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Anisotropic control of thermal transport in graphene/Si heterostructures

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The cross-plane interaction across interface changes phonon kinetics and spectrum near the interface, and the interaction effects on both in-plane and cross-plane thermal transport are investigated in graphene/Si heterostructure. The interaction with substrates dramatically reduces the in-plane thermal conductivity of graphene by changing the behaviors of the out-of-plane phonons as well as adding phonon-substrate scatterings. Applying pressure up to 2.6 GPa to the sandwiched graphene reduces the cross-plane interfacial thermal resistance by 50% without altering the in-plane thermal conductivity in a significant way. The pressure increases the inter-layer coupling and creates a low-energy phonon transport channel between graphene and Si with minor effects on phonons propagating along the graphene. This study suggests the anisotropic control of thermal transport, and the physics and calculation results can be used to improve the thermal design and analysis in two-dimensional nano-electronic devices. Published by AIP Publishing.

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I. INTRODUCTION

As the electronic industry miniaturizes, the devices and electronic parts shrink to nanometers, the thermal management of these devices is becoming a challenge;1 effective heat dissipation is required for the device performance and stability.2 The reported high electrical3 and thermal conductivities4 of single-layer graphene (SLG) have made this material a very promising candidate for electronic applications such as field effect transistors.7 However, when graphene is supported by a substrate, the superior thermal conductivity is dramatically reduced due to the influence of contacting materials.8 Since graphene in micro- and nanoelectronics is mostly supported or sandwiched rather than suspended,8,10 both in-plane and cross-plane thermal transports, which are characterized by in-plane conductivity and interfacial conductance (inverse of resistance), should be studied with the consideration of the cross-plane interactions of contacting materials for the thermal design of graphene applications.8,11–16 In particular, the interfacial thermal transport between graphene and substrates plays a critical role in the saturation of graphene electronic devices.17–20 There are many recent studies on different methods of controlling interfacial thermal transport,14,21,22 however, for further enhancement of electronic device design and heat dissipation, we still need to find an effective, cheap, and feasible way of controlling the directional thermal transport, and improving the physical understanding on interfacial interactions.

In this research, we study the thermal transport in various configurations and conditions of graphene heterostructures for understanding the cross-plane interaction physics, finding methods to control the thermal transport, and enhancing the thermal design and analysis. Since quantized atomic vibrations, i.e., phonons, are the main thermal energy carriers in the graphene systems,9,10 we focus on thermal transport by phonons. Molecular dynamics (MD) is employed to study the phonon transport including the in-plane conductivity of SLG as well as the cross-plane interfacial conductance between SLG and substrate. Si is selected as the substrate because it is a representative of semiconductors and the most common material used in the electronic devices.

To identify the influence of cross-plane interaction on phonon behavior, which is expected to cause the change in thermal transport properties, we calculate the phonon density of state (DOS) of SLG under different conditions. Supported and embedded (sandwiched) forms of SLG are investigated, and the phonon transport properties and spectra in the heterostructures are compared with the suspended (free-standing) SLG. In the sandwiched SLG, we apply the compressing pressure, as a potential tool to control the strength of the interactions between SLG and Si substrates, and study the effects on the interfacial thermal resistance as well as in-plane thermal conductivity.

II. METHODOLOGY

Periodic boundary conditions are applied in x and y directions, while a free boundary condition is used in the z direction to avoid interactions between periodic cells. Here, x is the in-plane heat flow direction, and z is the flow direction for interfacial thermal transport [Figs. 1(a)–1(d)]. Our simulations showed that the width dependency of thermal conductivity is negligible for sample wider than 11 nm, so we choose 11.6 nm for the simulation sample width (w in y direction) to reduce the computational cost and minimize the lattice mismatch in Si/SLG heterostructures. Various lengths (l) are simulated for the length dependence of thermal conductivity (k) and the calculation of bulk thermal conductivity (k0). l ranges from 40 to 2400 nm in simulations for the suspended SLG; however l’s in the supported and sandwiched SLG structures are less than 300 nm due to the computational
cost. For the cross-plane transport, a fixed, mid-size sample length ($\frac{l}{C24} 200 \text{ nm}$) is selected. The width and length of the Si block are the same as the graphene, and the size of Si in $z$ direction is 3.8 nm. Based on our simulations on different Si block sizes, this thickness of Si block is large enough to avoid the size dependence of simulation results. All the simulations were performed in Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS), all results in this work are the average of five independent simulations, and the time step for all our simulations is chosen to be 0.5 fs.

