Phonon and electron transport in Janus monolayers based on InSe

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Abstract

We systematically investigated the phonon and electron transport properties of monolayer InSe and its Janus derivatives including monolayer In$_2$SSe and In$_2$SeTe by first-principles calculations. The breaking of mirror symmetry produces a distinguishable A$_1$ peak in the Raman spectra of monolayer In$_2$SSe and In$_2$SeTe. The long-range harmonic and anharmonic interactions play an important role in the heat transport of the group-III chalcogenides. The room-temperature thermal conductivity ($\kappa$) of monolayer InSe, In$_2$SSe and In$_2$SeTe are 44.6, 46.9, and 29.9 W (mK)$^{-1}$, respectively. There is a competition effect between atomic mass, phonon group velocity and phonon lifetime. The $\kappa$ can be further effectively modulated by sample size for the purpose of thermoelectric applications. Meanwhile, monolayer In$_2$SeTe exhibits a direct band gap of 1.8 eV and a higher electron mobility than that of monolayer InSe, due to the smaller electron effective mass caused by tensile strain on the Se side and smaller deformation potential. These results indicate that 2D Janus group-III chalcogenides can provide a platform to design the new electronic, optoelectronic and thermoelectric devices.

Keywords: Janus group-III chalcogenides, thermal transport, Boltzmann transport equation, mobility

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

1. Introduction

Indium Selenide (InSe), as an experimentally accessible layered group-III metal chalcogenide, has been receiving much attention [1]. The synthesized few-layer InSe exhibits small electron effective mass ($\sim 0.14 m_0$) and room-temperature electron mobility ($\mu$) higher than $10^3$ cm$^2$ (V s)$^{-1}$ [2, 3]. Nano-devices based on two dimensional (2D) InSe had good ambient stability [4]. In addition, 2D InSe holds promise for the application of bendable photodetectors with broadband response [5], 2D ferromagnets [6], excitonic dynamics [7], magneto-optical effects [8] and topological insulator [9].

Recently, a new Janus-type monolayer MoSSe has been synthesized through the replacement of the S atoms at one side of monolayer MoS$_2$ by Se atoms [10, 11]. Both band gap and thermal conductivity $\kappa$ of monolayer MoSSe are between that of monolayer MoS$_2$ and MoSe$_2$ [12]. As far as Janus structures based on 2D InSe are concerned, Kandemir et al found that the band structure of the monolayer In$_2$SSe was formed by the superposition of the strained band structures of the binary analog single layers [13]. Guo et al predicted that the 2D Janus group-III chalcogenides had enhanced piezoelectric coefficients compared to that of perfect ones [14]. The out-of-plane symmetry breaking in 2D Janus structures can also induce unusual properties such as enhanced Rashba effect [15], catalytic activity for hydrogen evolution [16], the valley polarization upon magnetic doping [17], excitons with long lifetime [18] and so on. Besides the electronic properties, thermal properties become a crucial issue for the device’s performance, as on-going miniaturization of electronic devices. For example, a high and low $\kappa$ are desirable for the efficient heat dissipation in integrated electronic devices and high
W Wan et al predicted that the $\kappa$ of monolayer InSe can be effectively reduced by boundary scattering [19]. Nissimagoudar et al predicted that the $\kappa$ of monolayer InSe, In$_2$SSe and In$_2$SeTe, respectively [19]. Pandey et al calculated the $\kappa$ of monolayer GaSe, GaSe, and InSe, and found that the increase of mass gave both decreasing acoustic phonon velocities and increasing scattering of heat-carrying modes [20]. The thermal properties of 2D materials can be tuned by doping or alloying [21], strain [22], chemical functionalization [23] and so on, expanding its application prospect. Therefore, Janus structures can not only expand the family of 2D materials but also offer another way to modulate the thermal properties of 2D materials. Though much efforts have been devoted to the physical properties of 2D Janus group-III chalcogenides, the mechanism of its phonon and electron transport are still not well understood.

