Research Article

Hot Carriers in CVD-Grown Graphene Device with a Top h-BN Layer

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We investigate the energy relaxation of hot carriers in a CVD-grown graphene device with a top h-BN layer by driving the devices into the nonequilibrium regime. By using the magnetic field dependent conductance fluctuations of our graphene device as a self-thermometer, we can determine the effective carrier temperature \( T_e \) at various driving currents \( I \) while keeping the lattice temperature \( T_L \) fixed. Interestingly, it is found that \( T_e \) is proportional to \( I \), indicating little electron-phonon scattering in our device. Furthermore, the average rate of energy loss per carrier \( P_e \) is proportional to \( (T_e^2 - T_L^2) \), suggesting the heat diffusion rather than acoustic phonon processes in our system. The long energy relaxation times due to the weak electron-phonon coupling in CVD graphene capped with h-BN layer as well as in exfoliated multilayer graphene can find applications in hot carrier graphene-based devices.

1. Introduction

Recently, researchers in the graphene community are much interested in hot carriers in graphene-based systems since they determine the performance of high-power and high-frequency electronics, thermal management of electronic devices, optoelectronic devices, the quantum Hall metrology, and bolometric detectors [1–6]. Most of the hot carrier graphene devices in the high carrier density limit (the Bloch-Gruneisen temperature \( T_{BG} \) > the lattice temperature \( T_L \)) show the dominant cooling power from a weak coupling of carriers to acoustic phonon processes [7–10], which is represented by a heat flow power law equation \( P = \Sigma(T_e^\delta - T_L^\delta) \) [11], where \( \delta = 4 \) is a characteristic exponent, \( T_c \) is the carrier temperature, \( T_L \) is the lattice temperature, and \( \Sigma \) is the coupling constant. Furthermore, disordered-enhanced properties in hot carrier graphene devices revealed the supercollision cooling processes, where \( \delta = 3 \) [12–14]. Recently, hexagonal Boron Nitride (h-BN) bottom substrate can be a great heat drained material for disordered graphene so as to extremely reduce the carrier-phonon scattering via Wiedemann-Franz law heat diffusion, which is \( \delta = 2 \) [15–17]. However, the carriers of energy relaxation in such h-BN/disordered graphene systems with less substrate phonon interactions following the Wiedemann-Franz law heat diffusion are rarely studied. A key parameter for discussing the cooling process is the energy relaxation time \( \tau_c \), the characteristic time when the thermal energy is lost by the carriers [18–20].

To date, CVD-grown graphene appears to be a good candidate for large-scale graphene-based applications. However, as such a system may not be air stable, it is highly desirable to cap the CVD graphene with an inert layer so
as to experimentally realize stable, large-scale, and scalable devices. To this end, we have prepared CVD graphene with a top h-BN layer and studied the hot carrier effects in such a device. In this report, we studied the conductance fluctuations and hot carrier effects using a large measurement current \( I \) so as to raise the effective \( T_e \) above \( T_L \) on h-BN/CVD graphene and multilayer exfoliated graphene due to their disordered properties. Based on the self-thermometer linear properties between \( T_e \) and \( I \) due to Wiedemann-Franz law heat diffusion, we can investigate energy relaxation cooling processes under high carrier density limit \( (T_{BG} > T_L) \) between the h-BN/CVD graphene and multilayer exfoliated graphene for future graphene-based applications. By determining \( \tau_e \) from \( T_e \) and input power \( P_e \), we can find that \( \tau_e \) of the CVD graphene with a top h-BN layer is almost two orders of magnitude longer than those in exfoliated pristine monolayer/bilayer graphene. Such a result may find applications in hot carrier graphene-based transistors as a result of the weak electron-phonon coupling.

2. Experimental Section

2.1. Preparation of the Samples. As shown in the schematic diagram in Figure 1(a), we used the scotch tape method to mechanically exfoliate high purity and homogenous h-BN crystals synthesized by high pressure techniques [21] and transferred them by Gel-Pak polymer using the viscoelastic effect [22] on commercial chemical vapor deposition (CVD) graphene/285 nm SiO\(_2\)/Si substrate [23]. The CVD graphene region outside h-BN sheet was etched by oxygen plasma so as to confine to our CVD graphene region under the h-BN sheet. CF\(_4\) gas was used to etch h-BN sheet protected by photoresist for 8 terminals and Cr/Au metal depositions as shown in Figure 1(b).

2.2. Electrical Measurements. An ac driving current from lock-in amplifiers passed into the source and drain contacts through the graphene devices for Hall-bar measurements. The magnetoresistance was measured in a He\(^3\) cryostat equipped with a superconducting magnet.

