Dimensional crossover of thermal conductance in graphene nanoribbons: a first-principles approach

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Abstract
First-principles density-functional calculations are performed to investigate the thermal transport properties in graphene nanoribbons (GNRs). The dimensional crossover of thermal conductance from one to two dimensions (2D) is clearly demonstrated with increasing ribbon width. The thermal conductance of GNRs of a few nanometers width already exhibits an approximate low-temperature dependence of $T^{1.5}$, like that of 2D graphene sheets which is attributed to the quadratic nature of the dispersion relation for the out-of-plane acoustic phonon modes. Using a zone-folding method, we heuristically derive the dimensional crossover of thermal conductance with the increase of ribbon width. Combining our calculations with the experimental phonon mean-free path, some typical values of thermal conductivity at room temperature are estimated for GNRs and for 2D graphene sheet. Our findings clarify the issue of the low-temperature dependence of thermal transport in GNRs and suggest a calibration range of thermal conductivity for experimental measurements in graphene-based materials.

1. Introduction

Since the first exfoliation of graphene [1], many exotic properties have been discovered in two-dimensional graphene sheets [2]. Besides graphene, the electronic states of graphene nanoribbons (GNRs) [3–6], thin strips of graphene, can be tuned through controlling the ribbon width [3, 4] and the edge chirality [5, 6]. Graphene-based materials can be a technological alternative to silicon semiconductors because of their unique properties [1–6]. Apart from its electronic properties, graphene turns out to be an excellent heat-conduction material [7–21]. However, some discrepant values of thermal conductivity are experimentally reported. Thermal conductivities for suspended single-layer graphene flakes were first measured [8] using Raman spectroscopy with values in the range from $4.84 \pm 0.44 \times 10^3$ to $5.30 \pm 0.48 \times 10^3$ W m$^{-1}$ K$^{-1}$ at room temperature. Using a similar Raman scattering method or utilizing the conventional heat bath method, successive measurements on graphenes [7–21] have reported thermal conductivities varying from $\sim 170$ W m$^{-1}$ K$^{-1}$ [17] to $\sim 5000$ W m$^{-1}$ K$^{-1}$ [9, 12]. A monolayer of graphene in contact with silicon dioxide has a thermal conductivity of about 600 W m$^{-1}$ K$^{-1}$ [19]. The room-temperature thermal conductivity [20] decreases from 2800 to 1300 W m$^{-1}$ K$^{-1}$ as the number of atomic planes increases from two to four
in few-layer graphenes. Some different theoretical results for the thermal conductivity with various approaches have also appeared [23–30].

Furthermore, there is another confusing aspect with regard to the low-temperature dependence of thermal transport in two-dimensional graphenes. For one-dimensional quantum atomic chains [22], the thermal conductance at low temperatures is proportional to the temperature \( T \) with the quantized universal coefficient \( \pi^2 k_B^2 / 3h \), where \( k_B \) is the Boltzmann constant and \( h \) is the Planck constant. For two-dimensional materials, it is well known that the thermal conductance depends on temperature as the Debye \( T^2 \) law. A calculation by the valence-force field demonstrates that thermal conductivities at low temperature conform [23] to the conventional relation of \( T^2 \). However, a theoretical estimation [24] and the continuum model [25] propose a \( T^{1.5} \) temperature dependence of the thermal conductivity for graphene, followed by some recent experimental observations [15–18].

An understanding of the thermal transport in graphene-based materials is not only important for the possible technological applications but also crucial for a fundamental understanding of the thermal properties in low-dimensional systems. The reduced-dimension effects on thermal transport are becoming critical to both device reliability and intrinsic physics. The temperature dependence of thermal transport, especially in the low-temperature range, is essential for understanding the fundamental physics behind the measured thermal conductivities. An accurate knowledge of the temperature dependence of thermal transport is important because it helps to identify the phonon branches that have the dominant contributions to heat conduction. The low-frequency acoustic phonon branches which determine the behavior of low-temperature thermal transport are sensitive to the weak-bonding interactions between the long-range atoms. The valence-force field considering only the nearest neighbors or the continuum model may not be an appropriate model. Moreover, experimental measurements of temperature dependence alone cannot provide evidence in favor of one or another phonon contribution because the temperature dependence in graphite depends strongly on the material quality [7]. In contrast, the first-principles approach can yield accurate phonon dispersion [32] relations without empirical parameters, including the long-range interactions among the atoms.