In this work, we studied the structural, optical and transport properties of monolayer InSe, In$_2$SSe and In$_2$SeTe by first-principles calculations. Because of the breaking of mirror symmetry, Raman spectra of Janus structures exhibit a special $A_1'$ peak compared to that of monolayer InSe. The harmonic and anharmonic interaction in these group-III chalcogenides are long-range. The $\kappa$ of monolayer In$_2$SSe and In$_2$SeTe are larger and smaller than that of monolayer InSe, respectively. The mechanism of heat transport are carefully analyzed. At last, we calculated the electron mobility $\mu$ of monolayer In$_2$SeTe and its Janus structures. The $\mu$ of monolayer In$_2$SeTe is superior to that of monolayer InSe.

Table 1. The lattice constant ($a$), cation–cation bond length ($d_{cc}$), anion–cation bonding length ($d_{ac}$), effective thickness of monolayer ($l$), band gap ($E_g$), the frequency and irreducible representations of Raman peak of monolayer InSe, In$_2$SSe and In$_2$SeTe. The previous results [9] of monolayer InSe are displayed in bracket for a comparison.

|        | $a$ (Å) | $d_{cc}$ (Å) | $d_{ac}$ (Å) | $l$ (Å) | $E_g$ (eV) | $\omega_{\text{Raman}}$ (cm$^{-1}$) |
|--------|---------|--------------|--------------|---------|------------|----------------------------------|
| InSe   | 4.093   | 2.817        | 2.690        | 5.386   | 2.147      | $109 (A'_1)[110]$ [9] 178 ($E'$)[178] [9] 223 ($A'_1$)[225] [9] |
| In$_2$SSe | 4.015   | 2.819        | 2.671/2.585  | 5.289   | 2.297      | 124 ($A_1$) 214 ($A_1$) 257 ($A_1$) |
| In$_2$SeTe | 4.244   | 2.815        | 2.729/2.856  | 5.483   | 1.967      | 96 ($A_1$) 167 ($A_1$) 211 ($A_1$) |

Figure 1. (a) A top view and (b) a side view of crystal structure of monolayer InSe, In$_2$SSe and In$_2$SeTe, with the shaded area showing the unit cell. The $d_{cc}$ and $d_{ac}$ are the cation-cation and anion-cation bonding length, respectively. (c) The phonon dispersion of monolayer InSe, In$_2$SSe and In$_2$SeTe.
Based on the phonon Boltzmann transport function, lattice thermal conductivity $\kappa$ was estimated by [25]

$$\kappa_{\alpha \beta} = \frac{1}{N \Omega} \sum_{\mathbf{q},s} C_{\mathbf{q},s} v_{\mathbf{q},s,\alpha} v_{\mathbf{q},s,\beta} \tau_{\mathbf{q},s},$$

where $\alpha$ and $\beta$ are Cartesian indices. $N$ and $\Omega$ are the number of $\mathbf{q}$ point and volume of the unit cell, respectively. $C_{\mathbf{q},s}$, $\mathbf{v}_{\mathbf{q},s,\alpha}$ and $\tau_{\mathbf{q},s}$ are the mode specific capacity, group velocity and lifetime in the single-mode relaxation time approximation (RTA) of the phonon with wavevector $\mathbf{q}$ and branch index $s$, respectively. The phonon lifetime $\tau_{\mathbf{q},s}$ was estimated by combining the anharmonic scattering, isotopic impurities scattering and boundary scattering according to the Matthiessen rule [25]

$$\frac{1}{\tau_{\mathbf{q},s}} = \frac{1}{\tau_{\text{an,q,s}}} + \frac{1}{\tau_{\text{iso,q,s}}} + \frac{1}{\tau_{\text{b,q,s}}},$$

where boundary roughness scattering rate is $1/\tau_{\text{b,q,s}} = |v_{\mathbf{q},s}|/L$ with $L$ be the sample size.

