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A Revisit to High Thermoelectric Performance of Single-layer MoS₂

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Both electron and phonon transport properties of single layer MoS₂ (SLMoS₂) are studied. Based on first-principles calculations, the electrical conductivity of SLMoS₂ is calculated by Boltzmann equations. The thermal conductivity of SLMoS₂ is calculated to be as high as 116.8 Wm⁻¹K⁻¹ by equilibrium molecular dynamics simulations. The predicted value of ZT is as high as 0.11 at 500 K. As the thermal conductivity could be reduced largely by phonon engineering, there should be a high possibility to enhance ZT in the SLMoS₂-based materials.

Thermoelectric materials are essential for converting waste heat to electricity and solid-state cooling, which have attracted much attention recently^{1–10}. The dimensionless figure of merit (ZT) is utilized to evaluate the efficiency of the thermoelectric conversion, defined as: $ZT = S^2\sigma T/\kappa$, where S is the Seebeck coefficient, σ is the electrical conductivity, T is the absolute temperature, and κ is the total thermal conductivity. The κ is composed of electrons' contribution (κ_e) and phonons' contribution (κ_p). The ZT value for most commercial materials are around one, which is far below the critical value of three that is comparable with the traditional energy conversion in efficiency³. In the past two decades, nano-materials and nano-structured materials are expected to have excellent energy conversion efficiency due to the higher power factor ($S^2\sigma$)^{11,12} and lower κ_p ^{13–15}, which are also known as the electron-crystal and phonon-glass.

The graphene, as the first two dimensional material, has extraordinary electronic property as well as super high thermal conductivity¹⁶. However, the pristine graphene, a semi-metal, has zero band gap and very small S ¹⁷. Different from graphene, single layer MoS₂ (SLMoS₂) is a semiconductor and has a direct band-gap¹⁸, which enables its wide applications in electronic and optical devices, such as field effect transistor¹⁹.

Recently, some works have studied the electronic and phononic properties of SLMoS₂. Eugene *et al.* have calculated the electronic structure of SLMoS₂ which is compared with that of bulk MoS₂²⁰, and revealed the transition mechanism from the direct band gap of SLMoS₂ to the indirect band gap of bulk MoS₂. Emilio *et al.* have shown that, after applying compressive or tensile bi-axial strain, the electronic structure of SLMoS₂ transitions from semiconductor to metal²¹. Li *et al.* calculated the intrinsic electrical transport and electron-phonon interaction properties of SLMoS₂²². Moreover, the thermoelectric potential of SLMoS₂ has been explored and a maximum ZT , at room temperature, is obtained as 0.5 by Huang *et al.*²³ using the ballistic model. The scatterings of electrons are not considered in their ballistic model, which should have led to an over-estimation of ZT . Fu *et al.* studied SLMoS₂ ribbons and calculated the ZT value to be up to 3.4²⁴. Besides theoretical predictions, Wu *et al.* has experimentally reported a value of S as 30 mV/K for SLMoS₂²⁵, which indicates an appealing potential for thermoelectric applications.

Besides electron properties, some works focused on the phonon properties of SLMoS₂. The SLMoS₂ nanoribbon has a low thermal conductivity due to the size effect. Jiang *et al.* claimed that κ_p of SLMoS₂ nanoribbon was around 5 Wm⁻¹K⁻¹ at room temperature by molecular dynamics (MD) simulations²⁶. Zhang *et al.* reported three results for SLMoS₂ nanoribbons which were 1.35 Wm⁻¹K⁻¹ by equilibrium molecular dynamics (EMD)²⁷, 23.2 Wm⁻¹K⁻¹ by non-equilibrium Green's function²⁸, and 26.2 Wm⁻¹K⁻¹ by Boltzmann transport equation²⁹. However, there are also reports on the thermal conductivities for MoS₂ with higher values. Li *et al.* predicts the κ as 83 Wm⁻¹K⁻¹ from *ab initio* calculations³⁰. With high-quality sample, the κ of suspended few layers MoS₂ has been measured as 52 Wm⁻¹K⁻¹³¹ and 35 Wm⁻¹K⁻¹³². Liu *et al.* claimed that the basal-plane thermal conductivity

