Ballistic to diffusive crossover of heat flow in graphene ribbons

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Heat flow in nanomaterials is an important area of study, with both fundamental and technological implications. However, little is known about heat flow in two-dimensional devices or interconnects with dimensions comparable to the phonon mean free path. Here we find that short, quarter-micron graphene samples reach ~35% of the ballistic thermal conductance limit up to room temperature, enabled by the relatively large phonon mean free path (~100 nm) in substrate-supported graphene. In contrast, patterning similar samples into nanoribbons leads to a diffusive heat-flow regime that is controlled by ribbon width and edge disorder. In the edge-controlled regime, the graphene nanoribbon thermal conductivity scales with width approximately as \( \sim W^{1.8 \pm 0.3} \), being about 100 W m\(^{-1}\) K\(^{-1}\) in 65-nm-wide graphene nanoribbons, at room temperature. These results show how manipulation of two-dimensional device dimensions and edges can be used to achieve full control of their heat-carrying properties, approaching fundamentally limited upper or lower bounds.
he thermal properties of graphene are derived from those of graphite and are similarly anisotropic. The in-plane thermal conductivity of isolated graphene is high, $>2,000 \text{ W m}^{-1}\text{ K}^{-1}$ at room temperature, due to the strong $sp^2$ bonding and relatively small mass of carbon atoms. Heat flow in the cross-plane direction is nearly a 1,000 times weaker, limited by van der Waals interactions with the environment (for graphene) or between graphene sheets (for graphite). Recent studies have suggested that the thermal conductivity of graphene is altered when in contact with a substrate through the interaction between vibrational modes (phonons) of graphene and those of the substrate. However, an understanding of heat-flow properties in nanometre-scale samples of graphene (or any other two-dimensional (2D) materials) is currently lacking.

By comparison, most graphene studies have focused on its electrical properties when confined to scales on the order of the carrier mean free path (mfp) of graphene. For example, these studies have found that ‘short’ devices exhibit near-ballistic behaviour and Fabry–Perot wave interference, whereas ‘narrow’ nanoribbons display a steep reduction of charge-carrier mobility. Previous studies do exist for heat flow in three-dimensional (3D) structures, such as nanowires and nanoscale films. For instance, ballistic heat flow was observed in suspended GaAs bridges and silicon nitride membranes at low temperatures, of the order 1 K. Conversely, suppression of thermal conductivity due to strong edge-scattering effects was noted in narrow and rough silicon nanowires, up to room temperature. Yet, such effects have not been studied in 2D materials like graphene, and ballistic heat conduction has not been previously observed near room temperature in any material.

In this work we find that the thermal properties of graphene can be tuned in nanoscale devices comparable in size to the intrinsic phonon mfp. By ‘intrinsic’ thermal conductivity or phonon mfp, we refer to that in large samples without edge effects, typically limited by phonon–phonon scattering in suspended graphene and by substrate scattering in supported graphene; here $\lambda \approx 100 \text{ nm}$ at room temperature, as we will show. We find that the thermal conductance of ‘short’ quarter-micron graphene devices reaches up to 35% of theoretical ballistic upper limits. However, the thermal conductivity of ‘narrow’ graphene nanoribbons (GNRs) is greatly reduced compared with that of ‘large’ graphene samples. Importantly, we uncover that nanoeengineering the GNR dimensions and edges is responsible for altering the effective phonon mfp, shifting heat flow from quasi-ballistic to diffusive regimes. These findings are highly relevant for all nanoscale graphene devices and interconnects, also suggesting new avenues to manipulate thermal transport in 2D and quasi-one-dimensional systems.