In this paper, we report a systematic investigation on the temperature dependence of thermal transport in graphenes varying from quasi one-dimensional nanoribbons to infinite two-dimensional sheets via first-principles density-functional theory calculations [31]. The dimensional crossover of thermal conductance from one to two dimensions is clearly observed in GNRs with increasing ribbon width. We find that the thermal conductance of GNRs of a few nanometers wide already exhibits an approximate low-temperature dependence of \( T^{1.5} \). The ballistic thermal conductance of wide ribbons is also found to converge towards the corresponding values for 2D graphene sheet at low temperature. Combining our calculations with the experimental phonon mean-free path (MFP) [8], typical values of the thermal conductivity are estimated at room temperature for GNRs and 2D graphene sheet, respectively. We conclusively clarify the issue of the low-temperature dependence of thermal conductance for graphene-based materials and suggest a calibration range of thermal conductivity for experimental measurements. We anticipate that our first-principles results will provide some valuable insights into the experimental measurement of thermal transport in graphene-based materials.

2. Computational methods and formulas

To calculate the thermal conductance, we should obtain force constants in graphene nanoribbons and in 2D graphene sheet. The nanoribbons are classified [32] by the number of dimers in the unit cell using a common convention, where armchair-edged nanoribbons or zigzag-edged nanoribbons with \( n \) dimers are referred to as \( n \)-AGNRs or \( n \)-ZGNRs. To eliminate the dangling bonds at the edges of the ribbons, we passivated the nanoribbon edges with hydrogen atoms. The interatomic force constants are obtained from a series of supercell calculations using the software package SIESTA [31] based on the first-principles density-functional approach. The ribbons are periodic with a supercell of five unit cells along the ribbon axis to minimize the periodic continuation. In the other two directions, the periodic image of the ribbon is separated from its nearest neighbors by at least 18 Å to prevent interactions between them. We use a \( 7 \times 7 \times 1 \) supercell for calculations of the graphene sheet, with a space of 20 Å between periodic images of the sheet. For DFT calculations, Troullier–Martins pseudopotentials are used for both C and H atoms. The valence electrons are described by a double-\( \zeta \) basis set plus polarization orbits (DZP). The local-density approximation (LDA) with the exchange–correlation functional due to Ceperley–Alder is employed. To suppress the force fluctuations, we use a rather fine mesh cutoff of 400 Ryd during calculations.

We first perform geometrical optimization with a force tolerance of 0.001 eV Å\(^{-1}\) with the aforementioned parameters using a conjugate-gradient minimization method. Force constants are then derived from the optimized structures with the method of finite difference, where each atom is displaced from its equilibrium position by a distance of \( \pm 0.0212 \) Å. After the forces are evaluated, a central finite difference with respect to the displacement is utilized to compute the force constants. We have calculated force constants for \( n \)-AGNRs and \( n \)-ZGNRs of \( n = 2, 3, \ldots, 14 \) dimers in width and for the two-dimensional graphene sheet, respectively.

The thermal conductivity \( \kappa \) along one direction, e.g. the \( z \)-axis direction, can be expressed by the well-known Boltzmann–Peierls formula [33]

\[
\kappa_z = \sum_{\mathbf{p}, \mathbf{q}>0} \int \frac{d^2 \mathbf{q}}{(2\pi)^2} \hbar \omega_{\mathbf{p}} \frac{\partial f_B}{\partial T} \mathbf{v}_{\mathbf{p}} \mathbf{v}_{\mathbf{q}},
\]