All the first-principles calculations were performed by the Vienna $Ab$ initio Simulation Package (VASP) [26, 27] with the projector augmented wave (PAW) [28] pseudopotentials and Perdew, Burke, and Ernzerhof (PBE) [29] exchange-correlation functionals. An energy cut of 450 eV was used. A vacuum spacing of 15 Å was adopted to hinder the fictitious interactions between the adjacent supercells. The crystal lattice and ions positions were fully relaxed with a force convergence threshold of $10^{-4}$ eV $\cdot$ Å$^{-1}$. The band gap was modified by the Heyd–Scuseria–Ernzerhof (HSE06) screened
A hybrid functional [30]. A $6 \times 6 \times 1$ supercell was used to calculate the harmonic (2nd) and anharmonic (3rd) interatomic force constants (IFCs) with a finite displacement difference method [25]. The phonon dispersion was obtained by diagonalizing the dynamical matrix using the Phonopy code [31]. The dielectric constant tensor and Born effective charge tensor were considered to make a non-analytic correction to the dynamical matrix. The intensities of peaks in Raman spectra are calculated using density functional perturbation theory (DFPT) [32]. Based on the convergence test, the $\kappa$ was calculated by iteratively solving the phonon BTE using the ShengBTE code [25] with a $100 \times 100 \times 1$ Q-grid (see figure A1(a)). An cut-off distance of 16th nearest neighbors was adopted in the calculation of the anharmonic IFCs.

The phonon-limited carrier mobility of 2D material was estimated by [33]

$$\mu = \frac{e \hbar^3 C_{2D}}{k_B T m^* m' E_f^2},$$

where $e$, $\hbar$, $k_B$ and $T$ are the electron charge, Planck's constant, Boltzmann's constant and temperature, respectively. $m^*$ and $m'$ are the effective mass in the transport direction and average effective mass determined by $m^* = \sqrt{m_x m_y}$. The deformation potential constant $E_f$ is defined by $E_f = \Delta E / (\Delta l / l_0)$. Here, $\Delta E$ is the energy change of band edge under uniaxial strain of $(\Delta l / l_0)$. $l_0$ is equilibrium lattice constant and $\Delta l$ is the deformation along the transport direction. The 2D effectively elastic modulus $C_{2D}$ is fitting by $C_{2D} = [\partial^2 E / \partial (\Delta l / l_0)^2] / S_0$, where $E$ and $S_0$ are the total energy and the area of the unit cell, respectively. The band edge was aligned according to the vacuum level.

3. Results and discussion

3.1. The structure, phonons and Raman spectrum

Monolayer InSe consists of a quadruple layer in a stacking sequence of Se–In–In–Se and its lattice has $D_{3h}$ crystal symmetry (see figures 1(a) and (b)). Janus monolayer In$_2$SSe and In$_2$SeTe are built by replacing top-layer Se by S or Te atoms. Janus structures have $C_{3v}$ crystal symmetry without out-of-plane mirror symmetry ($z \rightarrow -z$). The thickness ($l$) of monolayer was estimated by the distance between the atoms on the outmost layer (see figure 1(b)). The optimized structural parameters are listed in table 1. The cation-cation bonding ($d_{cc}$) have a slight change compared to that of monolayer InSe. The anion-cation bonding length ($d_{ac}$) of In$_2$SSe and In$_2$SeTe are built by replacing top-layer Se by S or Te atoms. Janus structures have $C_{3v}$ crystal symmetry without out-of-plane mirror symmetry ($z \rightarrow -z$). The thickness ($l$) of monolayer was estimated by the distance between the atoms on the outmost layer (see figure 1(b)). The optimized structural parameters are listed in table 1.

Figure 4. (a) The temperature dependence of the $\kappa$ of monolayer InSe, In$_2$SSe and In$_2$SeTe. (b) The corresponding room-temperature $\kappa$. (c) The temperature dependence of contribution of $\kappa$ from different branches in monolayer In$_2$SSe. (d) The sample size $L$ dependence of the $\kappa$ contributed by different branches in monolayer In$_2$SSe.
Janus structures experiences a tensile or compressive strain, compared to its perfect binary analogs [13]. For example, the bonding length of $d_{\text{In-Se}}$ on Se side and $d_{\text{In-Te}}$ on Te side of monolayer In$_2$SeTe are larger and smaller than that of monolayer InSe and InTe ($d_{\text{In-Te}} = 2.890$ Å in our calculation), respectively (see table 1).