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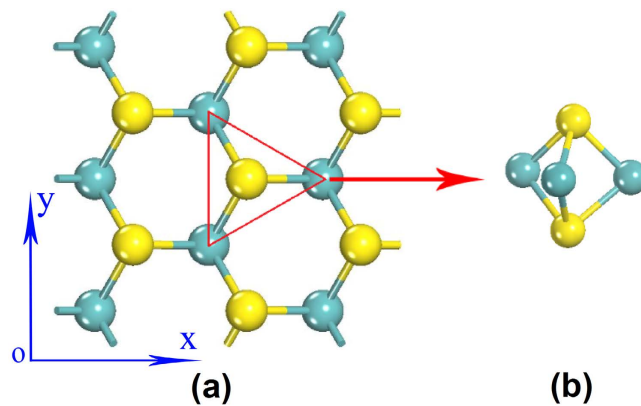


Figure 1. The structure of single layer MoS₂. (a) The top view, a hexagonal lattice structure. (b) The side view of the inset triangle. Each sulfur atom has three molybdenum atoms as its first nearest neighbor atom. Each molybdenum atom has six sulfur atoms as its first nearest neighbor atom.

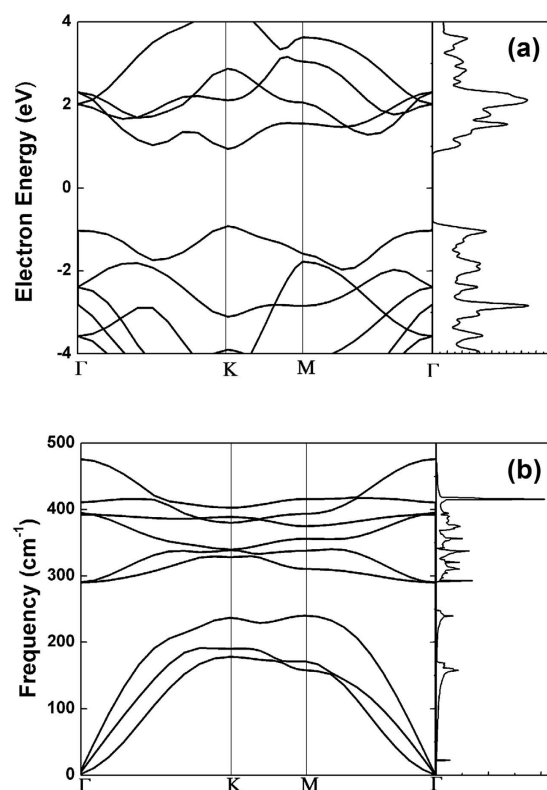


Figure 2. (a) Electron band structure and density of state along the symmetry line. The Fermi energy is set in the middle of the gap. (b) The phonon dispersion for SLMoS₂ and the phonon density of state in the whole Brillouin zone.

of single crystal MoS₂ would be 85–110 Wm⁻¹K⁻¹³³. There is not an agreement on the κ of SLMoS₂, and it needs more works on this issue.

In this paper, both electron and phonon transport properties of SLMoS₂ are studied (the structure as shown in Fig. 1). Based on the electronic band structure from first-principles calculations, the electrical conductivity of SLMoS₂ is calculated by Boltzmann equations. Both the electronic structure and phonon dispersion relation are calculated. Together with κ_p calculated from classical EMD simulations, the thermoelectric properties are obtained. The results show that SLMoS₂ is a promising material for thermoelectric engineering.

Results and Discussions

The electronic band structure of SLMoS₂ along the high-symmetry points in Brillouin zone is shown in Fig. 2(a). At the K point, there is a direct band gap as 1.86 eV which agrees well with previous calculations (1.69 ~ 1.98 eV)^{20–23,34,35}. Another characteristic in the SLMoS₂ band structure is that there is a Q valley along the Γ -K path. The Q valley