Adaptive intermolecular reactive empirical bond order (AIREBO)$^{24}$ is used to model the C-C bonded interactions, and three-body Stillinger-Weber$^{25}$ potentials for Si-Si interactions. Primarily, short-range, van der Waals (vdW) interaction$^{26,27}$ between C and Si atoms is modeled by the 12–6 Lennard-Jones (LJ) potential, $\phi_{LJ} = 4\varepsilon[(\frac{\sigma}{r})^12 - (\frac{\sigma}{r})^6]$, where $\varepsilon$ is the energy at equilibrium position, $\sigma$ is the distance at which the intermolecular potential between the two particles is zero, and $r$ is the distance of separation between particles.

The potential parameters used in our simulations are based on the vdW interactions in the universal force field (UFF) model by Rappe et al.$^{28}$

Thermal conductivity is calculated by using the non-equilibrium molecular dynamics (NEMD).$^{29-35}$ After the equilibration in $NVT$ ensemble (constant number of particles, volume, and temperature) at a measured $T$ for 0.5 ns employing velocity rescaling, the hot and cold Langevin thermostats are added at $x = l/4$ and $3l/4$ with a width of $l/20$ (i.e., distance between two thermostats, $d_{H-C} = l/2$) [Fig. 1(a)].$^{30,31,34}$ Temperature is averaged for a long enough period of time (1 ns) to minimize the noises in temperature distribution.

Thermal conductivity $k$ (W/m-K) is calculated by using Fourier’s law $q = -k \nabla T$, with a heat flux from the hot to the cold region $q$ (W/m$^2$), and a temperature gradient $\nabla T (= dT/dx$ in 1D transport). $q$ is the average heat addition and removal over 2 ns on thermostats per unit cross-sectional area. The interlayer spacing of graphite, (0.335 nm$^{36}$), is selected as the thickness of a single-layer graphene for the

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**FIG. 1.** Simulation models for the calculation of thermal transport properties. (a) Suspended graphene thermal conductivity. The hot and cold thermostats are placed to create temperature gradient ($dT/dx$) and heat flux ($q$) along the sample for the in-plane thermal conductivity. (b) Supported graphene thermal conductivity. The hot and cold thermostats are placed on graphene and Si, separately. (c) Interfacial thermal resistance for supported graphene. The hot thermostat is placed on the graphene, and the temperature and energy evolution during thermal relaxation are recorded to calculate the interfacial thermal resistance. (d) Interfacial thermal resistance for sandwiched graphene. In addition to the thermal relaxation, the $z$-direction force is applied to the upper Si block for the pressure dependence of the thermal transport.
cross-sectional area. For $\nabla T$, $T$-distribution is calculated from the local kinetic energy average, and to avoid the thermostat effects and $T$-dependence, the linear part of $T$-profile near a measured $T$ is used.

In order to calculate the thermal conductivity of graphene with a Si substrate, hot and cold thermostats are placed in graphene and Si for direct calculation of graphene heat flux as in Fig. 1(b) separately. The hot and cold thermostats in graphene and Si are set as the same temperatures, and after equilibration of MD simulations, Si and graphene temperature distributions are parallel and almost identical; thus, heat leakage to or an addition from the Si substrate is negligible. In addition to heat flux at thermostats (hence, heat leakage to or an addition from the Si substrate is negligible), the temperatures and total energies of the graphene and Si are recorded and used for the calculation of (areal) interfacial thermal resistance, $R$ (m$^2$ K/W) given as

$$R = \frac{1}{V} \left[ \sum_{i=1}^{N} m_i (u_i \cdot u_i) u_{i,x} + \sum_{j=1}^{N} q_j u_{j,x} \right],$$

where $V$ is the volume, $m_i$ is the mass of particle $i$, $u_i$ is the velocity vector, $u_{i,x}$ is the $x$ component of $u_i$, $q_j$ is the potential energy, $x_{ij}$ is the $x$ component of the interatomic separation ($r$), and $F_{ij}$ is the interaction force between $i$ and $j$ particles. In all cases, heat fluxes calculated at thermostats agree well with the local heat flux calculation and the local heat flux is almost constant ($\leq 5\%$ difference), which confirms a negligible heat leakage, i.e., 1D thermal transport under this thermostat setup. In this research, a local heat flux at the center ($l/2$) will be employed in conductivity calculation. To estimate the contributions of out-of-plane and in-plane phonon modes to thermal transport separately, either $z$-direction polarization only or $xy$-direction polarization only is included in the heat flux calculation. For the sandwiched graphene simulation, the graphene layer is placed between two Si blocks [Fig. 1(d)] and the same methodology as in the supported case is employed for the calculation of $q_T$ and $k$.

