INTRODUCTION

Thermoelectric technology is considered as a promising conversion strategy between electricity and heat, and it has wide applications, such as waste-heat recovery, self-powered electronic device, radioisotope thermoelectric generator, thermoelectric refrigerator, etc.1–3 The thermoelectric conversion efficiency is mainly determined by the performance in materials, which closely depends on its dimensionless figure of merit (ZT), \( ZT = \frac{S^2 \sigma T}{\kappa_{\text{tot}}} \), where \( S \), \( \sigma \), \( \kappa_{\text{tot}} \), and \( T \) represent Seebeck coefficient, electrical conductivity, total thermal conductivity, and absolute temperature, respectively.4–6 Obviously, high-performance thermoelectric materials require large absolute Seebeck coefficient, high electrical conductivity, and low thermal conductivity simultaneously. However, the thermoelectric performance is limited by these strongly coupled relationships between thermoelectric parameters. To develop high-performance thermoelectric materials, researchers can decouple these thermoelectric parameters in a given material by using band structure engineering7–9 and nanostructure designing,10–12 or exploit new highly effective thermoelectric materials with intrinsic low conductivity and superior electrical transport properties.13–15

Recently, indium selenide (InSe) has gained wide attention due to its exceptional plasticity as a semiconductor, making it promising to be applied in deformable or flexible electronics.16,17 More importantly, InSe compound presents wide bandgap \( (E_g \sim 1.18 \text{~V}) \) and low-symmetry layered crystal structure (space group: \( \text{P6}_3/mmc \)), which are consistently complete with the selection rules to seek highly effective thermoelectrics.21 Therefore, semiconductor InSe is considered as a potential thermoelectric and might have promising applications in flexible and wearable thermoelectric devices. However, previous results found that InSe owns very low carrier concentration around \( \sim 3.2 \times 10^{13} \text{~cm}^{-3} \) at room temperature that largely restricts its thermoelectric performance.22,23 Thus, both anion (Si and Sn) doping22–25 and S alloying in InSe, the carrier concentration can be further enhanced from \( \sim 3.2 \times 10^{13} \text{~cm}^{-3} \) in InSe to \( \sim 4.8 \times 10^{15} \text{~cm}^{-3} \) in InSe\(_{0.97}S_{0.03} \) at 300 K, because S \((\chi_p \sim 2.58)\) with larger Pauling electronegativity than Se \((\chi_p \sim 2.55)\) can induce more In\(^{3+}\) state to increase carrier concentration in matrix. This boosted dynamic carrier transport property benefits an obviously enhanced power factor. Additionally, InSe compound presents intrinsically low thermal conductivity \( \sim 1.6 \text{~W m}^{-1} \text{~K}^{-1} \) at 300 K due to low-symmetry crystal structure and strong anharmonicity. This work indicates that the special dynamic carrier transport property and intrinsically low thermal conductivity in InSe make it as a worthwhile-expecting thermoelectric material.

KEYWORDS

amphoteric indium, carrier concentration, InSe thermoelectric, lattice thermal conductivity
RESULTS AND DISCUSSION

Crystal structure and electronic band structure in InSe

The crystal structure of InSe is illustrated in Figure 1A–C. It is crystalized in hexagonal system with a space group of P63/mmc, and presents a low-symmetry layered crystal structure along c axis. The adjacent layers are connected by weak van der Waals forces, which results in strong anharmonicity and intrinsically low lattice thermal conductivity in InSe, as discussed below. The XRD patterns of InSe in Figure 1D are indexed into pure phase with the lattice parameters of $a = b = 4.004$ Å and $c = 16.682$ Å, which is consistent with the theoretical value of $a = b = 4.005$ Å and $c = 16.640$ Å. In Figure 1E, the optical absorption measurement shows a large bandgap ($E_g$) of $\sim 1.18$ eV in InSe compound. Noticeably, these intrinsic characters of wide bandgap, low-symmetry and layered crystal structure in InSe are well consistent with the rules to select new highly effective thermoelectric materials. Therefore, the thermoelectric performance in InSe is worthy to being systematically investigated.