where \( p \) denotes the polarization of the phonon, \( \mathbf{q} \) is the wavevector, \( \omega_{\mathbf{p}} \) is the angular frequency and \( f_B \) is the
Bose–Einstein distribution at temperature $T$. The phonon group velocity $v_p^g$ along the transport direction $z$ is given by $v_p^g = \partial \omega_p / \partial q_z$, and $l_p$ represents the length of the phonon mean-free path (MFP). The number $n = 1, 2$ and 3 denotes the system dimension. The integration over the wavevector is carried out within the first Brillouin zone. Graphene has a long phonon MFP, which is estimated to be about 775 nm near room temperature [8]. Therefore, at low temperatures, it is reasonably expected that the phonon transport in GNRs or in 2D graphene sheets will be dominantly ballistic or quasi-ballistic rather than diffusive. In the regime of ballistic or quasi-ballistic transport, the thermal conductivity is not well defined because the thermal conductivity $\kappa$ depends on the length of the system. Assuming that the MFP $l_p$ for ballistic or quasi-ballistic transport is independent of the polarization since it can be compared with the length of the system in experiments, we thus rewrite equation (1) using the mean value theorem as

$$\kappa_z = \bar{\sigma} l, \quad (2\alpha)$$

with

$$\sigma \equiv \sum_{p,v_p>0} \int \frac{d^2q}{(2\pi)^2} \frac{\hbar \omega_p}{\partial T} \frac{\partial f_B}{\partial T} v_p^g. \quad (2b)$$

Here $\bar{l}$ is the average length of the phonon MFP, $p$ denotes the polarization and $\sigma$ is defined as the thermal conductance per unit area. In particular, the thermal conductance $\sigma$ in equation (2) for one-dimensional ballistic transport is reduced to the following expression:

$$\sigma_{1D} = \frac{1}{S} \sum_{p,v_p>0} \int \frac{d\mathbf{q}}{2\pi} \frac{\hbar \omega_p}{\partial T} \frac{\partial f_B}{\partial T} v_p^g, \quad (3)$$

where $S = \hbar w$ is the cross-section, $h$ is the thickness and $w$ is the width. In contrast with thermal conductivity, thermal conductance is uniquely defined in the regime of ballistic transport. Here we concentrate on the salient features of the temperature dependence of the thermal transport at low temperature in both GNRs and graphenes such that nonlinear scattering characterized by the MFP is first neglected. The phonon transmission is assumed to be ballistic such that the thermal transport at low temperature is distinguished by the thermal conductance $\sigma$ as defined in equation (2). This is a valid approximation for thermal transport in graphenes at low temperatures, where the length of the MFP is approaching the system size. Nonlinear scattering is later included through the average length of the phonon MFP at the end of the paper.

3. Results and discussion

3.1. Phonon dispersion relations

Since the thermal conductance is intrinsically related to the phonon dispersion relations, we first present the calculated phonon dispersions from first principles. Figure 1 shows the calculated phonon dispersion for the 8-ZGNR and 2D graphene sheet. It can be seen from the figure that the stretching modes due to the C–H bonding in GNRs are located at about 3110 cm$^{-1}$. We can also find that the highest frequency of the longitudinal–optical (LO) mode at the $\Gamma$ point is about 1650 cm$^{-1}$ for both GNRs and the two-dimensional graphene sheet. For GNRs, there are four acoustic phonon branches near the $\Gamma$ point, including an out-of-plane mode (ZA), an in-plane transverse mode (TA), an in-plane longitudinal mode (LA) and a torsion branch that will disappear with increasing width. In comparison, similar acoustic branches except for the torsion one are obtained for the 2D graphene sheet. As shown in figure 1, the ZA mode shows a quadratic energy dispersion around the $\Gamma$-point as a consequence of the point-group symmetry of graphene [34] while the TA and LA modes display a linear dispersion. The phonon dispersion curves for both the GNRs and the 2D graphene sheet are consistent with the previous reported results [32].

The phonon dispersion between the GNRs and the 2D graphene sheet can be mutually transformed through the unfolding of the nanoribbons’ Brillouin zone to that of graphene [32]. The phonon dispersions of the 2D graphene sheet comprise three acoustic branches and three optical branches as indicated in figure 1(b) because there are two carbon atoms in the unit cell of graphene. In GNRs, a group of six modes can be found equivalent to the six phonon modes of graphene, with respect to the phonon eigenvectors near the $\Gamma$-point. Hence, all phonon modes of the GNRs can be interpreted as these six fundamental modes and their overtones [32].