The interfacial thermal resistance is calculated by analyzing the temperature relaxation. The system is equilibrated first at 300 K, and then hot and cold thermostats are placed on graphene and Si, respectively. On reaching a stable temperature on graphene and Si, the thermostats are removed to relax the system thermally. During the relaxation period, the temperatures and total energies of the graphene and Si are recorded and used for the calculation of (areal) interfacial thermal resistance, $R$ (m$^2$ K/W) given as

$$R = \frac{\partial E_T}{\partial T} = \frac{A \left( T_{\text{SLG}} - T_{\text{Si}} \right)}{E_T},$$

where $E_T$ is the total energy of the supported graphene, $A$ is the thermal transport interface area, and $T_{\text{SLG}}$ and $T_{\text{Si}}$ are the graphene and Si temperatures. In our simulations and previous studies, $R$ is almost constant for the temperature range of simulation (200–500 K), so approximating $R$ as the constant, Eq. (2) is integrated as

$$E_T = E_0 + \frac{R}{A} \int_0^t (T_{\text{SLG}} - T_{\text{Si}})dt,$$

where $E_0$ is the initial total energy of graphene. In the sandwiched graphene case, $A$ is doubled since heat flows in both up- and downward direction.

The sandwiched structure is selected to investigate the effect of the pressure ($p$) on the interfacial thermal resistance and in-plane thermal conductivity. We apply a uniform force to the upper Si block in negative $z$ direction while constraining the $z$ direction movement of the bottom-layer atoms of lower Si block to keep it in its place [Fig. 1(d)]. Phonon density of states ($D_p$) is calculated through the Fourier transform of the velocity autocorrelation function. For this calculation, we use velocities of sampled atoms exported at every time step from NVE ensemble simulations.

III. RESULTS AND DISCUSSIONS

Since the distance between hot and cold reservoirs ($d_{H-C}$) is comparable to or smaller than the SLG phonon mean free path ($\lambda_p \approx 750$ nm), thermal conductivity ($k$) is dependent on $d_{H-C}$ due to the ballistic effect; i.e., shorter $d_{H-C}$, lower $k$ in NEMD simulations as in Fig. 2(a). If $d_{H-C} \ll \lambda_p$, transport between two reservoirs will be almost scattering-free, and $k$ will be proportional to the product of ballistic conductance ($G_B$) and sample length (i.e., $k = G_B d_{H-C}$). However, the size-dependence of $k$ is negligible in diffusive regime ($d_{H-C} \gg \lambda_p$) and the diffusive $k$ is approximated as $G_B \pi \lambda_p/2$. Combining both ballistic and diffusive behaviors, $k$ for intermediate $d_{H-C}(\sim \lambda_p)$ is suggested as follows in the previous studies:

$$k \sim G_B \left( \frac{1}{d_{H-C}} + \frac{2}{\pi \lambda_p} \right)^{-1}.$$  

However, $k$s from the NEMD simulations is not fitted well to Eq. (4), and especially a larger deviation from the fitting equation is observed with a longer length ($d_{H-C}$) than $\lambda_p$. This anomalous length-dependence of $k$ has been studied, and is attributed to the additional participation of low-frequency, long-wavelength phonons in transport, which is restricted by the system size. In Fig. 2(a), we used three aforementioned equations (for three length ranges) to fit the thermal conductivity results from our simulations. The first region (red, dashed-dot line) is for the ballistic thermal transport regime and the linear relation between $k$ and $G_B$, the second region (purple, dot line) represents the range of intermediate sample length, and the third region (black, long-dashed line) is for a long sample length, which incorporates more long-wavelength phonon contributions.

