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Hot carriers in epitaxial graphene sheets with and without hydrogen intercalation: role of substrate coupling

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The development of graphene electronic devices produced by industry will rely on efficient control of heat transfer from the graphene sheet to its environment. In nanoscale devices, heat is one of the major obstacles to the operation of such devices at high frequencies. Here we study transport of hot carriers in epitaxial graphene sheets on 6H-SiC (0001) substrates with and without hydrogen intercalation by driving the device into the non-equilibrium regime. Interestingly, we demonstrate that the energy relaxation time of the device without hydrogen intercalation is two orders of magnitude shorter than that with hydrogen intercalation, suggesting applications of epitaxial graphene in high-frequency devices which require outstanding heat exchange with the outside cooling source.

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1. Introduction

Graphene, which can approximate an ideal two-dimensional system, has extraordinary electrical,[1, 2] optical,[3, 4] mechanical[5, 6] and thermal[7, 8] properties and has been considered for numerous applications such as field-effect transistors,[9] ultrasensitive gas sensors[10] and electromechanical resonators.[11] To date, mechanical exfoliation,[12] chemical vapor deposition (CVD)[13] and epitaxial growth[14] are the three major fabrication methods for graphene-based electronic devices. Although mechanically exfoliated graphene is of the best quality, the small size of its domains appears to limit practical device applications. On the other hand, CVD methods provide a way to produce large-area films but contamination from polymer residues may be inevitable when transferring the sheet onto a substrate. Epitaxial graphene, in contrast, can be of wafer size and does not require subsequent transfer procedures, which is ideal for applications in high-frequency devices.[15]

Functionalized graphene,[16-20] functionalized substrates,[21, 22] and quasi-free-standing graphene production methods[23-28] can improve the quality of graphene by reducing its coupling with the substrate and may make graphene-based field-effect circuits realizable by opening a band gap in some cases. However, the long-term prospects for most practical applications of graphene require efficient removal of waste heat produced during operation, which is especially important when a graphene-based device works at high frequencies. Therefore investigations of heat transfer from hot carriers in graphene are desirable.

In a semi-classical picture, an electron and phonon system can be described statistically by the Fermi-Dirac (FD) distribution and the Bose-Einstein (BE) distribution:

$$f_{FD}(\varepsilon) = \frac{1}{\exp[(\varepsilon - \mu)/k_B T_e] + 1}, \tag{1}$$

$$f_{BE}(\varepsilon) = \frac{1}{\exp[\varepsilon / k_B T_L] - 1}, \tag{2}$$

where ε, μ, k_B, T_e, and T_L are the single-particle electron energy, the chemical potential, the Boltzmann constant, the electron temperature and the temperature of phonon system, respectively. In the low current regime T_e and T_L are the same, reflecting the equilibrium between the phonons and charge carriers via electron-phonon scattering. As the injected current is increased, the external electric field will raise the total kinetic energy of the electron system. This will raise the value of T_e and lead to energy transfer from the electron system to the substrate lattice. The magnitude of this energy transfer depends on the temperature difference between T_e and T_L, as is the case for contact between two heat reservoirs. In this situation T_e can be substantially higher than T_L and this non-equilibrium heating phenomenon is known as the charge heating effect.[29]

In this work we study charge heating in epitaxial graphene sheets with and without hydrogen intercalation (H-intercalation) in order to understand their energy relaxation properties. Without intercalation, the buffer layer sample has essentially no conductivity, and can be compared as in this study only when it is converted into a useful electronic material by the saturation of the dangling bonds of the Si atoms. The electronic transport in the H-intercalated sample is then of high quality, when compared to the naturally conducting sample. Interestingly, however, our results show that the H-intercalated graphene sheet has a much longer energy relaxation time compared to that of epitaxial graphene without H-intercalation. This can be attributed to the reduced coupling of the sheet with the SiC substrate in the absence of covalent bonds, which are broken by H-intercalation.[25, 30, 31]

