Thermal conductivity of graphene grain boundaries along arbitrary in-plane directions: A comprehensive molecular dynamics study

Andy Fox, Upamanyu Ray, and Teng Li

Citation: Journal of Applied Physics **125**, 015101 (2019); doi: 10.1063/1.5059561 View online: https://doi.org/10.1063/1.5059561 View Table of Contents: http://aip.scitation.org/toc/jap/125/1 Published by the American Institute of Physics

Articles you may be interested in

Cu diffusion in CdTe detected by nano-metal-plasmonic enhanced resonant Raman scattering Journal of Applied Physics **125**, 013101 (2019); 10.1063/1.5051191

Photoinduced heat conversion enhancement of metallic glass nanowire arrays Journal of Applied Physics **125**, 015102 (2019); 10.1063/1.5059423

On the domain size effect of thermal conductivities from equilibrium and nonequilibrium molecular dynamics simulations Journal of Applied Physics **121**, 044301 (2017); 10.1063/1.4974884

Inter-layer and intra-layer heat transfer in bilayer/monolayer graphene van der Waals heterostructure: Is there a Kapitza resistance analogous? Applied Physics Letters **112**, 233104 (2018); 10.1063/1.5025604

The magnetic tunnel junction as a temperature sensor for buried nanostructures Journal of Applied Physics **124**, 174501 (2018); 10.1063/1.5049890

Strain engineering in functional 2-dimensional materials Journal of Applied Physics **125**, 082402 (2019); 10.1063/1.5053795



scitation.org/journal/jap

ARTICLE

Thermal conductivity of graphene grain boundaries along arbitrary in-plane directions: A comprehensive molecular dynamics study

Cite as: J. Appl. Phys. **125**, 015101 (2019); doi: 10.1063/1.5059561 Submitted: 20 September 2018 · Accepted: 7 December 2018 · Published Online: 27 December 2018



Andy Fox, ២ Upamanyu Ray, and Teng Li^{a)} ២

AFFILIATIONS

Department of Mechanical Engineering, University of Maryland, College Park, Maryland 20742, USA

^{a)}Author to whom correspondence should be addressed: lit@umd.edu

ABSTRACT

The thermal conductivity of polycrystalline graphene is expected to be lower than that of pristine graphene, due to the existence of defects, such as grain boundaries (GBs). To study the thermal transport behavior in polycrystalline graphene, it is crucial to understand the thermal conductivity of graphene GBs as a function of the tilt GB misorientation angle and in-plane thermal loading angle. However, existing studies of thermal conductivity of graphene GBs only consider the case where the thermal flux is perpendicular or parallel to the graphene GB. To address this issue, here we perform systematic non-equilibrium molecular dynamics simulations and investigate the thermal conductivity of graphene GBs for all possible tilt GB misorientation angles (23 cases) under arbitrary in-plane thermal loading directions. The findings from the present study can offer quantitative guidance for using polycrystalline graphene in thermal devices and flexible electronics applications.

Published under license by AIP Publishing. https://doi.org/10.1063/1.5059561

I. INTRODUCTION

Graphene, a planar sheet of sp²-bonded carbon atoms arranged in a honeycomb lattice, has become a promising candidate material^{1,2} for future-generation electronics due to its remarkable mechanical,^{3,4} electrical,^{5,6} optical,^{7,8} and thermal^{9,10} properties. The ever-miniaturizing electronic devices lead to ever increasing power density, which motivates a growing need for high performance thermal management in such devices. The high thermal conductivity^{11,12} of graphene makes it extremely attractive as a possible solution for next generation thermal management devices. On the other hand, thermal conductivity is sensitive to defect concentration, and the fabrication process of graphene inevitably results in defects such as vacancies,^{13,14} Stone-Wales defects,^{15,16} isotope effects,¹⁷⁻²¹ wrinkles,²² grain boundaries (GBs),^{23,24} and GB loops,²⁵ which in turn influence the thermal properties of graphene substantially.