Using the fitting method, $G_B_{\text{sus}}$, bulk conductivity $k_{0_{\text{sus}}}$, and $\lambda_{p_{\text{sus}}}$ in suspended SLG are obtained as 4.286 GW/m$^2$ K, 2235 W/m K, and 332 nm, respectively, as in Fig. 2(a). The discrepancies from the other experimental and theoretical studies (reported $k_{0_{\text{sus}}} = 3000–5000$ W/m K, $G_B_{\text{sus}} = 4.167$ GW/m$^2$ K, and $\lambda_{p_{\text{sus}}} = 750$ nm) are attributed to the limited size and approximations in MD simulations and AIREBO model; however, the overall phonon
Properties and transport behaviors are reproduced well with our current approaches. The in-plane thermal conductivities of SLG in the supported and sandwiched structures ($k_{\text{sup}}$ and $k_{\text{sw}}$) are calculated, and their length dependence for relatively short-size range ($<150$ nm, due to the computational cost) is also studied. As in Fig. 2(b), thermal conductivity is dramatically reduced by the contact with a Si substrate in the supported structure; however, the interaction with additional contact surface in the sandwiched structure does not have a significant effect on $k$. $k_{\text{sw}}$ is slightly smaller than $k_{\text{sup}}$, and their length dependencies are also similar to each other, showing that both relative thermal conductivities compared with the suspended SLG (i.e., $k_{\text{sup}}/k_{\text{sus}}$ and $k_{\text{sw}}/k_{\text{sus}}$) decreases with $d_{\text{H-C}}$ [Fig. 2(c)].

The high thermal conductivity of suspended SLG is attributed to a larger density of states ($D_p$) of out-of-plane modes ($ZA$ and $ZO$) at low energies, which results in higher $c_p$, and low phonon scattering rates of those phonon modes ($Z$: $z$-direction polarization, $A$: acoustic, and $O$: optical). The low scattering rates originate from the fact that approximately 60% of the normal ($N$) and Umklapp ($U$) scattering of the out-of-plane phonons are prohibited by the selection rules; thus, the long mean free path of $ZA$ and $ZO$ phonons in comparison with the in-plane phonons leads to a dominant contribution to in-plane thermal conductivity of suspended graphene (more than 77%$^{10}$). However, these $ZA$ and $ZO$ phonons are significantly affected by the contact, whereas in-plane phonons with LA, TA, LO, and TO polarizations ($L$: longitudinal, and $T$: transverse) show only a slight change, as $D_p$s for in-plane and out-of-plane modes in suspended and supported graphene show in Fig. 3(a). In particular, the $ZA$ mode in supported or sandwiched graphene does not have nonzero $D_p$ at the phonon energy of zero any more, while the suspended graphene does due to the quadratic dispersion by flexural bending rigidity.$^{59}$ Since low-energy (long-wavelength) $ZA$ phonons have less scattering rates or longer relaxation time,$^{50}$ the smaller population of the near zero energy phonons will cause a shorter average effective mean free path $(\lambda_p)$, reducing the thermal conductivity. Additionally, the $ZO$ mode is shifted to the energy of substrate Si optical mode peaks by the cross-layer interaction, which also enhances scattering with substrate phonons. Compared to the clear difference between suspended and supported $D_p$s in out-of-plane polarization, additional contact in the sandwiched structure has a minor effect on phonon spectrum as in Fig. 3(b). The relative contribution of each phonon polarization (in-plane or out-of-plane) to thermal conductivity [Fig. 3(c)] shows that out-of-plane phonon contribution is dominant in the in-plane thermal transport of suspended graphene, while in-plane phonons carry most of the thermal energy in supported and sandwiched cases. This confirms that the dramatic reduction of thermal conductivity stems from increased scattering of out-of-plane phonons. Very close participation percentage of $xy$ and $z$ phonons in supported and sandwiched graphene and analogous $D_p$s for these cases, can explain the small difference between supported and sandwiched graphene in-plane thermal conductivity.

Using $D_p$ in Figs. 3(a) and 3(b) with the equilibrium phonon (Bose-Einstein) distribution function $f_{\text{BE}} = \exp[h\nu/k_B T] - 1^{-1}$, the phonon specific heat capacities ($c_p$) of suspended and supported SLGs are calculated as.
where $k_B$ is the Boltzmann constant. The $c_p$ of SLG does not vary dramatically with different structures; moreover, the out-of-plane modes in the supported case have a slightly higher $c_p$ than the suspended at temperatures around 300 K in spite of its lower $k_{sup}$ as in Fig. 3(d).