3.2. Thermal conductance of graphene nanoribbons

Using the complete phonon dispersion relations, we have systematically calculated the thermal conductance $\sigma$ per unit area of equation (2) for GNRs with increasing width and for the 2D graphene sheet, as shown in figure 2. During the calculation, the layer thickness $h$ for both the GNRs and the graphene sheet is taken as a typical value $h = 0.335$ nm. The ribbon widths $w$ for the ZGNRs and AGNRs are computed by the formulas $w_{\text{ZGNR}} = \sqrt{3}(n - 1)a_0/2$ and $w_{\text{AGNR}} = (n - 1)a_0/2$, respectively. Here $n$ is the number of dimers in the unit cell and $a_0$ denotes the graphene lattice constant given by $a_0 = 0.246$ nm. For simplicity, a very small deviation of the ribbon width after the structural relaxation is not considered in the formula. Figure 2 shows the thermal conductance per unit area as a function of the temperature for $n$-ZGNRs with an increasing number of dimers $n$. The open circles in figure 2 represent the thermal conductance along the $\Gamma$–K direction, corresponding to the axis direction of the ZGNRs. For a clear display of the low-temperature dependence of thermal conductance, a log–log plot of the thermal conductance for $T < 200$ K is further illustrated in figure 3.
Figure 1. Phonon dispersion relations calculated using first-principles density-functional calculations. (a) The dispersion relation for an 8-ZGNR. The wavenumber $q$ is given in units of the length of the unit cell $T$. (b) The phonon dispersion curves plotted along high-symmetry directions for a two-dimensional graphene sheet.

the value for the two-dimensional graphene sheet $\beta = 1.58$, when the number of dimers for the ZGNRs grows from $n = 2$ to 14. This dimensional crossover of the thermal conductance from one to two dimensions is clearly demonstrated in figure 3(a). It is well known that the thermal conductance at low temperatures is proportional to the temperature $T$ for one-dimensional quantum atomic chains [22] whereas the low-temperature dependence of $T^2$ for the two-dimensional phonon gas is described by the conventional Debye law. For the 2D graphene sheet, a calculation by the valence-force field demonstrates that the thermal conductivity at low temperature conforms [23] to the conventional relation of $T^2$. However, a theoretical estimation [24] proposes a $T^{1.5}$ temperature dependence of the thermal conductance for an infinite 2D graphene sheet. An analysis of vibrational modes [25] using continuum mechanics shows that GNRs with width $w > 500$ nm will show a low-temperature dependence of $T^{1.5}$. In comparison with the approaches of empirical valence-force and continuum mechanics, the density-functional theory can yield accurate phonon dispersion relations without empirical parameters. Our first-principles calculations shown in figure 3(a) clarify that the two-dimensional graphene sheet will display a low-temperature dependence of $T^{1.5}$ and that dimensional crossover of the thermal conductance from one to two dimensions will be expected for GNRs of only a few nanometers in width. We conclude that the thermal conductance for most samples of graphene flakes in experiments with widths beyond a few nanometers will be characterized by a low-temperature dependence of $T^{1.5}$. Our calculated results are consistent with the experimental results [15–18], where the widths of the graphene samples were of the order of micrometers.

Next we elucidate the reasons for the dimensional crossover of the thermal conductance in GNRs through a
Figure 2. Temperature dependence of thermal conductance per unit area for ZGNRs with increase of ribbon width $n$, which indicates the number of dimers per unit cell. The open circles in the figure represent the thermal conductance along the $\Gamma$–$K$ direction, corresponding to the axis direction of the ZGNRs.