**Results**

**Test structures and measurements.** Figure 1 illustrates several of our experimental test structures, showing graphene and GNR arrays supported on a SiO$_2$/Si substrate (see Methods and Supplementary Note S1). Long, parallel metal lines serve as heater and thermometer sensors, electrically insulated from the graphene by a thin SiO$_2$ layer. We perform heat-flow measurements from 20 to 300 K on unpatterned graphene (Fig. 1a), control samples with the graphene etched off (Fig. 1b) and arrays of GNR widths $W \approx 130$, 85, 65 and 45 nm (Fig. 1c,d and Supplementary Fig. S2). Figure 1f shows the Raman spectra of representative samples, with no discernible D peak (no defects) in unpatterned graphene and a D/G peak ratio of GNRs consistent with the presence of edge disorder.

The measurement proceeds as follows. We pass a heating current through one metal line, which sets up a temperature gradient across the sample, and we monitor changes in electrical resistance of the opposite electrode (see Methods and Supplementary Note S2). Both electrode resistances are calibrated over the full temperature range for each sample, allowing us to convert measured changes of resistance into changes of sensor temperature $\Delta T_s$, as a function of heater power $P_H$ (Supplementary Fig. S5). We also perform measurements after removing the exposed graphene with an oxygen plasma etch (Fig. 1b). This allows us to obtain the thermal properties of the parallel heat-flow path through the contacts, supporting SiO$_2$ and substrate (Supplementary Figs S4 and S8). As a check on our method, we find the thermal conductivity of our SiO$_2$ layer in excellent agreement with well-known data from the literature (Supplementary Note S4 and Supplementary Fig. S8) over the full temperature range. As a result of this exercise, we were also able to fit the thermal resistance of the SiO$_2$–Si interface (Supplementary Fig. S6c and Supplementary Eq. S1), generating one of the few available data sets on this quantity, to our knowledge.

To extract the thermal properties of our samples, we use 3D simulations of the structures with dimensions obtained from measurements by scanning electron microscopy and atomic force microscopy, as shown in Fig. 1d,e and Supplementary Fig. S7. The model matches the measured and simulated $\Delta T_s$ and $P_H$, fitting the thermal conductance $G$ between the heater and the thermometer. The 3D simulations automatically include all known contact-resistance effects, including those of the graphene–SiO$_2$ and SiO$_2$–metal interfaces, matched against data from the literature and our control experiments (Supplementary Note S3). To provide some simple estimates, the contact thermal resistance (per electrode width) is $R_C \approx 0.7 \text{ mK}^{-1}$, the ‘wide’ unpatterned graphene thermal resistance is $R_C \approx 2.5 \text{ mK}^{-1}$ and that of the GNR arrays is in the range $R_{G_{\text{GNR}}} \approx 4–32 \text{ mK}^{-1}$ (from widest to narrowest). The graphene is not patterned under the electrodes; thus, the contact resistance remains the same for all samples. The 3D simulations also account for heat spreading through the underlying SiO$_2$ and our error bars include various uncertainties in all parameters (Supplementary Note S6).

Figure 2a displays in-plane thermal conductance per area $G/A$ for our GNRs, for one of our unpatterned ‘short but wide’ samples ($L = 260 \text{ nm}$, $W = 12 \text{ nm}$), and for the ‘large’ sample ($L = 10 \text{ nm}$) of Seol et al. Here $A$ is the cross-sectional area of heat flow, $A = WH$, where $W$ is the width and $H = 0.335 \text{ nm}$ is the thickness of the graphene samples. At the same time Fig. 3 displays schematics of the size effects and the three transport regimes expected, corresponding to the samples measured in Fig. 2. Figure 2a also shows the theoretical ballistic thermal conductance of graphene, $G_{\text{ball}}/A$, calculated with the approach listed in Supplementary Note S9. By comparison, our ‘short’ sample (schematic in Fig. 3b) has a thermal conductance $\sim 35\%$ of $G_{\text{ball}}/A$ at 200 K and $\sim 30\%$ at room temperature, indicating a regime of quasi-ballistic phonon transport (other similar samples are shown in Supplementary Fig. S9d). In contrast, the ‘large’ sample from Seol et al. (schematic in Fig. 3a) has a conductance per cross-sectional area $< 2\%$ of the ballistic limit, being in the diffusive transport regime as expected ($W, L \gg \lambda$).