Monolayer InSe and its Janus derivatives are semiconductors (see figure 6). Figure 1(c) displays the phonon dispersive relations and indicates their structural stability. The acoustic branches consist of the longitudinal (LA) branch, transverse (TA) branch and flexural branch (ZA) branch. Acoustic branches intersect with optical branches. The phonon modes at the $\Gamma$ point of monolayer InSe are decomposed into $\Gamma_D^{3v} = 2A_{\Gamma}^{\prime} \oplus 2A_{\Gamma}^{\prime\prime} \oplus 2E^\prime \oplus 2E^\prime\prime$ according to the analysis of group theory. $A_{\Gamma}^{\prime}$, $E^\prime$ and $E^\prime\prime$ modes are Raman active. The calculated Raman spectra exhibits three prominent peaks (see figure 2(a)). Two Raman peaks at frequencies $\omega$ of 109 and 223 cm$^{-1}$ arise from two out-of-plane $A_{\Gamma}^{\prime}$ modes and a smaller peak at $\omega = 178$ cm$^{-1}$ originates from in-plane $E^\prime$ mode, consistent with the previous work [9] (see table 1).

On the other side, the $\Gamma$-point phonon modes of Janus monolayer In$_2$SSe and In$_2$SeTe are decomposed into $\Gamma_{C_{\mu}} = 4A_{\Gamma} \oplus 4E$. All the optical modes are Raman active. There are two prominent $A_{\Gamma}^{\prime}$ peaks at the low- and high-frequency zone (see figures 2(b) and (c)). Its eigenvectors are similar to that of two $A_{\Gamma}^{\prime}$ peak of monolayer InSe. In addition, a conspicuous $A_{\Gamma}^{\prime}$ mode, which is absent in monolayer InSe, appears in the intermediate frequency zone (labeled by slant lines). The phonon mode with the similar eigen-displacements (see inset of figure 2(c)) in monolayer InSe belongs to $A_{\Gamma}^{\prime\prime}$ representation. The change of polarizability with corresponding normal coordinate at the equilibrium configuration is zero due to the mirror symmetry, so the $A_{\Gamma}^{\prime\prime}$ mode is Raman inactive in monolayer InSe. Therefore, the $A_{\Gamma}^{\prime}$ Raman peak due to the breaking of mirror symmetry can serve as the characterisic signal of 2D In$_2$SSe and In$_2$SeTe during its synthesis.

3.2. The long-range force constant

The potential energy of crystal can be expanded as the Taylor series in the atomic displacement

$$V = V_0 + \frac{1}{2!} \sum_{\alpha,\beta} \Phi_{\alpha,\beta} u^\alpha_i u^\beta_j + \frac{1}{3!} \sum_{\alpha,\beta,\gamma} \Phi_{\alpha,\beta,\gamma} u^\alpha_i u^\beta_j u^\gamma_k + \ldots,$$

(4)

where $u^\alpha_i$ is the $\alpha$ ($\alpha = x, y, z$) component of the displacement of atom $i$ from the equilibrium position $r^\alpha_i$. The $\Phi_{\alpha,\beta}$ and $\Phi_{\alpha,\beta,\gamma}$ are the 2nd and 3rd IFCs, respectively.
We found that both 2nd IFCs and 3rd IFCs in group-III chalcogenides are long-range. For the 2nd IFCs, we adopted the absolute and normalized trace of 2nd IFCs tensor which is defined as \[ |\text{Tr}(2\text{nd IFCs})| = \frac{\sum_{\alpha} \Phi^{\alpha,0}_0}{\sum_{\alpha} \Phi^{\alpha,0}_0} \] (5)

