conclusion, we systematically study the TE properties of the 2D XSe by using DFT and Boltzmann transport equations. Results show that these materials own ultralow lattice thermal conductivities, especially for monolayer PbSe ($\sim 0.50$ W/mK at 300 K), which stem from the inherent phonon transport factors including small phonon velocities, large Grüneisen parameters, and strong optical-acoustic phonon coupling. Furthermore, a high value of the Seebeck coefficient ($\sim 1150 \mu$V/K) at room temperature is observed for the PbSe sheet. Combining with ultralow thermal conductivities and outstanding electrical properties, high ZT values of 1.76, 2.32, and 3.95 are found in p-type doping for 2D GeSe, SnSe, and PbSe, respectively. Such high ZT values demonstrate that XSe monolayers are excellent TE materials that could be applied in nanodevices. Our results offer a valuable perspective for exploring new TE materials in experiments.

### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jpcc.9b09787.

Discussion and some details of $\partial \ln(e)/\partial e$, the Lorenz number, and thermal stability of XSe monolayers (PDF)
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REFERENCES

The calculations were performed at the supercomputer center in the China Spallation Neutron Source. This work was
complete contact information is available at: https://pubs.acs.org/10.1021/acs.jpcc.9b09787

Notes
The authors declare no competing financial interest.

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