**Length dependence of thermal conductivity.** We recall that in the ballistic limit ($L \ll \lambda$), the conductance, rather than the conductivity, approaches a constant at a given temperature $G_{\text{ball}}(T)$. Nevertheless, the thermal conductivity is the parameter typically used for calculating heat transport in practice, and for comparing different materials and systems. Thus, the well-known relationship $k = (G/A)L$ imposes the conductivity $k$ to become a function of length in the ballistic regime and to decrease as $L$ is...
reduced. This situation becomes evident when we plot the thermal conductivity in Fig. 2b, finding $k \approx 320 \, \text{W m}^{-1} \, \text{K}^{-1}$ for our ‘short’ and wide samples at room temperature (schematic Fig. 3b), almost a factor of two lower than the large graphene sample (schematic Fig. 3a). We note that both unpatterned samples here and in Seol et al. were supported by SiO$_2$, showed no discernible defects in the Raman spectra and the results were repeated over three samples (Supplementary Note S5 and Supplementary Fig. S9), with similar results obtained each time.

The transition of 2D thermal conductivity from diffusive to ballistic can be captured through simple models, similar to quasi-ballistic charge transport observed in short-channel transistors:

$$k(L) = \sum_{p} \left( \frac{A}{LG_{p, \text{ball}}} + \frac{1}{k_{p, \text{diff}}} \right)^{-1} \approx \frac{G_{\text{ball}}}{A} \left[ \frac{1}{L} + \left( \frac{1}{\pi/2} \right) \lambda \right]^{-1}. \quad (1)$$

The first equality is a ‘three-colour’ model with $p$ the phonon mode (longitudinal acoustic, transverse, flexural), $G_{p, \text{ball}}$ calculated using the appropriate dispersion and $\sum k_{p, \text{diff}} = k_{\text{diff}}$ the diffusive thermal conductivity ($\sim 600 \, \text{W m}^{-1} \, \text{K}^{-1}$ at 300 K). A simpler ‘gray’ approximation can also be obtained by dropping the $p$ index, $k(L) \approx \left[ A/(LG_{\text{ball}}) + 1/k_{\text{diff}} \right]^{-1}$, where $G_{\text{ball}}/A \approx 4.2 \times 10^{9} \, \text{W K}^{-1} \, \text{m}^{-2}$ at room temperature (see Supplementary Note S9). The second expression in equation 1 is a Landauer-like model with $\lambda$ accounting for angle averaging in 2D to obtain the phonon backscattering. For convenience, we note that the ballistic thermal conductance of graphene can be approximated analytically as $G_{\text{ball}}/A \approx 1/(4.4 \times 10^{5} \, \text{T}^{4.68}) + 1/(1.2 \times 10^{10}) \, \text{W K}^{-1} \, \text{m}^{-2}$ over the temperature range 1–1,000 K, as a fit to full numerical calculations (Supplementary Fig. S16).

We compare the simple models in equation 1 with the experiments in Fig. 2c and find good agreement over a wide temperature range. The comparison also yields our first estimate of the intrinsic phonon mfp in SiO$_2$-supported graphene, $\lambda \approx (2/\pi)k_{\text{diff}}/(G_{\text{ball}}/A) \approx 90$ nm at 300 K and 115 nm at 150 K. (The same argument estimates an intrinsic phonon mfp $\lambda \approx 300–600$ nm in freely suspended graphene at 300 K, if a thermal conductivity $2,000–4,000 \, \text{W m}^{-1} \, \text{K}^{-1}$ is used.) This phonon mfp is the key length scale which determines when the thermal conductivity of a sample becomes a function of its dimensions, in other words when $L$ and $W$ become comparable to $\lambda$. On the basis of Fig. 2c, we note that quasi-ballistic heat-flow effects should become non-negligible in all SiO$_2$-supported graphene devices shorter than $\sim 1 \, \mu \text{m}$.