heuristic derivation. The phonon wavevector $\mathbf{q} \equiv (q_t, q_l)$ for GNRs consists of the transverse direction component $q_t$ perpendicular to the ribbon axis and the longitudinal component $q_l$ along the ribbon axis. The longitudinal wavevector $q_l$ represented by the $x$-axis in figure 1(a) is continuous because the ribbon length goes to infinity. However, the transverse wavevector $q_t$ is discrete, due to the finite ribbon width which only allows the standing waves with the boundary condition $q_tw = k \cdot \pi$, $k = 0, 1, \ldots, n - 1$. Here $w$ denotes the ribbon width and $n$ is the number of dimers in width. Therefore, the transverse wavevector $q_t$ can only take the quantized values given by $q_t = k\pi/w$. Each discrete value of $q_t$ corresponds to one branch of the over-tone phonon mode [32]. The vibrational modes in GNRs with sufficient width can be classified [32] into six fundamental modes equivalent to the phonon modes in a 2D graphene sheet, over-tone modes of the fundamental modes and the C–H modes resulting from the passivation with hydrogen at the edges. The contribution to the thermal conductance from the C–H modes can be neglected at low temperature on account of their high frequencies. Hence, the summation $p$ in equation (3) over different polarization branches can be separated into the terms of six basic modes plus their over-tone modes, expressed by $\Sigma_p \rightarrow \Sigma_p' \Sigma_{k=0,n-1}$. Here $p'$ denotes the summation over six fundamental phonon modes. When the number of dimers $n$ in the unit cell is large enough, we can transform the discrete summation $k$ into an integration over the continuum $q_t$, mathematically described by $\frac{1}{w} \Sigma_{k=0,n-1} \rightarrow \int dq_t$. In other words, for a nanoribbon with sufficient width, the thermal conductance for one-dimensional transport in equation (3) will transition into the two-dimensional expression $\sigma_{2D} = \frac{1}{h} \Sigma_{p'} \frac{1}{(2\pi)^2} \int dq_t dq_l \hbar \omega_{p'} \frac{\partial f}{\partial T} v_z^{p'}$ with the constraint of $v_z^{p'} > 0$, where $h$ is the thickness. Here $p'$ denotes the basic phonon mode, ranging from the LA, TA, and ZA to the LO, TO and ZO modes shown in figure 1(b) for the 2D graphene sheet. With the increase of ribbon width, this zone-folding method can well reproduce the phonon dispersion between the GNRs and the 2D graphene sheet. From our numerical calculations, we find that the number of dimers $n = 14$ already leads to low-temperature thermal conductance fairly close to that of 2D graphene, demonstrating a dimensional crossover from one to two dimensions.

3.2.2. Temperature dependence of the thermal conductance.

We now turn to the causes of the low-temperature dependence of $T^{1.5}$ in 2D graphene sheet through an analytic derivation. The key reason lies in the different contributions to the thermal conductance made by each of the polarized phonon branches with different dispersion relations. Note that we only need to consider the acoustic branches during calculation of the thermal conductance at low temperature. It can be observed from figure 1(b) that both the LA and the TA branches exhibit linear phonon dispersion relations at low frequencies while the ZA branch displays an approximately

Figure 3. (a) The scaling behavior of thermal conductance with temperature for the 2-ZGNRs, 14-ZGNRs and two-dimensional graphene sheet, respectively. The dashed, dotted, and solid lines in the figure are numerically fitted as $T^\beta$ at low temperatures. (b) The 3D-view and its projection of the phonon dispersion relations for the out-of-plane acoustic branch (ZA) in the first Brillouin zone.
density of states (vDOS), which can be defined as vDOS $\propto \delta(\omega - \omega_p)$. It can be easily verified that the vDOS in two dimensions is proportional to $\omega$ for the quadratic phonon branch and to its constant vibrational density of states. By contrast, the phonon frequency tends to zero, the number of vibrational modes expressed by the vDOS also decreases to zero for low frequencies. At low temperature, only low-frequency phonons mainly contribute to the thermal conductance. Therefore, the ZA phonon branch with the quadratic dispersion makes a dominant contribution to the low-temperature thermal conductance due to its constant vibrational density of states. Namely, the thermal conductance at low temperature for the two-dimensional graphene sheet increases with temperature as $T^{1.5}$ on account of the dominant contributions made by the ZA phonon branch.

3.2.3. Convergence of the low-temperature thermal conductance and anisotropy. We now discuss another interesting feature observed in figure 2. It can be seen from the figure that the thermal conductance of GNRs at low temperatures converges to that of the 2D graphene sheet with the increase of ribbon width. A systematic investigation of the thermal conductance as a function of the ribbon width for the zigzag and armchair graphene nanoribbons at a temperature of $T = 200$ K is shown in figure 4. We can find that the thermal conductance per unit area for both ZGNRs and AGNRs decreases with increasing number of dimers as illustrated in figure 2. At low temperatures, the value of the convergence agrees well with that of 2D graphene sheet along the corresponding direction, as shown by the open circles in figure 2. Such agreement can be explained by the fact that with increasing width the phonon dispersion relations of GNRs approach that of the 2D graphene sheet.
3.3. Nonlinear scattering and thermal conductivity