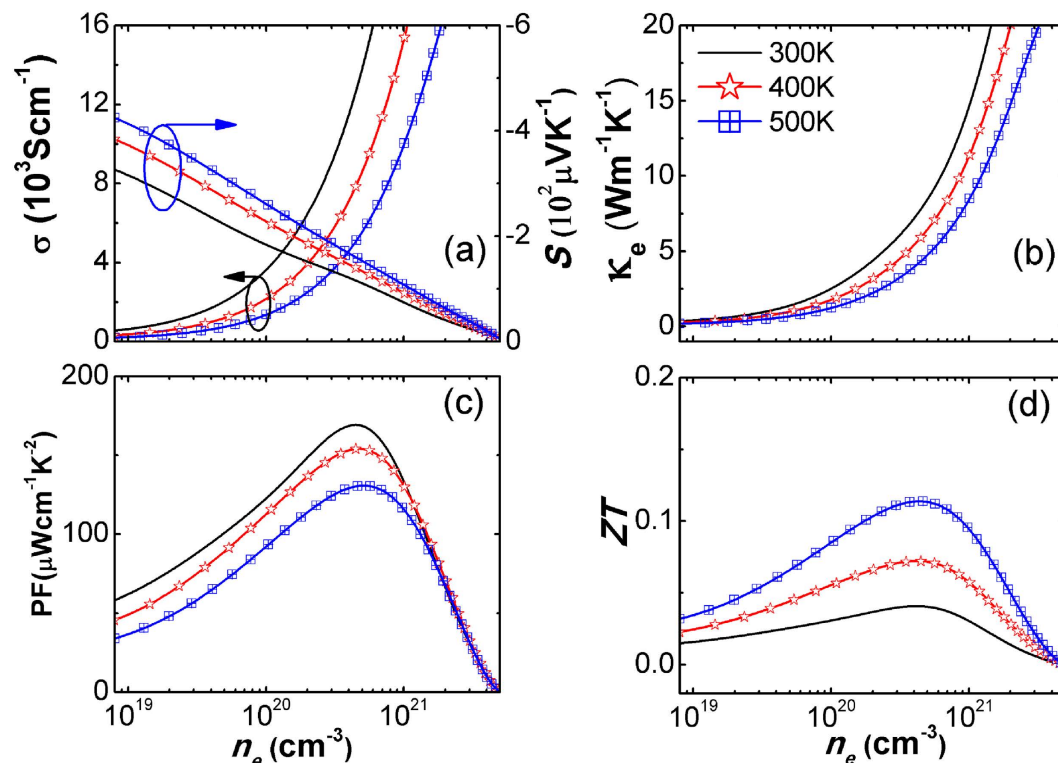


Figure 3. The thermoelectric transport properties of n-type SLMoS₂ at 300K, 400K and 500K. (a) The electrical conductivity and Seebeck coefficient; (b) The electrical thermal conductivity; (c) The power factor; (d) The figure of merit. The thermoelectric transport properties of p-type is shown in supporting information.

yields a larger effective mass than the K valley, which leads to strong electron-phonon interactions in MoS₂ at this point²². The large effective mass of carriers and multi-valleys band structure are favorable for a high ZT³⁶. As shown in the density of state (DOS) electrons (Fig. 2), there are sharp gradients at the edges of both conduction and valence band and several peaks near band edges, due to the quantum size effects in the 2D structure, which may enhance ZT as the prediction of Mahan and Sofo¹².

We made a full calculation of the thermoelectric properties of SLMoS₂ at 300 K, 400 K and 500 K. As shown in Fig. 3(a,b), κ_e and σ increase as the increasing of carrier concentration (n_e). When the Fermi level is in the band gap, n_e and σ is much smaller. As the Fermi level moves up into the conduction band, n_e and σ increases quickly (more details shown in Fig. S4 and S6 in supporting information). Shown in Fig. 3(a), the Seebeck coefficient has a large value and decreases with the increase of n_e . The Fermi level for ZT peak locates around the first DOS peak, and this is consistent with the prediction that a delta DOS would result in an optimum ZT¹². It leads to a power factor as high as several hundreds of $\mu\text{Wcm}^{-1}\text{K}^{-2}$ (shown in Fig. 3(c)), which is compared with those of high ZT thermoelectric materials, such as BiTe³⁷ and PbTe³⁸.

The phonon dispersion relation of SLMoS₂ is also calculated and shown in Fig. 2(b). In the vicinity of Γ point, the out-of-plane transverse acoustic branch (ZA) has a quadratic relation, both the transversal acoustic branch (TA) and longitudinal acoustic branch (LA) have linear relations. The group velocities at Γ point along Γ -M direction are around 667.5 m/s (TA) and 1080.2 m/s (LA), which are much smaller than the group velocities in graphene³⁹, as 3743 m/s (TA) and 5953 m/s (LA).