**Width dependence of thermal conductivity.** We now turn to the width dependence of heat flow in narrow GNRs. Our experimental data in Fig. 2b,d show a clear decrease of thermal conductivity as the width $W$ is reduced to a size regime comparable to the intrinsic phonon mfp. For instance, at room temperature $k \approx 230$, 170, 100 and 80 $\, \text{W m}^{-1} \, \text{K}^{-1}$ for GNRs of $W \approx 130$, 85, 65 and 45 nm, respectively, and same $L \approx 260$ nm. To understand this trend, we consider $k$ limited by phonon scattering with edge disorder through a simple empirical model with a functional form suggested by previous work on rough nanowires and GNR mobility.

$$k_{\text{eff}}(W, L) \approx \frac{1}{L} \left( \frac{\Delta}{W} \right)^{n} + \frac{1}{\kappa(L)}^{-1}. \quad (2)$$

Here $\Delta$ is the root-mean-square (r.m.s.) edge roughness (Fig. 3c) and $\kappa(L)$ is given by equation 1. The solid lines in Fig. 2d show...
good agreement with our GNR data \((L \approx 260 \, \text{nm})\) using \(\Delta = 0.6 \, \text{nm}\) and a best-fit exponent \(n = 1.8 \pm 0.3\). The parameter \(c = 0.04 \, \text{Wm}^{-1} \text{K}^{-1}\) can be used to fit the room-temperature data set and additional fitting discussion is provided in the Supplementary Note S9. (Note that we cannot assign an overly great physical meaning to the parameter \(c\), because the empirical model can only fit \(\Delta^2 / c\), not \(\Delta\) or \(c\) independently). The simple model appears to be a good approximation in a regime with \(\Delta \ll W\), where the data presented here were fitted. However, it is likely that this simple functional dependence would change in a situation with extreme edge roughness\(^{18}\), where the roughness correlation length (which cannot be directly quantified here) could also have an important role.

Nevertheless, the nearly \(W\)-squared dependence of thermal conductivity in narrow GNRs with edge roughness is consistent with previous findings for rough nanowires\(^{32,33}\), and also similar to that suggested by theoretical studies of GNR electron mobility\(^{34}\). The precise scaling with \(\Delta\) is ostensibly more complex\(^{30,31}\) than can be captured in a simple model, as it depends on details of the phonon dispersion, the phonon wave vector and indirectly on temperature. However, the \(\Delta\) estimated from the simple model presented above is similar to that from extensive numerical simulations below, and to that measured by transmission electron microscopy on GNRs prepared under similar conditions\(^{35}\). Thus, the simple expressions given above can be taken as a practical model for heat flow in substrate-supported GNRs with edge roughness \((\Delta \ll W)\) over a wide range of dimensions, corresponding to all size regimes in Fig. 3.

**Discussion**

We first revisit the effects of measurement contacts and how they relate to the interpretation of sample length in the quasi-ballistic heat-flow regime. As in studies of quasi-ballistic electrical transport\(^{26,27}\), we defined the ‘channel length’ \(L\) as the inside edge-to-edge distance between the heater and thermometer electrodes (Fig. 3c). Simple ballistic theory assumes contacts with an infinite number of modes and instant thermalization of phonons at the edges of the contacts. The former is well approximated here by electrodes two hundred times thicker than...
the graphene sheet; however, phonons may travel some distance below the contacts before equilibrating. The classical, continuum analogue of this aspect is represented by the thermal transfer length $L_T$ of heat flow from the graphene into the contacts3,36, which is automatically taken into account in our 3D simulations (Fig. 1e). However, a subcontinuum perspective37 reveals that graphene phonons only thermally equilibrate after travelling one mfp below the contacts. Previous measurements of oxide-encased graphene3 had estimated a thermal conductivity $k_{enc}$ = 50–100 W m$^{-1}$ K$^{-1}$, which suggests a phonon mfp $\lambda_{enc} = (2k_{enc}/\pi)/(G_{ball}/A) \approx 8$–15 nm under the contacts. This adds at most 12% to our assumption of edge-to-edge sample length (here $L \approx 260$ nm), a small uncertainty which is comparable to the sample-to-sample variation from fabrication, and to the size of the symbols in Fig. 2c. (The relatively low thermal conductivity of encased monolayer graphene3 is due to scattering with the SiO$_2$ sandwich, although some graphene damage from the SiO$_2$ evaporation38 on top is also possible.)