Currently, chemical-vapor deposition (CVD) is the most common approach to fabricating large-area monolayer graphene on metal foils such as nickel^{26,27} and copper.^{28,29} However, the CVD approach invariably creates defects such as

GBs in graphene³⁰ because individual crystals nucleate on each grain of the metallic foil and then randomly grow in different directions. Such crystals coalesce at the later stage of grain growth to form graphene GBs^{31,32} which are usually modeled in pairs of pentagon and heptagon rings.² Graphene GBs have been studied both experimentally^{23,3} ⁴ and theoretically^{35,36} and shown to significantly degrade many properties of graphene, such as electrical transport³⁷ and thermal conductivity.38 ⁴⁰ Several investigations perform defect engineering to modulate the thermal conductivity by tuning GB defect density,^{38,41,42} GB energy,⁴³ and material size.^{44,45} Molecular dynamics (MD) simulations reveal that different symmetric tilt GBs exhibit an anomalous expansion in thermal conductivity along GBs of bicrystalline graphene nanoribbons.⁴⁶ The thermal transport in polycrystalline graphene is governed by the thermal conductivity of graphene GBs along various in-plane directions, given the random orientations of GBs in polycrystalline graphene. However, existing studies consider the cases in which the thermal flux is either parallel or perpendicular to graphene tilt GBs. A comprehensive understanding of the thermal conductivity of



FIG. 1. Schematic of (a) the misorientation angle (θ) between 2 graphene grains and (b) the thermal loading angle (ϕ).

graphene tilt GBs as a function of in-plane thermal loading angle and GB misorientation angle is desirable but still absent.

Aiming to address this gap, in the present study, we investigate the thermal conductivity (k) of graphene GBs for all possible tilt GB misorientation angles (23 cases) under arbitrary in-plane thermal loading directions, using non-equilibrium molecular dynamics (NEMD)^{22,41,46,47} simulations.

II. METHODOLOGY

We consider a graphene tilt GB between two crystalline grains having a misorientation angle θ [Fig. 1(a)]. The thermal loading angle ϕ [Fig. 1(b)] is defined as the angle between the tilt GB and the direction perpendicular to the thermal flux. Therefore, when $\phi = 0^{\circ}$, the heat flux is perpendicular to the graphene tilt GB, and when $\phi = 90^{\circ}$, the heat flux is parallel to the graphene tilt GB.

Figure 2 shows all possible graphene tilt GB configurations (23 in total) grouped into three categories termed as armchair [Fig. 2(a)], transition [Fig. 2(b)], and zigzag [Fig. 2(c)] oriented GBs. Each configuration comprises of an arrangement of one pentagon and one heptagon ring of carbon atoms, both of which together constitute a disclination dipole (dp). For the range of misorientation angle θ from 2.1° to 21.8°, the GB features a periodic array of disclination dipoles (1dp) made of a pentagon-heptagon defect or dipole clusters (made of 2 or 3 dipoles, e.g., 2dp, 3dp) [Fig. 2(a)]. GBs in this category are termed as armchair oriented GBs (AC GBs). For the range of θ from 32.2° to 54.3°, the GB features a periodic array of disclination clusters made of a pentagon-heptagon-pentagonheptagon defect [Fig. 2(c)]. GBs in this category are termed as



FIG. 2. The plots of possible tilt GB configurations (23 in total), showing (a) the armchair (AC) GBs with a misorientation angle (θ) ranging from 2.1° to 21.8°, (b) transition GBs with θ from 23.3° to 30.2°, and (c) zigzag (ZZ) GBs with θ ranging from 32.2° to 54.3°. GB structures are modeled in clusters of pentagon (red) and heptagon (blue) membered rings as shown in each of the plots.

zigzag oriented GBs (ZZ GBs). For the intermediate range of misorientation angle from 23.3° to 30.2°, the GBs show mixed features of AC and ZZ GBs, termed as transition GBs [Fig. 2(b)]. This is the second part of our previous study⁴⁸ which used the same atomic coordinates for the GBs.