For semiconductors, the thermal conductivity is mainly contributed by phonons (κ_p). We calculated κ_p by EMD and show in Fig. 4. The κ_p of SLMoS₂ exhibits a size dependence on the simulation cell and reaches a converged value when the simulation cell is larger than $8 \times 8 \times 1$ units³ ($8.66 \times 7.50 \times 0.616$ nm³) (Fig. 4(a)). A weak anisotropy is observed in thermal conductivities along armchair and zigzag direction. The average value of κ_p along armchair and zigzag directions is $116.8 \text{ Wm}^{-1}\text{K}^{-1}$ for simulation cell as large as $32 \times 32 \times 1$ units³ ($34.7 \times 30.0 \times 0.616$ nm³) at 300K. In Fig. 4(b), the κ_p of SLMoS₂ decreases with the increasing temperature ($79.6 \text{ Wm}^{-1}\text{K}^{-1}$ and $52.9 \text{ Wm}^{-1}\text{K}^{-1}$ at 400 K and 500 K, respectively), because there are more three phonon Umklapp scatterings for high temperature. A lower κ_p is good for enhancing thermoelectric properties.

Comparing with previous results (details in Table 1), we obtained a maximum value of κ_p of SLMoS₂. Some of these works focused on the SLMoS₂ nanoribbons^{26,27,29} which have very low thermal conductivities, because the phonon confinement effect in nanostructures^{40,41}. Using the same empirical potential in MD simulations, our results for SLMoS₂ is around 20 times larger than that of nanoribbon with $34.6 \times 30 \times 0.61$ nm³ in size. Besides, due to the interlayer coupling by van Der Waals forces⁴², the multilayer structures^{31,33} should be lower than the single layer^{27,28,30,32} in thermal conductivity. Due to the absence of impurities, defects and interlayer scatterings in MD simulations, the κ_p of SLMoS₂ is a little higher than the measurements of bulk multilayer SLMoS₂³³, $85 \sim 110 \text{ Wm}^{-1}\text{K}^{-1}$. Our value is comparable to the result predicted from *ab initio* calculation³⁰, where stated that the lower bound

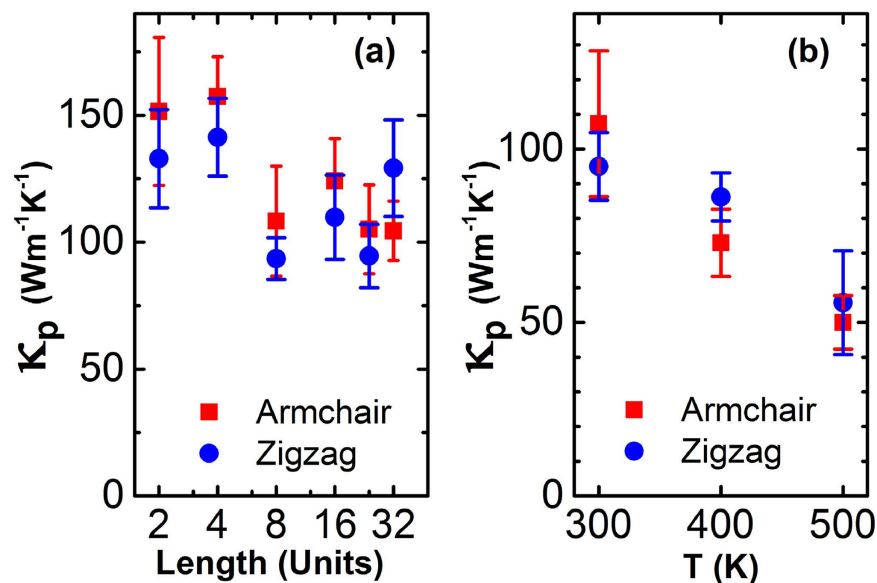


Figure 4. (a) The dependence of thermal conductivity (κ_p) of SLMoS₂ upon the size of simulation cell. The size of simulation cell size equals the length (L) times a supercell ($1.083 \times 0.938 \times 0.616 \text{ nm}^3$). (b) The thermal conductivity of SLMoS₂ for three different temperature as 300K, 400K and 500K, respectively.