Since the phonon conductivity $k$ is proportional to $c_p$, $u_p$, (average velocity), and $\lambda_p$ according to the kinetic theory,\cite{61} we can conclude that the large reduction in $k$ is mainly due to the increasing phonon scattering (shorter $\lambda_p$). The phonon mean free path ($\lambda_p$) increases with the time constant of phonon scattering ($\tau_p$), which decreases with more scatterings. In the simulation of supported graphene, SLG phonons are scattered by other SLG phonons $p-p$, substrate interaction $p-sub$, and thermostats $p-d$, due to the limited transport distance $d_{H-C}$, and these scattering mechanisms are characterized by three time constants; $\tau_{p-p}$, $\tau_{p-sub}$, and $\tau_{p-d}$, respectively. Using the Matthiessen rule,\cite{62} the overall time constant of phonon scattering ($\tau_p$) is given as

$$\tau_p^{-1} = \sum_j \frac{1}{\tau_{p,j}} = \frac{1}{\tau_{p-p}} + \frac{1}{\tau_{p-sub}} + \frac{1}{\tau_{p-d}}.$$  

Here, $\tau_{p-d}$ increases with the sample size ($d_{H-C}$), while other main mechanisms are not size-dependent; thus, with a longer $d_{H-C}$ (or $\tau_{p-d}$), the relative contribution of $\tau_{p-sub}$ and $\tau_{p-p}$ to overall $\tau_p$ is more significant. Since the shorter $\lambda_p$ (or larger scattering rate) for lower $k$ in the supported or sandwiched structures originate from the increased $p-p$ or $p-sub$ scattering when considering a common $d_{H-C}$, the change in conductivity by substrate [(k$_{sus}$ - k$_{sup}$)/k$_{sus}$] increases with $d_{H-C}$.
(i.e., $k_{\text{sup}}/k_{\text{sw}}$ decreases). This negative length-dependence of relative conductivity is observed both in the supported and sandwiched structures as in Fig. 2(c).

The phonons in the supported and sandwiched SLGs have an almost identical DOS [Fig. 3(b)] (thus, very similar $c_p$) and ballistic conductance from the similar linear dependence for a very short length of system [Fig. 2(b)]. Thus, a minor difference between $k_{\text{sup}}$ and $k_{\text{sw}}$ despite additional substrate demonstrates a small difference between the supported and sandwiched SLG scattering rates. This small difference implies that the substrate scattering ($\tau_{p-\text{sup}}$) is not significant although it can further reduce $k$, and the additional contact in the sandwiched structure increases only the minor substrate scattering. Therefore, we deduce that through the contact with a single substrate, the SLG loses its 2D characteristics (including the flexural bending motion and the mentioned prohibitions in selection rules), which results in low $p-p$ scattering rate (especially in out-of-plane modes) for higher $k$. The extensive change in the out-of-plane phonon modes, which have dominant contributions to the in-plane thermal transport in suspended SLG, and the minor difference between the supported and sandwiched phonon properties explain the observed behavior in both structures 

Thermal relaxation between hot SLG and cold Si is analyzed for the cross-plane thermal transport. The resulting equilibrium temperature in the sandwiched structure is lower than the supported structure, due to its doubled Si mass or doubled heat capacity as shown in Fig. 4(a). The decays in temperature difference ($\Delta T_{\text{SLG-Si}}$) and the energy in graphene ($E_{\text{SLG}}$) are fitted to the exponential function eliminating the noise from MD results. Using this fitted equation with Eq. (3), thermal boundary resistance ($R_{\text{SLG/Si}}$) is calculated using Eqs. (2) and (3), and the fitting parameters are adjusted in a way not to affect the simulation results [Fig. 4(b)].

The interfacial thermal resistance near the room temperature is almost constant in this range of temperatures as in Refs. 22 and 41–43, and our calculations also show that the $R_{\text{SLG/Si}}$ is almost consistent during the relaxation despite a variation of average interfacial temperature [$T_{\text{Si}} + T_{\text{SLG}}]/2$. The minor temperature dependence of interfacial transport in spite of a larger population of participating phonons at higher temperatures demonstrates the dominance (high interfacial transmission) of low-frequency phonons, excited already at low temperatures. The high transmission of low-frequency phonons is observed also in the other graphene heterostructures.