Figure 2A presents the electronic band structure of InSe obtained by density functional theory (DFT) calculation. The theoretical calculation shows that InSe is a direct bandgap semiconductor with a large bandgap ($E_g = \sim 0.73$ eV). It is worthy to note that the theoretical bandgap is calculated at 0 K without considering the effect of temperature, which is smaller than the optical measured result ($E_g \sim 1.18$ eV) at room temperature. InSe can be regarded as multiple band model with band offsets of $\sim 0.4$ and $\sim 0.3$ eV in conduction band and valence band, respectively. Notably, the conduction band owns much sharper shape than valence band, which is consistent with the density of states (DOS) in Figure 2B. Different band shapes in conduction and valence bands can lead...
to a lower carrier effective mass in $n$-type InSe than its $p$-type counterpart. Relatively low carrier effective mass in $n$-type InSe is capable to achieve high carrier mobility and electrical conductivity, while high Seebeck coefficient can be obtained in $p$-type InSe due to large carrier effective mass. The DFT calculation unveils a favorable electronic band structure in InSe, and signifies potential thermoelectric performance in both $n$-type and $p$-type InSe.

**Dynamic carrier transport property in InSe**

InSe is a sister compound of $\text{In}_2\text{Se}_3$, and the difference between these two compounds is that indium element presents valence fluctuation ($\text{In}^+$ and $\text{In}^{3+}$) in InSe, while $\text{In}_2\text{Se}_3$ contains full $\text{In}^{3+}$ valence state.\cite{30,31} To shed light on the effect of amphoteric indium, the carrier transport properties between InSe and $\text{In}_2\text{Se}_3$ are compared in Figure 3. All these carrier transport properties compared here are from in-plane direction, because InSe and $\text{In}_2\text{Se}_3$ possess layered crystal structure and high anisotropy. It is shown that both InSe and $\text{In}_2\text{Se}_3$ exhibit intrinsic $n$-type transport behaviors. Differently, $\text{In}_2\text{Se}_3$ exhibits a much higher electrical conductivity and lower Seebeck coefficient than InSe, shown in Figure 3A,B, because of its large carrier concentration around $1.9 \times 10^{17}$ cm$^{-3}$ in $\text{In}_2\text{Se}_3$ and low carrier concentration around $3.2 \times 10^{13}$ cm$^{-3}$ in InSe at room temperature. The different carrier concentrations in these two compounds
leads to great discrepancy in power factor, and In$_2$Se$_3$ shows higher power factor than InSe in the whole temperature range, as shown in Figure 3C. Notably, the turning point at around 470 K in electrical transport properties of In$_2$Se$_3$ originates from its phase transition from α-In$_2$Se$_3$ to β-In$_2$Se$_3$. Additionally, the temperature-dependent Hall measurement in Figure 3D proves that In$_2$Se$_3$ has orders magnitude higher carrier concentration than that in InSe in the entire temperature range, which might arise from more In$^{3+}$ valence state in In$_2$Se$_3$. More importantly, the Hall measurement uncloses that InSe has a dynamic carrier transport property and the carrier concentration continuously increases with rising temperature, from $3.2 \times 10^{13}$ cm$^{-3}$ at 300 K to $6.2 \times 10^{16}$ cm$^{-3}$ at 723 K. This particularly dynamic carrier concentration in InSe is attributed to the amphoteric indium (In$^+$ and In$^{3+}$), which has been observed in In-doped PbTe.\textsuperscript{32,33} With thermal activation, the In$^+$ state in InSe will transmit into In$^{3+}$ state and simultaneously release two free carriers into matrix, thus causing increasing temperature-dependent carrier concentration in the entire temperature range. This dynamic carrier transport property in InSe motivates us to enhance its carrier concentration by manipulating valence state of indium. Compared with the electronegativity of Se atom ($\chi_\text{P} \sim 2.55$), S atom possesses a larger electronegativity of $\chi_\text{P} \sim 2.58$, indicating that S atom is more capable to attract electrons and can induce more In$^{3+}$ valence state. Therefore, S alloying is conducted in InSe, and the carrier concentration in InSe$_{0.97}$S$_{0.03}$ is significantly enhanced as expected (shown in Figure 3D). Compared with the carrier concentration in InSe, InSe$_{0.97}$S$_{0.03}$ obtains an obvious enhancement in the whole temperature range, and the maximum carrier concentration increases from $\sim 6.2 \times 10^{16}$ cm$^{-3}$ in InSe to $\sim 1.0 \times 10^{17}$ cm$^{-3}$ in InSe$_{0.97}$S$_{0.03}$ at 723 K. This boosted dynamic carrier concentration is expected to further optimize its thermoelectric properties in InSe matrix.

**Electrical transport properties in S-alloyed InSe**

A series of S-alloyed InSe samples (InSe$_{1-x}$S$_x$, $x = 0, 0.01, 0.03, 0.05$) are prepared and their phases are first identified by X-ray diffraction (XRD). From the XRD patterns in Figure 4A, all the S-alloyed samples can be indexed into single phase with hexagonal structure (P6$_3$/mmc). The calculated lattice parameter in Figure 4B shows a decreasing tendency with increasing S content along $a$ and $c$ axes, proving that S can fully alloy in InSe within 0.05 S content.