Struct. & Ref.	Method	T (K)	Carrier type	σ (Scm ⁻¹)	S (μVK ⁻¹)	κ_e	κ_{ph}	ZT
						(Wm ⁻¹ K ⁻¹)		
SL	DFT + BTE + MD	300	n	14625	-110	8.94	116.8	0.04
			p	16957	72.9	11.39		0.02
		500	n	11714	-161	9.69	52.9	0.26
			p	8853	150	8.40		0.16
SL ²³	DFT + Ballistic model	300	n	54	-202	0.021	0.243	0.25
			p	108	215	0.040	0.244	0.53
SLR ^{24,27}	DFT + BTE + MD	300	n	7770	-204	2.89	1.02	2.5
			p	14300	223	5.20		3.4
SL CVD ²⁵	Experiment	300	-	-	≤30000	-	-	-
SL FET ⁵⁹	Experiment	300	-	-	400-100000	-	-	-
Bulk ⁶⁰	Experiment	90-873	-	-	500-700	-	-	-
SL ²⁷	EMD	300	-	-	-	-	1.35	-
SL ³⁰	DFT + BTE		-	-	-	-	>83	-
SL ²⁸	DFT + NEGF		-	-	-	-	23.2	-
SLR ²⁹	DFT + BTE		-	-	-	-	26.2	-
SLR ²⁶	NEMD		-	-	-	-	5	-
FL ³¹	Experiment		-	-	-	-	52	-
SL ³²	Experiment		-	-	-	-	35.4	-
Bulk ³³	Experiment		-	-	-	-	85-110	-

Table 1. The comparison of thermoelectric properties for different MoS₂ structures, including single layer (SL), few layers (FL), single layer ribbon (SLR), and bulk MoS₂.

of κ_p as 83 Wm⁻¹K⁻¹ at 300 K in the considering of phonon scatters and the simplification in calculation of BTE model. Another advantage for our results is that both the nonequilibrium Green's function calculation²⁸ and the Boltzmann transport equation²⁹ adopt artificial relaxation time approximations for phonon-phonon Umklapp scatterings, which is not required in the MD simulations.

Generally, there are two types of commonly used MD simulation methods, EMD and non-equilibrium MD (NEMD). The EMD is better than NEMD in predicting a bulk structure by applying the periodic boundary condition, because NEMD need impose artificial heat bath and use the extrapolating method. However, NEMD gets the advantage of EMD in predicting a structure with a finite size. There is a EMD report which showed a low value of κ_p of SLMoS₂ as 1.35 Wm⁻¹K⁻¹²⁷. The potential functions used in Ref. [27] are determined by tight-binding

quantum chemistry calculations and used to reproduce the crystal structure and Raman spectrum. The empirical potential function is important to obtain a reliable value of thermal conductivity. Differently, the Stillinger-Weber potential²⁶, used in this work, can reproduce a better phonon dispersion relations, which will describe the heat transfer properties with a better reliability (details in supporting information).

With the above calculations of electron and phonon properties, ZT profiles can be obtained and are shown in Fig. 3(d). There is a parabolic tendency for ZT in the whole carrier concentration range. The optimized ZT values are 0.04, 0.07, and 0.11 for 300 K, 400 K and 500 K, respectively. These values get bigger as temperature increases because of the improved power factors and the reduced thermal conductivity. These optimized ZT values correspond to the situations where the Fermi level moves up to the first peak in the conduction band.

As shown in Table 1, we list some recently results on thermoelectric properties of different SLMoS₂ structures. The value of ZT is in the same order of Ref. [23] and one order smaller than that of SLMoS₂ ribbon. Our results of κ_p is higher than others. As shown in Fig. 4(a), our results of κ_p overcome the size confinement effect and corresponds to an infinite SLMoS₂ sheet. Moreover, different from NEMD, it does not need the assumption of linear relationship between $1/\kappa_p$ and $1/L$.

Compared to nanoporous silicon analyzed by Lee⁴³, we get the similar ZT trend and magnitude of these transport values. As shown in Fig. 3(c), the power factor of SLMoS₂ is larger than that of nanoporous silicon. The large power factor of SLMoS₂ comes from a larger intrinsic σ and a comparable S . It indicates that the SLMoS₂ has comparable electron properties as the optimized nanoporous silicon. However, due to the high κ_e and κ_p , the SLMoS₂ has a modest ZT value. It is also worth noting that the ZT value here is smaller than the prediction from ballistic models by Huang *et al.* where neglects the phonon scatterings²³.