where the \( \Phi^{\alpha,0}_0 \) represents the 2nd IFCs with respect to the \( \alpha \)-directional displacement of the origin atom (\( u^0_\alpha \)) and the neighboring \( \alpha \)-th atom (\( u^\alpha_\alpha \)). When \( i = 0 \), it represents the self-interaction 2nd IFCs. The distance between two atoms is \( d = |r^0_\alpha - r^0_0| \). Figure 3(a) shows the interatomic distance \( d \) dependence of \( |\text{Tr}(2\text{nd IFCs})| \) of monolayer InSe. The 2nd IFCs is the largest for the nearest neighboring (\( d = 2.689 \) Å) In–Se pairs and decreases quickly from \( d = 4.093 \) Å, which is the lattice constant and represents distance of the Se–Se or In–In pairs between the neighbor cells. However, the non-negligible 2nd IFCs can exist at a distance as long as 10 Å which is corresponding to the 16th nearest neighbors (N.N.).

**Figure 6.** (a) The HSE06 band structure of monolayer (a) InSe, (b) In$_2$SSe and (c) In$_2$SeTe. The spatial distribution of electronic state at the conduction band minimum of monolayer (d) InSe, (e) In$_2$SSe and (f) In$_2$SeTe.

**Table 2.** The electron effective mass \( m^*/m_0 \) along \( x \) and \( y \) axis, 2D elastic module \( C_{2D} (J \text{m}^{-2}) \), deformation potential constant \( E_1 \) (eV) and room-temperature \( \mu \) (cm$^2$/V/s) of monolayer InSe and its Janus derivatives along the \( x \) axis. The previous result of [44] is listed for a comparison.

| Type     | \( m^*/m_0 \) | \( m^*/m_0 \) | \( C_{2D} \) | \( |E_1| \)  | \( \mu \)  |
|----------|---------------|---------------|-------------|-------------|------------|
| InSe     | 0.181         | 0.182         | 49.21       | 5.815       | 943.3      |
| [0.177]  | [0.182]       |               |             |             |            |
| In$_2$SSe| 0.211         | 0.212         | 52.69       | 5.331       | 884.8      |
| In$_2$SeTe| 0.175        | 0.176         | 44.22       | 5.075       | 1190.6     |

The previous result of [44] is listed for a comparison.
Besides InSe, long-range 2nd IFCs were also found in monolayer In$_2$SSe and In$_2$SeTe.

For each 3rd IFC, we extracted its maximum component $(\Phi_{ijkl}^{x,y})_{\text{max}}$ and the maximum bonding length $(d_{\text{max}})$ among three atoms ($i,j,k$) [35]. The distribution of $(\Phi_{ijkl}^{x,y})_{\text{max}}$ with respect to $d_{\text{max}}$ in monolayer InSe is shown in figure 3(b). We identified that the triplets $\Phi_{\text{Sc,Se,Se}}^{x,y}$ are particularly large at $d_{\text{max}} = 4.09$ Å and $8.19$ Å which are corresponding to the 2nd and 11th N.N., respectively. They describe the motion of Se atoms at the same atomic plane along the $y$ (zigzag) direction. Actually, the large triplet $\Phi_{\text{Sc,Se,Se}}^{x,y}$ only involves two Se atoms, indicating that it represents the derivatives of self-interaction 2nd IFCs $\Phi_{\text{Sc,Se}}^{x,y}$ with respect to displacements of another Se atom.

The population of the 3rd IFCs as the function of $d_{\text{max}}$ is shown in the inset of figure 3(b). The number of 3rd IFCs at $d_{\text{max}} = 8.19$ Å (11th N.N.) is much larger than that of $d_{\text{max}} = 4.09$ Å ($2^{\text{nd}}$ N.N.) at which the largest 3rd IFCs located, leading to strong anharmonic scattering when we include the 3rd IFCs with $d_{\text{max}}$ beyond that distance. Meantime, we found that though the triplets $(\Phi_{ijkl}^{x,y})_{\text{max}}$ at $d_{\text{max}} = 9.46$ Å (14th N.N.) are small (see figure 3(b)), it has a particular large population. This will lead to a visible influence on the thermal transport. Additionally, the existence of 3rd IFCs with large value or large population at the distance beyond the 11th N.N. had also been found in other monolayers. The test indicated that $\kappa$ will not reach convergence until one considered the 3rd IFCs as long as 16th N.N. (see figure A1(b)), indicating that the long-range 2nd and 3rd IFCs are a common feature in 2D group-III chalcogenides.