We use the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS)⁴⁹ for the molecular dynamics study of the strength of graphene GBs. The size of the simulation model, on the one hand, needs to be large enough to contain sufficient number of defects along the GB and, on the other hand, needs to be suitable so that parametric studies of GB properties are not computation time prohibitive. For such considerations, in all simulations, polycrystalline graphene is 300 Å×200 Å. First, the entire model structure is energy minimized, allowing the structure to relax to the lowest energy state and ensuring stable bonding among all the atoms. The optimized Tersoff potential⁵⁰ was used to model the interactions between C-C atoms in graphene, because it has been recently shown to produce values of the acousticphonon velocities that are in excellent agreement with the measured data for pristine graphene. The original Tersoff potential is made up of 12 parameters for carbon based systems and the optimized potential used for these simulations changed two of these parameters. Each of the polycrystalline structures is initialized at a temperature of 300 K using the canonical (NVT) ensemble [coupling time 0.1 picosecond (ps)] for 1.5 ps. Subsequently, the microcanonical ensemble (NVE) using the Berendsen thermostat is employed for a further 1 ps to equilibrate the system in the absence of imposed constraints. A time step of 0.0005 ps is used throughout in all the simulations. Here, we assume the validity of ergodic hypothesis, which states that the time average will be equal to the ensemble average.

To investigate the thermal transport of polycrystalline graphene, we apply the non-equilibrium molecular dynamics (NEMD) method³⁸ along the in-plane (x direction) using the NVT ensemble for 16 000 ps. There also exist other methods to determine k, such as Green's function techniques using phonon properties that involve the solution of solving lattice dynamics (LD) and Boltzmann Transport (BT) equations.⁵⁷ Green's function techniques are usually applicable to low temperatures (below the Debye temperature) and involve a rather small model system (e.g., up to 2000 atoms in Ref. 51) in comparison with that using MD techniques (e.g., around 20 000 atoms in our simulations). The atomic model is such that the boundaries are held fixed at two different temperatures of 320 K (left boundary) and 280 K (right boundary). The atomic positions and velocities of the hot zone at the left end and the cold zone at the right end are updated for each time step using the NVT ensemble (Nose-Hoover thermostat) in LAMMPS [Fig. 3(a)]. For the rest of the atoms in the central region, which are mobile, an NVE ensemble is used. This temperature difference initiates a kinetic energy exchange mechanism driving the heat transport process through the material model. After a steady state temperature distribution is achieved after 10 ps, the constant heat flux (J_x) through the material and the temperature gradient can be computed.



FIG. 3. (a) A representative schematic of a graphene nanostructure containing a tilt GB with a misorientation angle $\theta = 21.8^{\circ}$ and a thermal loading angle $\phi = 10^{\circ}$. In the atomic model of the graphene sheet, the left end (red) is set at a temperature higher than the right end (blue) which drives the heat transfer process across the material and thus imposes a heat flux J_x from left to right. (b) Steady-state temperature distribution along the heat transfer direction for the graphene sheet in (a). The spatial distribution of the temperature of the regions containing the two grains along with the GB zone is used to determine the best linear fit (red line) to evaluate the temperature gradient and hence compute the thermal conductivity of the graphene tilt GB along this specific thermal loading direction.

Then, we continue additional NEMD simulations to calculate the spatially distributed temperature values along with the average cumulative energy (E_{cum}) transferred between the hot and cold zones which is given by

$$E_{cum} = (E_{hot} - E_{cold})/2, \qquad (1)$$

where E_{hot} is the cumulative energy supplied to the hot region and E_{cold} is the cumulative energy extracted from the cold region and is in units of eV. This difference in energy at the two boundaries is thus directly proportional to J_x , which subsequently imposes the temperature gradient dT/dx. Finally, the thermal conductivity (k) is computed from 1D



FIG. 4. Thermal conductivity of graphene GBs as a function of thermal loading angle for (a) armchair GBs, (b) transition GBs, (c) zigzag GBs, and (d) pristine sheets. Error bars represent the standard deviations computed from ten simulations.

Fourier's law as

 $k = \frac{J_x}{dT/dx},$ (2)

where

$$J_x = \left(\frac{dE_{cum}}{dt}\right) / A.$$
(3)

Here, A is the cross-sectional area across which heat transfer occurs. For polycrystalline graphene, it is the rectangular area equal to the width of the sheet times the interlayer spacing of graphite, which equals 0.34 nm. The temperature gradient is determined by a linear fit algorithm of the spatial temperature distribution of the inner region of the graphene sheet. The extreme ends of the sheet where the temperature profile is nonlinear, due to finite size effects and the high thermal conductivity of graphene,³⁸ are excluded from this calculation. Figure 3(b) shows a typical steady state temperature profile in the simulation model.