To gain deeper insight into our experimental results, we employ a numerical solution of the Boltzmann transport equation (BTE) with a complete phonon dispersion31,39. Our approach is similar to previous work6,40, but accounting for quasi-ballistic phonon propagation and edge disorder scattering in short and narrow GNRs, respectively (see Methods and Supplementary Notes S7 and S8). Figure 4a finds good agreement of thermal conductivity between our measurements and the BTE model across all samples and temperatures. We obtained the best fit for GNRs of width 130 and 85 nm with r.m.s. edge roughness $\Delta = 0.25$ and 0.3 nm, where the gray bands in Fig. 4a correspond to $\pm 5\%$ variation around these values. For GNRs of widths 65 and 45 nm, the gray bands correspond to edge roughness ranges $\Delta = 0.35$–0.5 and 0.5–1 nm, respectively. We note that unlike the empirical model of equation 2, the best-fit BTE simulations do not use a unique value of edge roughness $\Delta$. This could indicate some natural sample-to-sample variation in edge roughness from the fabrication conditions, but it could also be due to certain edge-scattering physics (such as edge-roughness correlation18 and phonon localization41), which are not yet captured by the BTE model.

Figure 4b examines the scaling of mfps by phonon mode, finding they are strongly reduced as the GNR width decreases below ~200 nm, similar to the thermal conductivity in Fig. 2d. The mfp for each phonon mode is calculated as an average over the entire frequency spectrum, weighted by the frequency-dependent heat capacity and group velocity (Supplementary Eq. S19). We note that longitudinal acoustic (LA) and transverse (TA) modes, which have larger intrinsic mfps, are more strongly affected by the GNR edge disorder. On the other hand, flexural acoustic modes (ZA) are predominantly limited by substrate scattering and consequently suffer less from edge disorder, consistently with recent findings from molecular dynamics simulations2,8.

Increasing edge disorder reduces phonon mfps (Supplementary Fig. S15d), and the thermal conductivity is expected to scale as shown in Fig. 4c. In the BTE model, the edge-roughness scattering is captured using a momentum-dependent specularity parameter (Supplementary Eq. S11), meaning that small wavelength (large momentum $q$) phonons are more strongly affected by line edge roughness. However, as $\Delta$ increases the specularity parameter saturates, marking a transition to fully diffuse edge scattering, and also to a regime where substrate scattering begins to dominate long-wavelength phonons in substrate-supported samples. This transition cannot be captured by the simplified $\Delta^0$ dependence in the empirical model of equation 2.

To further illustrate such distinctions, Fig. 4d displays the energy (frequency $\omega$) dependence of phonon mfps for a ‘small’ GNR and a ‘large’ SiO$_2$-supported graphene sample (corresponding to Fig. 3c and 3a, respectively). Low-frequency substrate scattering (proportional to $\sim 1/\omega^2$) dominates the large sample6,7, whereas scattering with edge disorder affects phonons with wavelengths comparable to, or smaller than, the roughness $\Delta$ (see Supplementary Note S7). Therefore, larger $\Delta$ can affect more long-wavelength (low energy) phonons, but only up to $\Delta \sim 1$ nm, where the effect of the substrate begins to dominate in the long-wavelength region (also seen in Fig. 4c). Such a separation of frequency ranges affected by substrate and edge scattering could provide an interesting opportunity to tune both the total value and the spectral components of thermal transport in GNRs, by controlling the substrate and edge roughness independently.