Monolayer InSe, In$_2$SSe and In$_2$SeTe have a large Born effective charge and weak dielectric screening (see table A1). Hence, the aforementioned long-range IFCs can be attributed to the long-range dipole-dipole interaction (DDI), which has been proved in monolayer InSe [36]. Meanwhile, the long-range DDI produces the individual optical branch around the 100 cm$^{-1}$ that is completely separated from other phonon modes by frequency gaps (see figure 1(c)). This feature offers the potential applications of these Janus monolayers in phononic lasers and nanomechanical oscillators for mode control [36].

### 3.3. The lattice conductivity of Janus monolayers based on InSe

We first calculated the intrinsic $\kappa$ of monolayer InSe and its Janus derivatives without boundary scattering. The corresponding phonon lifetime was estimated by $(\tau_{q_s}^{-1}) = (\tau_{q_{s,\text{iso}}}^{-1}) + (\tau_{q_{s,\text{an}}}^{-1})$. The intrinsic $\kappa$ in the temperature range of $T = 150–750$ K are shown in figure 4(a). At $T = 300$ K, the $\kappa$ of monolayer InSe and In$_2$SSe are estimated as 44.6 and 46.9 W (m · K)$^{-1}$, respectively (see figure 4(b)), which are comparable to that of semiconductor GaAs (45 W (mK)$^{-1}$) [37], few-layer MoS$_2$ (40–50 W (mK)$^{-1}$) [38] and few-layer black phosphorus (10–20 W (mK)$^{-1}$) [39]. The result of monolayer InSe agrees with previous Pandey’s work [21]. In contrast, monolayer In$_2$SeTe with heavier atomic mass has a smaller $\kappa$ of 29.8 W (m · K)$^{-1}$.

Besides the acoustic branches, low-frequency optical (low-$\omega$) branches (labeled by blue in figure 1(c)) also make a non-negligible contribution to the $\kappa$ of monolayer InSe and its Janus derivatives (see figure 4(c)). The large phonon group velocity of low-$\omega$ branches is the main reason. In contrast, only the acoustic phonons are considered in the heat transport of conventional semiconductors. Meanwhile, the heat is mainly carried by phonons with frequencies less than 100 cm$^{-2}$, since the group velocities of higher optical modes are small.

Considering the limited sample size ($L$), the size effects of $\kappa$ and its contribution from different branches are discussed in the range of diffusive thermal transport. The representative result of monolayer In$_2$SSe is displayed in figure 4(d). The results of monolayer InSe and In$_2$SeTe are similar. At room temperature, the phonon mean free path (MFP) of monolayer In$_2$SSe is about 10$^2$ nm, which is mainly determined by the MFP of LA andZA phonons. The $\kappa$ can be decreased by 90% as the $L$ decrease down to 10nm. That indicates that nanostructuring might be an effective method to reduce the $\kappa$ of 2D group-III chalcogenide for the thermoelectric applications.