III. RESULTS AND DISCUSSIONS

Figure 4 presents the variation of thermal conductivity k as a function of thermal loading angle ϕ for all the 23 cases of graphene tilt GBs (armchair GBs, Fig. 4(a); transition GBs, Fig. 4(b); zigzag GBs, Fig. 4(c); and pristine graphene, Fig. 4(d)]. It is obvious that k exhibits an overall increasing trend as ϕ increases. This trend can be readily understood as follows. The thermal transport in polycrystalline graphene is mainly dominated by phonons^{52,53} and has negligible electronic contribution.⁵⁴ With higher defect density, the nanomaterial aggravates the phonon-phonon scattering in addition to shortening the phonon mean free path, leading to hindrance in the formation of vibrational modes responsible for the thermal transport, which consequently reduces k. As thermal loading angle ϕ increases from 0° to 90°, the orientation of the GB gradually varies from being perpendicular to being parallel to the thermal flux direction (Fig. 5). As ϕ increases, the perturbation to thermal transport due to the disclination dipole defects in the GB decreases, leading to an increasing k.

To further understand the parameters that govern the variation of k, Fig. 6 plots k as a function of misorientation angle θ for various thermal loading angle ϕ . It is evident that for a given ϕ , k reaches the lowest at θ = 32.2°. We suggest that



FIG. 5. Graphene sheets with a GB with (a) $\phi = 0^{\circ}$, (b) an intermediate ϕ , and (c) $\phi = 90^{\circ}$.

ARTICLE

this phenomenon is dependent on the defect density. Figure 1 shows that the defect density along the GB gradually increases as θ increases for armchair GBs [Fig. 1(a)] and transition GBs [Fig. 1(b)]. The highest defect density occurs at $\theta = 32.2^{\circ}$, corresponding to the lowest *k*. For zigzag GBs, the defect density decreases as θ increases from 32.2° [Fig. 1(c)].

Such a dependence of thermal conductivity on defect density can be further understood mechanistically as follows. There are two types of vibrational modes that are responsible for the conduction of thermal energy, namely, the low frequency (low wave number) long-range modes¹ and the high frequency, short-range stretching modes.55 Even though there is an ongoing debate to identify the exact vibrational mode which has a dominant contribution in the thermal transport of these carbon nanomaterials, it is clear that the influence of mass disorder from the defects along the GB results in the perturbation of these vibrational modes. This is because when any mass disorder is present in the nanomaterial, increased phonon-phonon interactions at different frequencies occur which facilitates increased energy scattering and thereby reduces the phonon velocities along with the energy carrying capacity of the phonons. As a result, the thermal conductivity of graphene decreases with increased defect density.

It is also evident from Fig. 6 that, for any given thermal loading angle, the dependence of *k* on misorientation angle θ is rather non-linear, with a sharp variation of *k* when θ deviates from 0 Å or 60 Å and a rather gradual variation of *k* for an intermediate range of θ (near 32.2 Å). Such a non-linear dependence can be possibly attributed to the defect-induced fluctuation of the graphene morphology. Defects such as the



FIG. 6. Thermal conductivity as a function of misorientation angle θ for various thermal loading angles ϕ starting from 0° to 90°.



FIG. 7. Defect-induced out-of-the-plane fluctuation in graphene. (a) θ = 5.5°; (b) θ = 32.2°; (c) θ = 50.5°. Top: perspective view; middle: top view; bottom: side view.

disclination dipoles along the GB cause lattice distortion near the defects, which in turn lead to out-of-the-plane fluctuation of the graphene. With such fluctuation of the graphene, the corresponding change in bond lengths and bond angles leads to instability in the lattice and phonon scattering, which consequently cause reduced energy carrying capacity and thus reduce thermal conductivity.⁵⁶ As shown in Fig. 7, GBs with a low defect density indeed result in out-of-the-plane fluctuation with an overall amplitude larger than that due to a GB with a high defect density. For example, for $\theta = 32.2$ Å the fluctuation amplitude is only about 7.6 Å, while for $\theta = 5.5$ Å and $\theta = 50.5$ Å, the fluctuation amplitude is about 20.5 Å and 16.6 Å, respectively. This can be attributed to the mutual constraint of out-of-the-plane fluctuations between neighboring disclination dipole defects. As a result, the reduction in k from the increase in defect density is offset by the reduction of the out-of-the-plane fluctuation of polycrystalline graphene as θ increases from 0 Å (or decreases from 60 Å), leading to the non-linear dependence as depicted in Fig. 6.