For the accuracy in the calculation of $R_{\text{SLG/Si}}$ at 300 K, we select data points around $[(T_{\text{Si}} + T_{\text{SLG}})/2]$ of 300 K. The calculated $R_{\text{SLG/Si}}$ in the supported case is $3.502 \times 10^{-8} \text{m}^2/\text{K/W}$, which is in good agreement with the previous reports ($3.57 \times 10^{-8} \text{m}^2/\text{K/W}$ through Raman method65 and $3.52 \times 10^{-8} \text{m}^2/\text{K/W}$ in other MD simulations66). As Fig. 4(b) shows, thermal relaxation in the sandwiched structure requires less time than the supported case; however, the calculated $R_{\text{SLG/Si}} (4.98 \times 10^{-8} \text{m}^2/\text{K/W})$ is larger than the supported one, and the faster decay in $\Delta T_{\text{SLG-Si}}$ and $E_{\text{SLG}}$ is attributed to the doubled interfacial area. Although the additional contact in the sandwiched structure does not make a significant change in the phonon spectrum and in-plane conductivity of SLG compared to the supported, more scatterings at the interface that are induced by stronger interaction with Si from both sides, leading to a larger $R_{\text{SLG/Si}}$ in the sandwiched structure.

The different effect of the interaction by additional contact on in-plane thermal conductivity and cross-plane resistance implies the possibility of the anisotropic control of thermal transport in SLG/Si heterostructure through the control of the cross-plane (i.e., out-of-plane direction) interaction. In this study, pressure normal to the interface ($p$), ranging from 0 to 2.6 GPa, is applied for the transport control in the sandwiched structure (Si/SLG/Si).

The in-plane thermal conductivity of the sandwiched SLG slightly decreases with increasing $p$, demonstrating minor effects of $p$ on $k_{\text{sw}}$ as in Fig. 5(a). $k_{\text{sw}}$ from the MD simulation under 2.6 GPa to the interface is 3.5% lower than without pressure, and the reduction using $k_{\text{sw}}$ from the linear fitting is 2.1%. In contrast, $R_{\text{SLG/Si}}$ significantly decreases with increasing $p$ (2.6 GPa reduces by around 50%) [Fig. 5(b)] as expected and observed also in the multi-layer graphene.21 This suggests the anisotropic thermal transport control (selective enhancement of cross-plane thermal transport) by applying $p$.

The $p$-increase reduces the distance between C in SLG and Si atoms in general; thus, a larger number of Si atoms are located within shorter distances with C atoms as shown in Fig. 6(a). The shorter distance induces a stronger interaction between C and Si, which is represented by the LJ potential. The average distance between the sandwiched SLG and the Si block is reduced by 7% in the presence of the pressure force.

FIG. 4. (a) Temperature evolution of SLG and Si during thermal relaxation. (b) The decays in temperature difference ($\Delta T_{\text{SLG-Si}}$) and the energy in graphene ($E_{\text{SLG}}$) (inset). The equilibrium temperature in the sandwiched structure is lower than the supported structure, due to its doubled Si mass or doubled heat capacity, and the relaxation is also faster in the sandwiched structure due to the larger contact area.
of 2.6 GPa, compared to one with no pressure \( (p = 0 \text{ GPa}) \). 

\( D_p \) of phonons with in-plane polarization, which is perpendicular to the pressure- (or force-) applied direction, does not vary with pressure as expected and shown in Fig. 6(b). In contrast, the phonon spectrum in the \( p \)-applied (out-of-plane or \( z \)) direction shows an additional low-energy peak in both the sandwiched SLG and interfacial Si as in Figs. 6(c) and 6(d). As \( p \) increases, this peak increases and \( D_p \)'s of SLG and Si are more synchronized. The stronger interaction and applied pressure constrain the motions of constituent atoms

FIG. 5. Variations of (a) in-plane thermal conductivity of SLG \( (k_{sw}) \) and (b) cross-plane interfacial thermal resistance \( (R_{SLG/Si}) \) in the sandwiched structure \( (\text{Si-SLG-Si}) \) with respect to the applying pressure \( (p) \). \( R_{SLG/Si} \) decreases as \( p \) increases, while \( k_{sw} \) does not vary significantly.