Figure 5 presents the electrical transport properties of InSe$_{1-x}$S$_x$ ($x = 0, 0.01, 0.03, 0.05$). The electrical conductivity in S-alloyed sample obtains an obvious enhancement, especially in InSe$_{0.97}$S$_{0.03}$ sample as shown in Figure 5A. The Seebeck coefficient in S-alloyed InSe is still negative value, indicating an electron-dominated $n$-type transport property in Figure 5B. Correspondingly, the absolute value of Seebeck coefficient in S-alloyed InSe decreases, which arises from the enhanced carrier concentration, as shown in Figure 5C. The room-temperature carrier concentration is largely improved after S alloying, from $3.2 \times 10^{13}$ cm$^{-3}$ in InSe to $2.7 \times 10^{16}$ cm$^{-3}$ in InSe$_{0.99}$S$_{0.01}$, and then slightly decreases to $2.8 \times 10^{15}$ cm$^{-3}$ in InSe$_{0.95}$S$_{0.05}$. The improved carrier concentration in S-alloyed InSe is originated from the large electronegativity of S that can attract two electrons from In$^{3+}$ state and then release more free carriers into matrix, as discussed above. Notably, large electronegativity in S atoms is double-edged sword, which will inversely capture the free carrier with high S content and finally suppress the carrier concentration with increasing S alloying content in InSe$_{1-x}$S$_x$ samples. Therefore, it is of great importance to optimize the S content in order to enhance its carrier concentration in InSe. As shown in Figure 5C, InSe exhibits very low carrier mobility, $\sim 1.9$ cm$^2$ V$^{-1}$ s$^{-1}$ at room temperature, and the carrier mobility in S-alloyed InSe continuously decreases due to its enhanced carrier concentration. As a result, the power factor achieves an improvement due to increased concentration in S-alloyed InSe, from $\sim 0.4 \mu$W$^{-1}$ cm$^{-1}$ K$^{-2}$ in InSe to $\sim 0.6 \mu$W$^{-1}$ cm$^{-1}$ K$^{-2}$ in InSe$_{0.97}$S$_{0.03}$.

**Thermal transport properties and ZT value in S-alloyed InSe**

To investigate the intrinsic thermal transport properties in InSe, this work conducted ultrasonic pulse echo measurements. With the measured sound velocity, series elastic properties can be calculated, including Poisson ratio ($\nu_p$), Young’s modulus ($E$), shear modulus ($G$), and Grüneneisen parameter ($\gamma$).\textsuperscript{34} Generally, low Young’s modulus, low shear modulus, and large Grüneneisen parameter are the indicators of low thermal conductivity.\textsuperscript{34–36} The relationships between sound...
Electrical transport properties in InSe$_{1−x}$S$_x$ ($x = 0, 0.01, 0.03, 0.05$): (A) electrical conductivity, (B) Seebeck coefficient, (C) carrier concentration and Hall mobility as a function of S content, and (D) power factor.

Velocity and elastic properties can be expressed:

\[ \nu_a = \left[ \frac{1}{3} \left( \frac{1}{\nu_l^3} + \frac{1}{\nu_s^3} \right) \right]^{-\frac{1}{3}}, \]

\[ \nu_p = \frac{1 - 2 \left( \frac{\nu_s}{\nu_l} \right)^2}{2 - 2 \left( \frac{\nu_s}{\nu_l} \right)^2}, \]

\[ E = \frac{\nu_s^2}{\nu_l^2} \left( 3\nu_s^2 - 4\nu_s \nu_p \right), \]

\[ G = \frac{E}{2(1 + \nu_p)}, \]

\[ \gamma = \frac{2}{3} \left( \frac{1 + \nu_p}{2 - 3\nu_p} \right). \]

where \( \nu_a, \nu_p, \) and \( \rho \) are average sound velocity, Poisson ratio, and density of samples, respectively.

The Debye temperature (\( \Theta_D \)) is estimated as follows:

\[ \Theta_D = \frac{h \nu_s}{k_B} \left( \frac{3N}{4\pi V} \right)^{\frac{1}{3}}, \]

where \( h, k_B, N, \) and \( V \) are the Plank constant, Boltzmann constant, number of atoms in a unit cell, and volume of a unit cell, respectively.