There typically will be a temperature jump between regions with differing thermal properties and we have also seen a sharp jump in temperature between Grain 1 and Grain 2 for $\phi = 20^{\circ}$ or less. The thermal conductivity for these two grains can also be quite different, as shown in Table I, since the chirality of Grain 1 and Grain 2 is different. However, even when *k* between Grain 1 and Grain 2 is similar, the temperature jump is about the same as when the difference in *k* is large. This implies that the temperature jump occurs mainly because of the GB region itself. This temperature jump produces a resistance to the heat flow. The Kapitza resistance, R, was determined for a few sheets, as shown in Table I, using

$$\mathbf{R} = \frac{\Delta T}{\mathbf{J}_x},$$

where ΔT is the temperature jump between Grain 1 and Grain 2 as shown in Fig. 8, and J_x is the heat flux through the entire sheet.

TABLE I.	Kapitza	resistance,	R	and	thermal	conductivity	k,	of	Grain	1 and	Grai	n 2	for	several	shee	ets
----------	---------	-------------	---	-----	---------	--------------	----	----	-------	-------	------	-----	-----	---------	------	-----

		φ = 0°		φ = 10°					
θ (°)	9.4	21.8	44.8	9.4	21.8	44.8			
k for Grain 1 (W/m K) k for Grain 2 (W/m K) ΔT (K) Heat flux J _x (W/m ²) Kapitza resistance R [K/(W/m ²)]	349 356 9.9 1.80 × 10 ¹¹ 5.60 × 10 ⁻¹¹	200 254 8.8 1.53 × 10 ¹¹ 5.80 × 10 ⁻¹¹	238 231 9.7 1.60 × 10 ¹¹ 6.10 × 10 ⁻¹¹	164.9 197 5 1.70 × 10 ¹¹ 3.00 × 10 ⁻¹¹	194 126 7.5 1.50 × 10 ¹¹ 5.10 × 10 ⁻¹¹	135 117 6.5 1.50 × 10 ¹¹ 4.40 × 10 ⁻¹¹			
		φ = 20°		φ = 30°					
θ (°)	9.4	21.8	44.8	9.4	21.8	44.8			
k for Grain 1 (W/m K) k for Grain 2 (W/m K) ΔT (K) Heat flux J _x (W/m ²) Kapitza resistance R [K/(W/m ²)]	155 128 3.8 1.60 × 10 ¹¹ 2.50 × 10 ⁻¹¹	82.5 105 3.1 1.30 × 10 ¹¹ 2.30 × 10 ⁻¹¹	109 104 3.5 1.40 × 10 ¹¹ 2.50 × 10 ⁻¹¹	199 111 1.7 1.60 × 10 ¹¹ 1.10 × 10 ⁻¹¹	100 109 1.2 1.40 × 10 ¹¹ 9.00 × 10 ⁻¹²	115 107 1.1 1.50 × 10 ¹¹ 6.80 × 10 ⁻¹²			



FIG. 8 . The temperature jump, ΔT , between Grain 1 and Grain 2 for a sheet with $\phi = 10^{\circ}$ and $\theta = 21.8$ Å.

Table I also shows that *k* for Grain 1 and Grain 2 is slightly larger when ϕ is small, then converges to the same *k* for the entire sheet as ϕ gets larger. The Kapitza resistance gets very small after $\phi = 20^{\circ}$, since ΔT becomes small after this and the temperature profile becomes linear through the entire sheet (excluding the nonlinear ends near the hot and cold zones).

Size effects are important as the NEMD approach employs finite-size simulation cells, which results in artificial scattering at the thermostat boundaries (the left and the right ends which are fixed at 320 K and 280 K, respectively) and at the GBs. As shown in Fig. 9, for a given width of the simulation model of 200 Å, *k* increases as the length of the simulated sheet increases. Such a trend can be explained as follows. The phonon mean free path of graphene is in the order of ~775–800 nm.^{38,57} When the nanostructure length (L) is smaller than the effective mean free path, the thermal transport is of ballistic type where the energy scattering due to phonon collisions in the boundary and the GBs dominates. With gradual increase in the model system size (length of graphene), the energy losses due to phonon scattering in the boundary and the GBs decrease and the phonons now have a greater likelihood of colliding against each other. The more the