Finally, we include the nonlinear phonon scattering effects through the phonon MFP. The above calculations have not taken into account phonon–phonon scattering. At high temperatures, nonlinear phonon scattering is inevitable for more realistic situations. However, calculation of the nonlinear phonon scattering for a GNR system with a few hundred of atoms is a formidable task for the first-principles approach and is also beyond the ability of any first-principles implementations. For simplicity, we phenomenologically introduce the phonon–phonon scattering into the present ballistic theory through the phonon MFP described by equation (2). It is estimated from experiments that the phonon MFP at room temperature [8] is about 775 nm on average for graphene samples. At $T = 300$ K, our calculated thermal conductance per unit area for the 2D graphene sheet is $\sigma_{\text{2D}} = 5.5 \times 10^5$ W m$^{-2}$ K$^{-1}$ along the $\Gamma$–K direction and the converged value of $\kappa_{\text{ZGNR}} = 3.4 \times 10^7$ W m$^{-2}$ K$^{-1}$ is obtained for the ZGNRs as shown in figure 2. In comparison with the previous estimation of thermal conductance [24] for 2D graphene at room temperature, $\sigma_{\text{2D}} = 3.1 \times 10^5$ W m$^{-2}$ K$^{-1}$, given by the formula $\sigma = 0.6 \times 10^8 \tau^{3/2}$ W m$^{-2}$ K$^{-5/2}$, our results are qualitatively consistent with this estimation. Combining the calculated thermal conductance with the experimental phonon MFP, the thermal conductivities at room temperature $\kappa_{\text{2D}} = 4263$ W m$^{-1}$ K$^{-1}$ for 2D graphene and $\kappa_{\text{GNR}} = 2635$ W m$^{-1}$ K$^{-1}$ for ZGNRs are achieved. Even though our calculated results cannot be directly compared with experimental results where some other factors exist, such as structure defects, supporting substrate and surface tensions, etc, a calibration range of the thermal conductivity in GNRs and two-dimensional graphene is suggested for experimental measurements from our calculations. We propose that the room-temperature thermal conductivity of GNRs may be estimated to be of the order of $\sim 2600$ W m$^{-1}$ K$^{-1}$ for GNRs and $\sim 4200$ W m$^{-1}$ K$^{-1}$ for 2D graphene sheet if only intrinsic phonon–phonon scattering is considered. We think that the measurement of intrinsic thermal conductivity in GNRs or 2D graphene sheet can be reduced to measuring the accurate phonon mean-free path [35].

4. Conclusions and remarks

In summary, we have investigated the low-temperature thermal transport in GNRs and 2D graphene sheet using a first-principles density-functional theory approach. A dimensional crossover of the thermal conductance from one-dimensional GNRs to two-dimensional graphene sheet is clearly demonstrated. We determine that the thermal conductance of GNRs with a width of a few nanometers will exhibit a low-temperature dependence of $T^{1.5}$. A heuristical derivation of the thermal conductance by the zone-folding method is carried out to elucidate the reason for this dimensional crossover. The reason for the temperature dependence of $T^{1.5}$ can be attributed to the quadratic nature of the dispersion relation for the out-of-plane acoustic phonon branch. In addition, we find that low-temperature thermal conductance in GNRs converges towards the corresponding values for 2D graphene sheet with increase of ribbon width. Combining our calculations with the experimental phonon MFP, typical values for the thermal conductivity for GNRs and 2D graphene sheet are estimated at room temperature. Our findings conclusively clarify the issue of the low-temperature dependence of the thermal conductance for GNRs and suggest a calibration value range of thermal conductivity in graphene-based material. We hope that our findings will offer some enlightening insights into the experimental measurement of thermal transport in graphene-based materials.

Our present conclusions are valid in the low-temperature range, where the thermal transport is dominated by ballistic phonon transmission. For high temperatures, an exact calculation of nonlinear phonon scattering is expected to give the temperature dependence of the phonon mean-free path. We will leave this problem for further pursuit.