To give a more deep understanding of heat transport in monolayer InSe and its Janus structures, we analyzed every term in determining the $\kappa$ in equation 1. The mode group velocity $v_{q_s}$ in the frequency zone of 0–100 cm$^{-1}$ are displayed in figure 5(a). The overall $\kappa_{q_s}$ of monolayer In$_2$SSe and In$_2$SeTe are higher and lower than that of monolayer InSe, due to the smaller and larger atomic mass than that of InSe, respectively. Figure 5(b) shows the mode relaxation time $\tau_{q_s}$ at room temperature. The $\tau_{q_s}$ of monolayer InSe is slightly larger than that of In$_2$SSe at frequency zone of $\omega < 4$ cm$^{-1}$ and 30 cm$^{-1} < \omega < 50$ cm$^{-1}$ (see figures 5(b) and A2). Meanwhile, the overall $\tau_{q_s}$ of monolayer In$_2$SeTe is smaller than that of monolayer InSe and In$_2$SSe. It was found that the anharmonic scattering (1/\tau_{q_s}^{an}) dominates the total phonon lifetime. In contrast, the isotopic impurities scattering, which is inversely proportional to atomic mass [25], has a small influence on $\kappa$, e.g. the room-temperature $\kappa$ of monolayer In$_2$SSe will increase only by 1.26% if we excluded the 1/\tau_{q_s}^{iso} from the total scattering rate 1/\tau_{q_s}.

The anharmonic 1/\tau_{q_s}^{an} depends on the square of Grüneisen parameter $\gamma_{q_s}^2$, and weighted phase space $W_{q_s}$ [25]. The former and latter one represent the anharmonicity strength and the number of channels available for a phonon to get scattered, respectively [25, 40]. The increase of $\gamma_{q_s}^2$ or $W_{q_s}$ has a decreased effect on $\tau_{q_s}$.

The overall $\gamma_{q_s}^2$ of monolayer InSe is smaller than that of its Janus derivatives (see figure 5(c)). The breaking of mirror
symmetry in Janus structures leads to the asymmetric chemical bonding and charge density distribution along the out-of-plane direction (see figures 6(d)–(f)), which will increase the bond anharmonicity [41, 42]. Figure 5(d) shows that the sequential of $W_{\text{qs}}$ from high to low is $W(\text{InSe}) > W(\text{InSe}) > W(\text{InSe})$. $W_{\text{qs}}$ of monolayer InSe, which has the mirror symmetry, is between that of monolayer In$_2$S$_2$Se and In$_2$SeTe. The phase space for anharmonic scattering decreases as the overall phonon frequency scale increases (see figure 1(c)), consistent with the observation in many bulk materials [43]. The integral effect of $\gamma_{\text{qs}}$, $\gamma_{\text{sq}}$, and $\gamma_{\text{q2}}$ lead to the aforementioned higher $\tau_{\text{qs}}$, $\tau_{\text{sq}}$, and $\tau_{\text{q2}}$ of monolayer InSe than that of monolayer In$_2$S$_2$Se at the frequency zone of $\omega < 4 \text{ cm}^{-1}$ and $30 \text{ cm}^{-1} \leq \omega < 50 \text{ cm}^{-1}$. As a result, monolayer In$_2$S$_2$Se, with smaller atomic mass, larger $v_{\text{qs}}$, but a smaller $\tau_{\text{q2}}$, has a slightly larger $\kappa$ than that of monolayer InSe. On the other side, the larger atomic mass, smaller $v_{\text{qs}}$, larger $\gamma_{\text{qs}}$, and $W_{\text{qs}}$ lead to the smaller $\tau_{\text{qs}}$, $\tau_{\text{sq}}$, and $\kappa$ in monolayer In$_2$SeTe than InSe.

3.4. The electron mobility of Janus monolayers based on InSe

Besides the heat transport, we also calculated the carrier mobility $\mu$ based on the deformation potential theory [33]. The band structure indicates that electron effective mass is smaller than hole one which has Mexican-hat valence band edge (see figures 6(a)–(c)), consistent with previous work [14]. The parameters involved in electron mobility are calculated (see figure A3) and shown in table 2. The order of $C_{2D}$ from large to small is $C_{2D}(\text{InSe}) > C_{2D}(\text{InSe}) > C_{2D}(\text{InSe})$, consistent with the order of $d_{\text{ac}}$ and $v_{\text{qs}}$ (see table 1 and figure 5(a)).