3. Results and Discussion

Figure 2(a) shows the longitudinal resistivity \( \rho_{xx} \) as a function of magnetic field with fixed current \( I = 20 \text{ nA} \) at different temperatures that are equivalent to \( T_L \). The conductance fluctuations are observed, and they decrease as \( T_L \) increase from \( T_L = 0.32 \) to \( 50 \text{K} \), which are typical properties in disordered mesoscopic graphene [24–27]. Figure 2(a) inset shows the corresponding Hall resistivity at \( T_L = 0.32 \text{K} \), which can calculate the carrier density \( n_A = 3.5 \times 10^{12} \text{cm}^{-2} \) from the Hall slope and Hall mobility \( \mu = 2092 \text{cm}^2/\text{Vs} \) from carrier density and zero field resistivity. Figure 2(b) shows the results of \( \rho_{xx} \) at various driving currents \( I \) from \( I = 20 \text{ nA} \) to \( 30000 \text{ nA} \) at a fixed \( T_L \) of \( 0.32 \text{K} \), which reveals similar conductance fluctuation characteristics by current heating in the nonequilibrium regime due to the hot carrier effects in disordered two-dimensional systems [18, 28–31].

By using a conductance fluctuations-based thermometer between \( \rho_{xx} (I) \) and \( \rho_{xx} (T) \), we are able to reveal the clear conductance variations for lattice temperature and current dependence between \( B = 0.5 \) and \( 3 \text{ T} \) by subtracting a smooth background that avoids the zero field weak localization peak and high field Shubnikov-de Haas-like oscillations as shown in Figures 3(a) and 3(b) [13]. Hence, we determine the root mean square (RMS) conductance fluctuation \( \delta g_{rms} \) in units of \( e^2/h \) for every \( \delta g_{rms} (T_L) \) and \( \delta g_{rms} (I) \) data set as a common graph (see Supporting Information (available...
Figure 2: (a) Magnetoresistivity $\rho_{xx}(B)$ at various $T_L$ for sample A. The inset shows the Hall resistivity $\rho_{xy}(B)$ at $T_L = 0.32$ K. (b) Magnetoresistivity $\rho_{xx}(B)$ at various driving currents $I$ at fixed $T_L = 0.32$ K for sample A.

Figure 3: (a) Conductance fluctuations in sample A as a function of magnetic field at various lattice temperatures with fixed $I = 20$ nA. (b) Conductance fluctuations in sample A as a function of magnetic field at various driving currents for $T_L = 0.32$ K.

By averaging conductance fluctuations over the range of $B$ so as to assign an effective $T_e$ to the driving current as shown in Figure 4(a) [13, 18]. Interestingly, $T_e$ shows a linear dependence on $I$. According to the seminal work done by Baker and coworkers about the energy transfer between carriers and the lattice [31], the following relation can be found:

$$T_e \propto I^\alpha,$$

where $\alpha = 2/(\rho + 2)$ and $\rho$ is the exponent for the inelastic scattering rate $\tau_{in}^{-1} \propto T^\rho$. Therefore, Figure 4(a) shows that $T_e(I)$ of our sample A follows (1) with $\rho = 0$ and $\alpha = 1$, which suggested little carrier-phonon scattering in two-dimensional material heterostructure systems [26]. Furthermore, one would be interested in whether the heat dissipation was transferred by another mechanism rather than carrier-phonon scattering. Under the low-temperature limit ($T_L < T_{BG}$), $T_{BG} = 2\hbar s k_F/k_B$, where $s$ is the sound
velocity and \( k_F \) is the Fermi radius. For graphene systems (\( s = 2.1 \times 10^5 \text{ ms}^{-1} \)), \( T_{BG} \sim 54n^{1/2}K \equiv 93.4K \) with the carrier density \( n \approx 3.5 \) in units of \( 10^{12} \text{ cm}^{-2} \) in sample A [13, 32]. The energy loss (\( P_e \)), the average rate of energy loss per carrier, is usually expressed to the carrier and lattice temperatures as

\[
P_e = A \left( T_e^\beta - T_L^\beta \right),
\]