Although the predicted ZT value of SLMoS₂ is not over one, SLMoS₂-based materials may be a good candidate for thermoelectric application. Our results show that SLMoS₂ has a much higher thermal conductivity ($\sim 116 \text{ W m}^{-1} \text{ K}^{-1}$, at 300 K) than other thermoelectric materials (on the order of $1 \text{ W m}^{-1} \text{ K}^{-1}$)^{6,44}. The higher thermal conductivity makes a bigger room for thermal conductivity reduction by phonon engineering. There are some conventional ways to reduce thermal conductivity by phonon engineering, such as isotope doping⁴⁵, nanoporous structure^{14,43,46,47}, nanoribbons⁴⁸, or folding⁴⁹ etc. The mechanism is to introduce more phonon scatterings which can shorten phonon mean free paths. For example, bulk silicon has a ZT value as low as 0.003. Then, with phonon engineering, Si-based nanomaterials, such as Si nanowires^{45,50}, nanoporous Si^{14,43,46,47}, and nanostructured Si⁵¹, may reach a two orders larger ZT . Another inspiration example is the graphene. The high pristine thermal conductivity of graphene can be reduced largely by phonon engineering^{48,49,52} which make a ZT as high as 3⁵².

The values of ZT and power factor of SLMoS₂ are much higher than those of silicon and graphene. With a reduced thermal conductivity and kept electron transport properties, the values of ZT of SLMoS₂-based materials may be larger than one. Generally, a side-effect of phonon engineering is the reduction of power factor. However, the side-effect is not obvious because the mean free paths of electrons are around two orders smaller than that of phonons, such as what is shown in the recent thermoelectric results on Si phononic crystals⁴⁷.

Conclusion

The thermoelectric properties of SLMoS₂ are explored using theoretical calculations. The electronic structure and phonon dispersion relation are calculated using DFT calculations. Combined with molecule dynamics simulations and Boltzmann equations, thermoelectric properties are predicted as a function of carrier concentration at room temperature. With the lattice thermal conductivity as $116.8 \text{ W m}^{-1} \text{ K}^{-1}$, $79.6 \text{ W m}^{-1} \text{ K}^{-1}$, and $52.9 \text{ W m}^{-1} \text{ K}^{-1}$, the optimized ZT of SLMoS₂ is found to be of 0.04, 0.07 and 0.11 at 300 K, 400 K and 500 K, respectively. As SLMoS₂ has a higher ZT than other pristine structure, like silicon and graphene, there will be a big room to enhance ZT in SLMoS₂-based materials by the developing phonon-engineering.

Methods

To calculate electronic properties, the first-principles calculation is implemented by QUANTUM ESPRESSO in the frame of density functional theory (DFT)⁵³. The local density approximation (LDA) is used in the exchange-correlation approximation while the semi-core valence for molybdenum is considered with the Goedecker-Hartwigsen-Hutter-Tetter method⁵⁴. The wave-functions in electronic calculation are cut off at 160 Ry, and the irreducible Brillouin zone is sampled with a $16 \times 16 \times 1$ Monkhorst-Pack grid.

The hexagon primitive cell is used to structure relaxation and property prediction in DFT calculation. Structure relaxation for SLMoS₂ yields lattice constant of about 3.13 Å, consistent with previous predictions of 3.12–3.16 Å^{20–22}. For the consistency of property evaluation, the thickness of SLMoS₂ is assumed to be 6.16 Å – the same as that of the single-sheet in bulk MoS₂⁵⁵. The calculations on both electrons and phonons are based on this optimized structure.

In the calculations of transport coefficients, a k-point mesh as $28 \times 28 \times 1$ (denser enough to obtain converged results) is used over the irreducible Brillouin zone. With the assumption of constant relaxation time, the transport coefficient for electrons can be calculated using BoltzTrap⁵⁶ which solves Boltzmann transport equation (more details in supporting information).

The thermal conductivity of SLMoS₂, κ_p , is calculated by EMD with the Green-Kubo approach⁵⁷. All the simulations are carried out utilizing the LAMMPS software package⁵⁸. The Stillinger-Weber potential with parameters fitted by Jiang *et al.*²⁶ is adopted in our simulations. The SLMoS₂ film is constructed by periodic arrangement of supercell illustrated in Fig. 1, and the sizes of $1 \times 1 \times 1$ units³ supercell corresponds to $1.083 \times 0.938 \times 0.616 \text{ nm}^3$. To study the finite size effect on thermal conductivities, we calculated the simulation cells with the volumes from $2 \times 2 \times 1$ to $32 \times 32 \times 1$ units³ at room temperature (more details in supporting information).

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Author Contributions

Z.J. and Z.D. carried out the first-principles calculations. Q.L. carried out the MD calculations. N.Y., Z.J., Q.L. and Z.D. analyzed the calculation data. H.F., Z.L., W.L. and T.L. discussed and commented on the manuscript. Z.J., Q.L., Z.D. and N.Y. edited the manuscript. N.Y., H.F. and Z.L. supervised the research.

Additional Information

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