FIG. 9. (a) Effect of sheet size on *k* for several θ 's, where it can be seen there is a significant increase in *k* for longer sheet sizes. For each sheet, $\phi = 10^{\circ}$. (b) Inverse of thermal conductivity (*k*) with respect to the inverse of length of graphene (*L*). The intercepts for $\theta = 9.4$ Å, $\theta = 21.8$ Å, $\theta = 32.2$ Å, and $\theta = 44.8$ Å are 0.0024, 0.0026, 0.0032, and 0.0021, respectively.

phonon-phonon collisions with each other, the more is the energy carrying capacity of the phonons which directly contribute to a higher k. This explains the increase in values of thermal conductivity with larger system sizes. This increase continues even when the thermal transport transitions from ballistic type to diffusive type (L>800 nm). However, an increase in k with an increase in the model system size is not unbounded. Values of k for defect-free graphene have been found to converge to around 3122 W/mK for graphene sheet lengths of $16\,\mu\text{m}$ as in the previous literature.⁵⁷ Such a large length-scale is outside the scope of our study. However, from the intercepts of the plots in Fig 9(b), we observe that the values of k for infinitely long graphene sheets are 416. 66 W/m K(for $\theta = 9.4$ Å), 384.62 W/m K (for $\theta = 21.8$ Å), 312.5 W/m K (for $\theta = 32.2$ Å), and 476.19 W/m K (for $\theta = 44.8$ Å). The results from Ref. 38 predict k for infinitely long pristine graphene along the ZZ direction to be around 2650 W/mK, which is also close to 3122 W/m K found in Ref. 57. Ours results show that k is lower because of the GB. For our simulations that analyzed size effects, the sheet sizes varied from lengths of 250 Å to 500 Å with a constant width of 200 Å, while k varied from 52.7 to 215.6 W/mK. This is in excellent agreement with a previous study that showed that k values are around 175 W/m K for graphene sheets that were 2000 Å×41 Å with similar GBs.⁴⁶ Moreover, we observe that for these sheets with varying lengths, the trend of change in k with changing defect density is the same for the results we presented for sheets that were all $300 \text{ Å} \times 200 \text{ Å}$ above. The maximum k always occurs at $\theta = 9.4 \text{ Å}$, intermediate at $\theta = 44.8$ Å, and the minimum k occurs at θ = 32.2 Å. This trend of change in k is consistent with the results of all the sheets presented in our study. Sheets longer than the ones in this study are also expected to show the same trend of change in *k* till the values converge for infinitely long sheets.

IV. CONCLUSION

In summary, our computational investigation deals with graphene structures having GBs formed from pentagon and heptagon rings and elucidates the dependence of its thermal conductivity on the thermal loading direction and the GB misorientation angle. The comprehensive study provides quantitative knowledge of the thermal conductivity of graphene GBs for the entire range of GB misorientation angles in arbitrary thermal loading directions. It was shown that thermal conductivity generally decreases with an increase in defect density along the GBs and increases with an increase in thermal loading angle or sheet length. The Kapitza resistance of the sheet can be determined because of the temperature jump between Grain 1 and Grain 2 for $\phi = 20^{\circ}$ or less but disappears for $\phi = 30^{\circ}$ or higher. Such knowledge can shed light on understanding the thermal conductivity of polycrystalline graphene and offer guidelines in designing graphene-based devices.

ACKNOWLEDGMENTS

The authors acknowledge the support from the National Aeronautics and Space Administration (NASA) National Institute

of Aerospace Langley Professor Program. The authors acknowledge the University of Maryland supercomputing resources (http://hpcc.umd.edu) made available for conducting the research reported in this paper.

REFERENCES

¹K. S. Novoselov, V. I. Fal'ko, L. Colombo, P. R. Gellert, M. G. Schwab, and K. Kim, Nature 490, 192 (2012).

- ²K. M. F. Shahil and A. A. Balandin, Solid State Commun. 152, 1331 (2012).
- ³C. Lee, X. Wei, J. W. Kysar, and J. Hone, Science 321, 385 (2008).