(2)

with a constant \( A \) and a characteristic exponent \( \beta \). In general graphene systems, \( \beta \approx 3 \) suggests the supercollision cooling mechanism in disordered systems [13, 18, 33] and \( \beta \approx 4 \) suggests the two-dimensional acoustic phonon cooling processes in clean systems [34, 35]. Particularly, \( \beta \approx 2 \), the heat diffusion described by the Wiedemann-Franz law, was found in graphene on bottom h-BN substrate systems, where bottom h-BN substrate acts as a thermal conduction layer, effectively reducing the electron-phonon coupling [14–16]. Such interesting bottom h-BN substrate underneath graphene can change graphene heat transport mechanism from electron-phonon interactions (\( \beta \approx 3 \sim 4 \)) to heat diffusion (\( \beta \approx 2 \)). One might be interested in what heat transport mechanism will be in encapsulated graphene between bottom insulating SiO\(_2\) and top h-BN sheet as our sample A rather than traditional graphene on h-BN substrate [14–16]. Based on the work of Baker et al. [31], \( \beta = p + 2 \), which suggested \( \beta \) in sample A should be close to 2 since the linear relation between \( T_e \) and \( I \) is \( p \approx 0 \) (\( 1 = \alpha = 2/(p + 2) \)). Apparently, our results in sample A are highly consistent with this speculation for \( \beta \approx 2 \) as shown in Figure 4(b), where \( P_e = I^2R_{xx}/nWL \) (\( W \) and \( L \) are the width and length of our sample A, resp.). Interestingly, the heat transfer mechanism in our sample A with top h-BN sheet on graphene/SiO\(_2\) was dominated by heat diffusion (\( \beta \approx 2 \)), which is the same as traditional graphene/h-BN substrate devices [14–16]. Such interesting results suggest that the top h-BN sheet coupled with graphene is strong heat transfer medium in comparison with other coupled materials, like SiO\(_2\) or air. Also, such structures in the top h-BN sheet on graphene not only protected graphene without air molecular adsorbing doping [2–36], but also screened graphene from electrostatic force [37, 38], a great advancement for graphene-based devices.

In order to further discuss the linear relation between \( T_e \) and \( I \) that indicates little carrier-phonon scattering in our sample A, we fabricate multilayer exfoliated graphene (sample B) that showed the same linear relation between \( T_e \) and \( I \) by zero field resistance as a thermometer and conductance fluctuations due to its disordered property as in our previous reports [23–26]. Again, we are able to measure the conductance fluctuations for temperature and current dependence as shown in Figure 5(a) and inset [13]. Consequently, we found the linear relation between \( T_e \) and \( I \) by utilizing \( \delta g_{\text{rms}}(T_L) \) and \( \delta g_{\text{rms}}(I) \) as a thermometer for determining \( T_e \) (see Supporting Information) [13] as shown in the inset of Figure 5(b), which suggested \( p \approx 0 \) and \( \beta \approx 2 \). Under the low-temperature limit (\( T_L < T_{BG} \)), \( T_{BG} \sim 54n^{1/2}K \equiv 188.6K \) with the carrier density \( n \approx 12.2 \) in units of \( 10^{12} \text{ cm}^{-2} \) in sample B [13, 32]. Consistently, \( P_e \) is proportional to \( (T_e^2 - T_L^2) \) in sample B as shown in Figure 5(b), which belonged to Wiedemann-Franz law heat diffusion and corresponded to the transport situation for little carrier-phonon scattering for \( \alpha \approx 1 \) [7, 26]. Interestingly, the multilayer exfoliated graphene (sample B) has the same heat transfer mechanism (\( \beta \approx 2 \)) as the h-BN/graphene (sample A) systems [14–16].

Although the heat transfer mechanism of multilayer exfoliated graphene is the same as h-BN/CVD graphene for heat diffusion (\( \beta \approx 2 \)), the energy relaxation time can describe this cooling process about the typical time on which energy is lost from carriers [39, 40]. Therefore, the energy relaxation time can be expressed by

\[
P_e = \frac{k_B(T_e - T_L)}{\tau_e}.
\]

(3)

Figure 6 compares the energy relaxation time \( \tau_e \) between the h-BN/CVD graphene (sample A) and the multilayer
4. Conclusion

We have studied conductance fluctuations and hot carrier effects caused by current heating on h-BN/CVD graphene and multilayer graphene as a self-thermometer. It has been shown that $T_e$ ($P_e$) is linearly proportional to $I (T_e^2 - T_L^2)$ in both of the disordered graphene devices, suggesting the little electron-phonon scattering and heat diffusion due to Wiedemann-Franz law. The extremely long energy relaxation time may find applications in graphene-based hot carrier devices. Such a result may be useful for other material systems [42, 43].

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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Supplementary Materials

This includes the method of determining the effective electron temperature using amplitudes of the observed conductance fluctuations. (Supplementary Materials)

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