The electron effective mass $m^*$ of monolayer In$_2$S$_2$Se and In$_2$SeTe are larger and smaller than that of monolayer InSe, respectively. The orbital analysis indicates that the electronic states at conduction band minimum (CBM) of monolayer InSe are dominated by In-5s orbital with symmetry spatial distribution (see figure 6(d)). In contrast, the electronic states at CBM of monolayer In$_2$S$_2$Se and In$_2$SeTe are mainly composed of 5x orbital of In atom on the S and Se side, respectively (see figures 6(e) and (f)). Taking monolayer In$_2$SeTe as an example, the Se side experiences aforementioned tensile strain compared to that of monolayer InSe. Actually, if we applied a biaxial tensile strain of 2% to the lattice of monolayer InSe, the $d_{\text{InSe}}$ becomes close to that of monolayer In$_2$S$_2$Se. Meanwhile, the $m^*$ changes from 0.188 $m_0$ to 0.174 $m_0$, consistent with the $m^*$ of In$_2$S$_2$Te (table 2). Similarly, $d_{\text{InSe}}$ of monolayer In$_2$S$_2$Se is about 2% larger than that of monolayer InS. The calculated $m^*$ of monolayer InS under 2% tensile strain is 0.227 $m_0$, which can also explain the larger $m^*$ of monolayer In$_2$S$_2$Se than that of monolayer InSe.

The $\mu$ of monolayer In$_2$S$_2$Se is smaller than that of monolayer InS, due to large $m^*$. Monolayer In$_2$SeTe has a higher $\mu$ than that of monolayer InSe, which can be attributed to smaller $m^*$ and $E_1$. Moreover, monolayer In$_2$SeTe has a direct band gap of 1.8 eV and thereby is more favorable for optoelectronics application than monolayer InSe and In$_2$S$_2$Se which have indirect band gaps (see figures 6(a)–(c)).

4. Conclusion

Based on the first-principles calculations, we investigated the thermal and electronic transport properties of monolayer InSe, In$_2$S$_2$Se and In$_2$SeTe. One side of monolayer In$_2$S$_2$Se and In$_2$SeTe undergo a tensile or compressive strain. A distinguishable $A_1$ peak in the Raman spectra of monolayer In$_2$S$_2$Se and In$_2$SeTe was identified due to the breaking of mirror symmetry. The existence of long-range harmonic and anharmonic interactions is a common feature in 2D group-III chalcogenides, which plays an important role in its heat transport. The
room-temperature $\kappa$ of monolayer InSe, In$_2$Se and In$_2$SeTe are 44.6, 46.9 and 29.8 W (m·K)$^{-1}$, respectively. Though with smaller atomic mass, the $\kappa$ of monolayer In$_2$Se is comparable to that of monolayer InSe, due to the competition effect between phonon group velocity and lifetime. Moreover, the electron mobility of monolayer In$_2$SeTe is higher than that of monolayer InSe, due to the smaller electron effective mass and deformation potential. Our work facilitates the understanding of the heat and carrier transport properties of 2D Janus group-III chalcogenides and offers the theoretical support to the corresponding designment of electronic device in future.

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Appendix

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References

Table A1. The Born effective charge $Z'$ (in unit of elementary charge $e$), dielectric constant $\varepsilon$ of monolayer InSe, In$_2$S$_2$Se and In$_2$Te.

| Atom     | $Z_{xx}'$ | $Z_{zz}'$ | $\varepsilon_{xx}$ | $\varepsilon_{zz}$ |
|----------|-----------|-----------|---------------------|---------------------|
| InSe     | 2.49      | 0.21      | 3.87                | 1.56                |
| Se       | -2.49     | -0.21     | -                    | -                    |
| In$_2$S$_2$Se | 2.45      | 0.27      | 3.62                | 1.54                |
| In$_2$Te | 2.57      | 0.35      | -                   | -                    |
| Se       | -2.44     | -0.26     | -                   | -                    |
| S        | -2.58     | -0.34     | -                   | -                    |
| In$_2$SeTe | 2.59      | 0.27      | 4.36                | 1.61                |
| In$_2$Te | 2.34      | 0.23      | -                   | -                    |
| Se       | -2.62     | -0.26     | -                   | -                    |
| Te       | -2.31     | -0.23     | -                   | -                    |

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