All the calculated elastic parameters are listed in Table 1 and compared with typical mid-temperature thermoelectric materials, PbTe and PbSe. InSe presents a large Poisson ratio \( \nu_p \sim 0.37 \) that indicates a deformable trend under stress. The values of the calculated Young’s modulus and shear modulus of InSe are \( \sim 20.5 \) GPa and \( \sim 7.5 \) GPa, which are lower than those of PbSe (\( E \sim 65.9 \) GPa and \( G \sim 25.6 \) GPa) and PbTe (\( E \sim 53.7 \) GPa and \( G \sim 20.9 \) GPa). Because Young’s modulus and shear modulus are related to the interatomic bonding strength, the low modulus implies a weak van der Waals force between layers in InSe. As known, Grüneisen parameter (\( \gamma \)) is a reflection of lattice vibration anharmonicity. The Grüneisen parameter of InSe is \( \sim 2.28 \), which is much larger than PbSe (\( \sim 1.65 \)) and PbTe (\( \sim 1.45 \)), indicating that InSe processes a strong anharmonicity in crystal lattice. The Debye temperature can be evaluated by using average sound velocity, and InSe has a lower Debye temperature of \( \sim 128 \) K compared with PbSe \( \sim 191 \) K and PbTe \( \sim 163 \) K. These evaluated elastic parameters clearly

**Table 1:** Lattice thermal conductivity, density, sound velocity, elastic parameters, and the Debye temperature of InSe and PbQ (Q = Se, Te).

| Parameters | InSe | PbSe | PbTe |
|------------|------|------|------|
| Lattice thermal conductivity, \( \kappa_L \) (W m$^{-1}$ K$^{-1}$) | 1.59 | 1.65 | 2.00 |
| Density, \( \rho \) (g cm$^{-3}$) | 5.30 | 8.27 | 8.18 |
| Longitudinal sound velocity, \( \nu_l \) (m s$^{-1}$) | 2590 | 3220 | 2900 |
| Shear sound velocity, \( \nu_s \) (m s$^{-1}$) | 1188 | 1760 | 1600 |
| Average sound velocity, \( \nu_a \) (m s$^{-1}$) | 1339 | 1960 | 1780 |
| Poisson ratio, \( \nu_p \) | 0.37 | 0.29 | 0.28 |
| Young’s modulus, \( E \) (GPa) | 20.5 | 65.9 | 53.7 |
| Shear modulus, \( G \) (GPa) | 7.5 | 25.6 | 20.9 |
| Grüneisen parameter, \( \gamma \) | 2.28 | 1.65 | 1.45 |
| Debye temperature, \( \Theta_D \) (K) | 128 | 191 | 163 |

Note: Room-temperature lattice thermal conductivity (\( \kappa_L \)), sample density (\( \rho \)), and sound velocity (\( \nu_l \) and \( \nu_s \)) of InSe are obtained by experimental measurement; elastic parameters are calculated by Equations (1)–(5), and Debye temperature is estimated by Equation (6). The data of PbQ (Q = Se, Te) are based on previous reports.


reveal the intrinsic thermal transport property in InSe and contribute to a comparably low lattice thermal conductivity of $\sim 1.6$ W m$^{-1}$ K$^{-1}$ at room temperature.

To discuss the possibilities of further reducing the thermal conductivity, the phonon mean free path (MFP) of InSe is evaluated with the following expression:

$$\kappa_{\text{lat}} = \frac{1}{3} C_V \nu_m l,$$

where $C_V$ is the heat capacity at constant volume, which can be replaced by $C_V = \rho C_P$ ($\rho$ represents sample density and $C_P$ represents the heat capacity at constant pressure), $l$ is the phonon MFP. The MFP of InSe calculated by Equation (7) is 26.49 Å, which is much larger than the lattice parameters in the unit cell ($a = b = 4.004$ Å and $c = 16.682$ Å). Thus, the lattice thermal conductivity of InSe could be further suppressed by nanostructure engineering. Using the interatomic distance as the minimum phonon MFP, the minimum lattice thermal conductivity ($\kappa_{\text{lat, min}}$) can be expressed as follows according to Cahill’s model:

$$\kappa_{\text{lat, min}} = \frac{1}{2} \left( \frac{\pi}{\delta} \right)^{\frac{1}{3}} k_B V^{-\frac{2}{3}} (\nu_1 + 2\nu_2),$$

where $V$ is the volume of per atom in the unit cell. Value of the minimum $\kappa_{\text{lat}}$ estimated by Equation (8) is $\sim 0.3$ W m$^{-1}$ K$^{-1}$. The evaluated minimum lattice thermal conductivity in InSe is much lower than the measured room temperature value of $\sim 1.6$ W m$^{-1}$ K$^{-1}$, suggesting that there is large space to further reduce its thermal conductivity.