Finally, it is instructive to examine some similarities and differences between our findings here versus previous results regarding size effects on charge-carrier mobility in GNRs with dimensions comparable to the phonon or electron mfp. The edge-limited thermal conductivity begins to fall off in GNRs narrower than ~200 nm (Fig. 2d), or twice the intrinsic phonon mfp. A similar trend was noted for the electrical mobility in GNRs3,41, but with a fall off at widths narrower than ~40 nm (Supplementary Fig. S11). These observations are consistent with the intrinsic electron mfp being several times shorter3,42 than the phonon mfp in SiO$_2$-supported graphene, that is, ~20 nm for the electron mfp versus nearly ~100 nm for the phonon mfp at room temperature. Thus, edge disorder affects thermal transport more strongly than charge transport in GNRs of an intermediate width (40 nm < W < 200 nm), an effect that could be used to manipulate charge and heat flow independently in such nanostructures.

In conclusion, we have investigated heat flow in SiO$_2$-supported graphene samples of dimensions comparable to the phonon mfp. Short devices (L ~ $\lambda$, corresponding to Fig. 3b schematic) have thermal conductance much higher than that previously found in micron-sized samples, reaching 35% of the ballisitic limit at 200 K and 30% (~1.2 GW K$^{-1}$ m$^{-2}$) at room temperature. However, narrow ribbons (W ~ $\lambda$, corresponding to Fig. 3c schematic) show decreased thermal conductivity due to
phonon scattering with edge disorder. Thus, the usual meaning of thermal conductivity must be carefully interpreted when it becomes a function of sample dimensions. The results also suggest powerful means to tune heat flow in 2D nanostructures through the effects of sample width, length, substrate interaction and edge disorder.

Methods
Sample fabrication. Graphene monolayers were deposited on SiO$_2$/Si (~290 nm/0.5 nm) substrates by mechanical exfoliation from natural graphite. Graphene thickness and GNR edge disorder were evaluated with Raman spectroscopy$^{2,15}$. Samples were annealed in Ar/H$_2$ at 400°C for 40 min. Electron (e)-beam lithography was used to pattern the heater and thermometer electrodes as long, parallel, ~200-nm-wide lines with current and voltage probes, with a separation of $L$=260 nm (Fig. 1). Electrodes were deposited by successive evaporation of SiO$_2$ (20 nm) for electrical insulation and Ti/Au (30/20 nm) for temperature sensing. Additional e-beam lithography and oxygen plasma etching were performed when needed, to define GNR arrays with pitch ~150 nm and varying widths.

Electrical and thermal measurements. The heater electrode is slowly ramped up (<0.2 mHz) to 1.5 mA. We measured the resistance change of the sensor electrode through a lock-in technique with a frequency of 2.147 Hz and r.m.s. current of 1 μA (carefully verified to avoid additional heating). All electrical measurements were performed in a four-probe configuration, inside a Physical Property Measurement System (Quantum Design).

Numerical simulation. We obtain the thermal conductivity by solving the Boltzmann transport equation in the relaxation time approximation, including scattering at the rough GNR edges$^{31}$. The simulation uses the phonon dispersion of an isolated graphene sheet, which is a good approximation for SiO$_2$-supported graphene within the phonon frequencies that contribute most to transport$^{9}$, and at typical graphene–SiO$_2$ interaction strengths$^{7}$. (However, we note that artificially increasing the graphene–SiO$_2$ coupling, for example, by applying pressure$^{29}$, could lead to modifications of the phonon dispersion and hybridized graphene–SiO$_2$ modes$^{7}$.) We assume a graphene monolayer thickness $H=0.335$ nm and a concentration of 1% 13C isotope point defects$^{2,6}$. The interaction with the SiO$_2$ substrate is modelled through perturbations to the scattering Hamiltonian$^{8}$ at small patches where the graphene is in contact with the SiO$_2$ with nominal patch radius $a=8.75$ nm. Anharmonic three-phonon interactions of both normal and umklapp type are included in the relaxation time (see Supplementary Note S7). An equivalent 2D ballistic scattering rate$^{25,29}$ $\sim 2v_F/L$ is used in the numerical solution ($v_F$ is the heat flow direction along graphene) to account for transport in short GNRs.

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