⁴D. Akinwande, C. J. Brennan, J. S. Bunch, P. Egberts, J. R. Felts, H. Gao, R. Huang, J. S. Kim, T. Li, Y. Li, K. M. Liechti, N. Lu, H. S. Park, E. J. Reed, P. Wang, B. I. Yakobson, T. Zhang, Y. W. Zhang, Y. Zhou, and Y. Zhu, Extrem. Mech. Lett. 13, 42 (2017).

- ⁵A. K. Geim and K. S. Novoselov, Nat. Mater. 6, 183 (2007).
- ⁶J. K. Wassei and R. B. Kaner, Mater. Today **13**, 52 (2010).

7R. R. Nair, P. Blake, A. N. Grigorenko, K. S. Novoselov, T. J. Booth, T. Stauber, N. M. R. Peres, and A. K. Geim, Science 320, 1308 (2008).

8X. Xu, N. M. Gabor, J. S. Alden, A. M. Van Der Zande, and P. L. McEuen, Nano Lett. 10, 562 (2010).

9S. Ghosh, W. Bao, D. L. Nika, S. Subrina, E. P. Pokatilov, C. N. Lau, and A. A. Balandin, Nat. Mater. 9, 555 (2010).

10 D. L. Nika, E. Pokatilov, and A. Balandin, Phys. Status Solidi (B) 248, 2609

(2011). ¹¹A. Balandin, S. Ghosh, W. Bao, I. Calizo, D. Teweldebrhan, F. Miao, and C. N. Lau, Nano Lett. 8, 902 (2008)

12S. Ghosh, I. Calizo, D. Teweldebrhan, E. P. Pokatilov, D. L. Nika, A. A. Balandin, W. Bao, F. Miao, and C. N. Lau, Appl. Phys. Lett. **92**, 151911 (2008). ¹³J. Hu, X. Ruan, and Y. P. Chen, Nano Lett. **9**, 2730 (2009).

- 14B. Mortazavi and S. Ahzi, Carbon 63, 460 (2013).
- ¹⁵T. Y. Ng, J. J. Yeo, and Z. S. Liu, Carbon **50**, 4887 (2012).

¹⁶J. Haskins, A. Kinaci, C. Sevik, H. Sevinçli, G. Cuniberti, and T. Cagin, ACS Nano 5, 3779 (2011).

¹⁷U. Ray and G. Balasubramanian, Chem. Phys. Lett. **599**, 154 (2014).

¹⁸S. Srinivasan, U. Ray, and G. Balasubramanian, Chem. Phys. Lett. 650, 88

(2016). ¹⁹S. Broderick, U. Ray, S. Srinivasan, K. Rajan, and G. Balasubramanian, ppl. Phys. Lett. 104, 243110 (2014).

²⁰H. Zhang, G. Lee, A. F. Fonseca, T. L. Borders, and K. Cho, J. Nanomater. 2010. 1 (2010).

²¹S. Chen, Q. Wu, C. Mishra, J. Kang, H. Zhang, K. Cho, W. Cai, A. A. Balandin, and R. S. Ruoff, Nat. Mater. 11, 203 (2012).

²²C. Wang, Y. Liu, L. Li, and H. Tan, Nanoscale 6, 5703 (2014).

23P. Y. Huang, C. S. Ruiz-Vargas, A. M. van der Zande, W. S. Whitney, M. P. Levendorf, J. W. Kevek, S. Garg, J. S. Alden, C. J. Hustedt, Y. Zhu,

J. Park, P. L. McEuen, and D. A. Muller, Nature 469, 389 (2011).

²⁴Z. L. Li, Z. M. Li, H. Y. Cao, J. H. Yang, Q. Shu, Y. Y. Zhang, H. J. Xiang, and X. G. Gong, Nanoscale 6, 4309 (2014).

²⁵N. Khosravian, M. K. Samani, G. C. Loh, G. C. K. Chen, D. Baillargeat, and B. K. Tay, Comput. Mater. Sci. 79, 132 (2013).

26 K. S. Kim, Y. Zhao, H. Jang, S. Y. Lee, J. M. Kim, K. S. Kim, J.-H. Ahn, P. Kim, J.-Y. Choi, and B. H. Hong, Nature 457, 706 (2009).

27Q. Yu, J. Lian, S. Siriponglert, H. Li, Y. P. Chen, and S. S. Pei, Appl. Phys. Lett. 93, 113103 (2008).