FIG. 6. (a) Atomic density distribution of Si with respect to the distance with C atoms. The increase in \( p \) decreases the average distance between C and Si, which induces the stronger interaction. (b) \( D_p \)'s of phonon modes polarized in the in-plane directions \( (xy) \) in the sandwiched SLG and interfacial-region Si under two different \( p \)s \((0 \text{ and } 2.6 \text{ GPa}) \). (c) Out-of-plane \( D_p \)s in the sandwiched SLG under the pressures of 0, 1.0 and 2.6 GPa. (d) \( D_p \)s of phonon modes polarized in the cross-plane direction \( (z) \) in the interfacial-region Si with respect to \( p \) \((0 \text{ GPa}) \) in the out-of-plane \( D_p \) in the sandwiched SLG under the maximum pressure is added as a reference). While the in-plane phonons do not change with \( p \), out-of-plane phonons in both interfacial Si and SLG have additional low-energy peaks in \( D_p \), which increases with \( p \). (e) \( D_p \)'s of silicon at different distances from the interface \((p = 2.6 \text{ GPa}) \). The peak near zero energy disappears after few layers of atoms far from the interface.
neighboring the interfaces, analogous to linkage to very heavy mass, which we attribute to the appearance and increase of low-energy phonon peak.

The peak is sharp and narrow, and its size in Si diminishes as the distance from the interface increases [Fig. 6(e)]; thus, we deduce that the phonon is localized (or not propagated) in the out-of-plane direction. Additionally, the out-of-plane mode, which the peak belongs to, has a minor contribution to the in-plane thermal transport as in Fig. 3(c), and the in-plane thermal conductivity is decreasing despite an increased phonon population by larger density of state at the peak. Therefore, the low-energy phonons in the peak do not contribute to the in-plane thermal transport, being localized. On the contrary, this low-energy vibration excitation in both Si and graphene can provide additional transport channels, enhancing local cross-plane interfacial transport.

As discussed above, phonons with a long wavelength (low frequency) dominantly contribute to the thermal transport due to their high transmission across interfaces, and this additional channel enhances the cross-plane thermal transport vividly; thus, the larger peak by higher pressure at the interface can lead to the lower interfacial resistance. Our additional simulations show that increasing $\varepsilon$ (characterizing the Si-SLG interaction strength) in the LJ potential induces similar effects of increasing $p$; a low-energy peak appears and increases with $\varepsilon$, in-plane $D_p$ is almost invariant, and in-plane thermal conductivity ($k_{\text{in-plane}}$) slowly decreases, while the decrease in interfacial thermal resistance ($R_{\text{SLG/Si}}$) is significant as shown in Figs. 7(a) and 7(b). However, large $\varepsilon$ is required to have a similar extent of change in phonon $D_p$ and transport (e.g., five times larger $\varepsilon$ leads to similar results of 1.0 GPa, $\varepsilon_0$ is the LJ potential parameter from UFF model by Rappe et al.\(^{28}\)), and very large $\varepsilon$ ($>10\varepsilon_0$) causes a large deformation of Si crystal structure near the interface. Therefore, the pressure control is more feasible in engineering, and more effective without undesirable effects (such as the unstable interfacial structures) than the control of interaction strength.

IV. CONCLUSIONS

Phonons polarized in the out-of-plane direction are significantly influenced by the cross-plane interaction with a contacting substrate, while the in-plane phonons are almost intact. The substrate effect is expected to activate the phonon scattering modes especially involving out-of-plane modes, which are prohibited in suspended 2D graphene, reducing the mean free path and thermal conductivity as a result. The additional contact surface in the sandwiched structure increases the substrate interaction in the out-of-plane direction, and applying a pressure induces the change in out-of-plane phonon modes; however, the change in conductivity ($k$) is not significant, indicating the minor effect of the cross-plane interaction control on the in-plane thermal transport of heterostructured graphene since the thermal energy is transported dominantly by in-plane phonons, which are almost invariant with pressure application. In contrast, the graphene/Si interfacial transport can be effectively controlled by the cross-plane interaction, thus the variation in out-of-plane phonon mode. Applying the pressure for the interaction control creates the additional low-frequency transport channel in the out-of-plane mode, which effectively reduces interfacial thermal resistance (by 50% with 2.6 GPa, while in-plane conductivity reduces by 3.5%).

This study offers insights on thermal transport properties in graphene heterostructures, which can be employed for the thermal system design and analysis of graphene devices. Effective, anisotropic thermal transport control proposed in this research is beneficial in many engineering applications, such as the space fission propulsion/power systems,\(^{65}\) electronic circuits,\(^{68}\) and energy storage systems.\(^{69}\) In a future study, thermal transport with different temperature and substrate, which leads to a different phonon mismatch and cross-plane interaction, and the phonon kinetics in those systems can be further studied for enhanced understanding.

\(^{1}\)A. Vassighi and M. Sachdev, Thermal and Power Management of Integrated Circuits (Springer Science and Business Media, 2006).