Figure 6 presents the thermal transport properties and final ZT values. The thermal diffusivity, the heat capacity, Lorentz constant, and electronic thermal conductivity are showed in Figures 6A, 6B, 6C, and 6D, respectively. The temperature-dependent thermal conductivities of InSe$_{1-x}$S$_x$ ($x = 0$, 0.01, 0.03, 0.05) samples are showed in Figure 6E. The total thermal conductivity is close to lattice thermal conductivity in S-alloyed InSe because its negligible electronic thermal conductivity ($\kappa_{\text{ele}}$) shown in Figure 6D caused by poor electrical conductivity, following the equation $\kappa_{\text{ele}} = \sigma T$. With the increasing S content, the $\kappa_{\text{lat}}$ decrease is obviously ascribed to more point defects scattering caused by S substitution. The minimum lattice thermal conductivity in S-alloyed InSe can be achieved at $\sim 0.6$ W m$^{-1}$ K$^{-1}$ in InSe$_{0.95}$S$_{0.05}$ at 723 K. Finally, combining the suppressed lattice thermal conductivity and optimized power factor above, the ZT value obtains a distinct improvement, as shown in Figure 6F.
CONCLUSION

In this work, the dynamic carrier transport property and intrinsic low lattice thermal conductivity in InSe are systematically investigated. Its particularly increasing temperature-dependent carrier concentration arises from the amphoteric indium in InSe, and this dynamic carrier transport property can be boosted by introducing S alloying due to large electronegativity. The intrinsic low lattice thermal conductivity in InSe is related to its low-symmetry layered crystal structure and strong anharmonicity of lattice vibration, which is unclosed by sound velocity. This work suggests that InSe might be a promising thermoelectric material and its properties can be further enhanced with well-optimized carrier concentration. Some potential approaches to further improve the carrier concentration in n-type InSe include halogen doping (Cl, Br, or I) in Se site, high-valent cation doping (Ti, V, or Ni) in In site, and interstitial atoms doping (Cu or Ag). Furthermore, excellent thermoelectric performance is expected to be achieved in InSe crystal owning to high carrier mobility, as realized in SnSe and SnSe crystals. Last but not least, this interesting dynamic carrier engineering proved in InSe can be regarded as a promising strategy to be extended to other thermoelectric systems.

EXPERIMENTAL SECTION

The samples in this work with nominal composition of InSe_{1-x}S_x (x = 0, 0.01, 0.03, 0.05) were synthesized by the melting reaction. Stoichiometric amounts of In (purity, 99.99%), Se (purity, 99.99%), and S (purity, 99.99%) powders were weighed and mixed in a vacuum-sealed quartz tube, heated to 1023 K in 12 h, kept at 1023 K for 10 h, and followed by furnace cooling to room temperature. The obtained ingots were ground into powders using an agate mortar and then densified by spark plasma sintering (SPS) at 773 K for 5 min under 50 MPa, resulting in polycrystalline samples with $\phi$ 12.7 $\times$ 12 mm dimensions.

The phase of sample was characterized by X-ray diffraction (D/MAX2200pc) with Cu Kα radiation ($\lambda$ = 0.15418 nm). The samples obtained by SPS were cut and polished into 3 $\times$ 3 $\times$ 8 mm bulks for measurements of Seebeck coefficient and electrical resistivity using Cryoall AGGREGATE measurement system at 300–723 K in the helium atmosphere. The carrier concentration ($n_{H}$) was calculated by $n_{H} = 1/(eR_{H})$, where $e$ denotes electron charge and $R_{H}$ denotes the Hall coefficient measured by Lake Shore 8400 series. The thermal conductivity of the samples was determined by $\kappa_{tot} = D\rho C_{p}$. Thermal diffusivities ($D$) was measured by Netzsch LFA457 at 300–723 K using the pieces cut and polished into 8 $\times$ 8 $\times$ 1.2 mm. The density ($\rho$) of samples was obtained by the ratio of mass to volume, and specific heat capacity ($C_{p}$) was estimated using the Debye model with room-temperature value of 0.25 J g$^{-1}$ K$^{-1}$. The optical bandgap was obtained by Shimadzu UV-3600 Plus UV–VIS NIRS spectrophotometer. The sound velocity of InSe was measured by ultrasonic pulse echo measurements at room temperature, and a series of mechanical parameters were calculated. The above measurements were conducted in a direction parallel to the pressure to obtain more excellent thermoelectric properties of polycrystal samples.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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