28X. Li, W. Cai, J. An, S. Kim, J. Nah, D. Yang, R. Piner, A. Velamakanni, I. Jung, E. Tutuc, S. K. Banerjee, L. Colombo, and R. S. Ruoff, Science 324, 1312 (2009)

²⁹M. P. Levendorf, C. S. Ruiz-Vargas, S. Garg, and J. Park, Nano Lett. 9, 4479 (2009).

 ³⁰S. N. Raja, D. Osenberg, K. Choi, H. G. Park, and D. Poulikakos, Nanoscale 9, 15515 (2017).

³¹Q. Yu, L. A. Jauregui, W. Wu, R. Colby, J. Tian, Z. Su, H. Cao, Z. Liu, D. Pandey, D. Wei, T. F. Chung, P. Peng, N. P. Guisinger, E. A. Stach, J. Bao, S. S. Pei, and Y. P. Chen, Nat. Mater. 10, 443 (2011).

- 32H. Wang, G. Wang, P. Bao, S. Yang, W. Zhu, X. Xie, and W. J. Zhang, J. Am. Chem. Soc. 134, 3627 (2012).
- ³³B. I. Yakobson and F. Ding, ACS Nano 5, 1569 (2011).
- 34K. Kim, Z. Lee, W. Regan, C. Kisielowski, M. F. Crommie, and A. Zettl, ACS Nano 5, 2142 (2011).
- ³⁵R. Grantab, V. B. Shenoy, and R. S. Ruoff, Science **330**, 946 (2010).
- ³⁶O. V. Yazyev and S. G. Louie, Phys. Rev. B 81, 1 (2010).
- ³⁷H. Zhang, G. Lee, C. Gong, L. Colombo, and K. Cho, J. Phys. Chem. C 118, 2338 (2014).
- ³⁸A. Bagri, S. P. Kim, R. S. Ruoff, and V. B. Shenoy, Nano Lett. **11**, 3917 (2011).
- 39T. B. Limbu, K. R. Hahn, F. Mendoza, S. Sahoo, J. J. Razink, R. S. Katiyar, B. R. Weiner, and G. Morell, Carbon 117, 367 (2017).
- 40 M. Shavikloo and S. Kimiagar, Comput. Mater. Sci. 139, 330 (2017).
- ⁴¹P. Wang, B. Gong, Q. Feng, and H. T. Wang, Acta Mech. Sin. 28, 1 (2012).
- ⁴²H. Y. Cao, H. Xiang, and X. G. Gong, Solid State Commun. **152**, 1807 (2012).
- 43H. K. Liu, Y. Lin, and S. N. Luo, J. Phys. Chem. C 118, 24797 (2014).

- 44A. Y. Serov, Z.-Y. Ong, and E. Pop, Appl. Phys. Lett. 102, 033104 (2013).
- ⁴⁵S. Tang and Y. Kulkarni, Appl. Phys. Lett. **103**, 213113 (2013).
- ⁴⁶T. H. Liu, S. C. Lee, C. W. Pao, and C. C. Chang, Carbon **73**, 432 (2014).
- 47A. Cao and J. Qu, J. Appl. Phys. 112, 013503 (2012).
- 48A. Fox, U. Ray, and T. Li, Carbon 142, 388 (2019).
- ⁴⁹S. Plimpton, J. Comput. Phys. **117**, 1 (1995).
- ⁵⁰L. Lindsay and D. A. Broido, Phys. Rev. B **81**, 205441 (2010).
- ⁵¹L. M. Sandonas, H. Sevinçli, R. Gutierrez, and G. Cuniberti, Adv. Sci. 5 (2018).
- ⁵²A. A. Balandin, Nat. Mater. **10**, 569 (2011).
- ⁵³D. L. Nika and A. A. Balandin, J. Phys. Condens. Matter **24**, 233203 (2012). ⁵⁴A. Glatz and I. S. Beloborodov, Phys. Rev. B 80, 245440 (2009).
- ⁵⁵J. Turney, E. Landry, A. McGaughey, and C. Amon, Phys. Rev. B **79**, 064301 (2009).

⁵⁶N. Wei, L. Xu, H. Q. Wang, and J. C. Zheng, Nanotechnology 22, 105705 (2011). ⁵⁷M. Park, S. C. Lee, and Y. S. Kim, J. Appl. Phys. **114**, 053506 (2013).