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References

[1] Novoselov K S, Geim A K, Morozov S V, Jiang D, Zhang Y, Dubonos S V, Grigorieva I V and Firsov A A 2004 Science 306 666
[2] Neto A H C, Guinea F, Peres N M R, Novoselov K S and Geim A K 2009 Rev. Mod. Phys. 81 109 and references therein
[3] Son Y-W, Cohen M L and Louie S G 2006 Phys. Rev. Lett. 97 216803
[4] Son Y W, Cohen M L and Louie S G 2006 Nature 444 347
[5] Li X, Wang X, Zhang L, Lee S and Dai H 2008 Science 319 1229
[6] Jia X T et al 2009 Science 323 1701
[7] Balandin A A 2011 Nature Mater. 10 569
[8] Ghosh S, Calizo I, Teweldebrhan D, Pokatilov E P, Nika D L, Balandin A A, Bao W, Miao F and Lau C-N 2008 Appl. Phys. Lett. 92 1531911
[9] Balandin A A, Ghosh S, Bao W, Calizo I, Teweldebrhan D, Miao F and Lau C-N 2008 Nano Lett. 8 902
[10] Cai W, Moore A L, Zhu Y, Li X, Chen S, Shi L and Ruoff R S 2010 Nano Lett. 10 1645
[11] Chen S S et al 2011 ACS Nano 5 321
[12] Yu Q et al 2010 ECS Trans. 28 73
[13] Faugeras C, Faugeras B, Orlifta M, Potemski M, Nair R R and Geim A K 2010 ACS Nano 4 1889
[14] Lee J-U, Yoon D, Kim H, Lee S and Cheong H 2011 Phys. Rev. B 83 081419
[15] Pettes M T, Jo I, Yao Z and Shi L 2011 Nano Lett. 11 1195
[16] Wang Z Q, Xie R G, Bui C T, Liu D, Ni X X, Li B W and Thong J L 2011 Nano Lett. 11 113
[17] Xu X et al 2010 arXiv:1012.2937 [cond-mat]
[18] Jiang W, Chen Z, Bao W, Lau C N and Dames C 2010 Nano Lett. 10 3909
[19] Seol J H et al 2010 Science 328 213
[20] Ghosh S, Bao W, Nika D L, Subrina S, Pokatilov E P, Lau C N and Balandin A A 2010 Nature Mater. 9 555
[21] Murali R, Yang Y, Brenner K, Beck T and Meindl J D 2009
    Appl. Phys. Lett. 94 243114

[22] Schwab K, Henrickson E A, Worlock J M and
    Roukes M L 2000 Nature 404 974

[23] Nika D L, Pokatilov E P, Askerov A S and Balandin A A 2009
    Phys. Rev. B 79 155415

[24] Mingo N and Broido D A 2005 Phys. Rev. Lett. 95 096105

[25] Muñoz E, Lu J X and Yakobson B I 2010 Nano Lett. 10 1652

[26] Xu Y, Chen X, Gu B-L and Duan W 2009 Appl. Phys. Lett. 95 233116

[27] Jiang J W, Wang J-S and Li B 2009 Phys. Rev. B 79 205418

[28] Tan Z W, Wang J-S and Gan C K 2011 Nano Lett. 11 214

[29] Hu J, Schiffli S, Vallabhaneni A, Ruan X and Chen Y P 2010
    Appl. Phys. Lett. 97 133107

[30] Evans W J, Hu L and Keblinski P 2010 Appl. Phys. Lett. 96 203112

[31] Soler J M, Artacho E, Gale J D, García A, Junquera J,
    Ordejón P and Sánchez-Portal D 2002 J. Phys.: Condens. Matter 14 2745

[32] Gillen R, Mohr M, Thomsen C and Maultzsch J 2009 Phys. Rev. B 80 155418

[33] Callaway J 1974 Quantum Theory of the Solid State (London: Academic)

[34] Saito R, Dresselhaus G and Dresselhaus M S 1998 Physical
    Properties of Carbons Nanotubes (London: Imperial College Press)

[35] Minnich A J, Johnson J A, Schmidt A J, Esfarjani K,
    Dresselhaus M S, Nelson K A and Chen G 2011 Phys. Rev. Lett. 107 095901