2. Experimental section

2.1 Preparation of the samples

A controlled sublimation method was used for graphene growth on a chemically-mechanically polished semi-insulating 6H-SiC (0001) surface. Before the growth process, standard procedures were applied to clean the SiC substrates. After the cleaning process, the 6H-SiC (0001) surface was placed face-to-face with a polished graphite disk and was arranged such that wide and uniform Newton rings could be observed in fluorescent light. The substrates were dehydrated and cleaned in the furnace at 725 °C for approximately 16 hours in Ar background gas at a pressure slightly higher than 1 100 kPa. After the substrate treatment, the temperature was increased and maintained at 1200 °C for 30 min. Then, for sample A, the furnace was heated to 1950 °C at a ramping rate of 100 °C/min and maintained at 1950 °C for 30 min to grow graphene having two layers. Here, the exposed Si atoms in the SiC (0001) lattice form partial covalent bonds to carbon atoms in the lower graphene layer or zero layer,[31-33] and only the top layer is conducting. Sample A was then patterned into Hall-bar geometry for electrical measurements by standard photolithography using reactive ion etching in O_2 plasma. Ti/Au contacts were deposited by optical lithography and a lift-off process.

The substrate and growth conditions for Sample B were the same, except that the final temperature was reduced to 1600 °C and the annealing time was increased to 1 hour. These conditions result in the production of so-called zero-layer graphene which is covalently bound to the top-most layer of Si atoms and is non-conducting. Hall bars were fabricated as described above, and the measured average sheet resistivity of the device described here was 1.08 MΩ at 23 °C. After the process for Hall-bar production, the sample was heated again in the furnace for 24 hours at 710 °C ± 10 °C with forming gas (96% Ar + 4% H_2) background at a total pressure slightly higher than 100 kPa. During this process hydrogen atoms were intercalated into the graphene-SiC interface.[25, 34, 35] The influence of covalent bonding between carbon atoms of bottom-layer graphene and Si atoms of the SiC substrate can be eliminated by this process, and moreover the dangling bonds at the interface are passivated by bonds with hydrogen atoms, effectively decoupling the graphene sheet from the SiC substrate. After H-intercalation, the sheet resistivity of Sample B was measured to be 22.7 kΩ at 23 °C, which was similar to that of Sample A and indicated substantial breaking of the covalent bonds.

2.2 Electrical measurements

Standard four-terminal electrical measurements were performed on Sample A and Sample B. The magnetic field was applied perpendicular to the plane of the graphene sheet. The dc current was injected by using a constant current source (Keithley 236). The voltage probes for the measurements of resistances R_{xx} and R_{xy} are separated from the source and drain to minimize the influence of contact resistance. For two-dimensional systems like graphene, one can then obtain resistivities ρ_{xx} and ρ_{xy} by $\rho_{xx} = R_{xx}w/l$ and $\rho_{xy} = R_{xy}$, where w is the width of the sample and l is the distance between the two voltage probes for R_{xx}. From the sign of Hall resistivity ρ_{xy}, the type of carriers for transport could be identified. The results show that Sample A is n-type whereas Sample B with H-intercalation is p-type. This behavior is consistent with the literature demonstrating that intercalation leads to the robust p-type doping.[25, 30] For the discussion here, only the magnitude of ρ_{xy} related to the carrier density is concerned.

3. Results and discussion

We note that in the AFM image in Fig. 1a for Sample A, the epitaxial graphene on SiC (0001) as shown has surface roughness on the order of 10 nm. This reflects the step-like terrace structure that was formed due to the miscut angle of the wafer relative to the crystalline basal plane, and by the sublimation of Si from the edges of the SiC atomic planes in the annealing stage of graphene growth. In sample B, where annealing occurred at lower temperature, there is no terrace formation and the surface is composed of atomic planes of one-half unit cell height (~ 0.75 nm) as observed in Fig. 1b.

Figures 2a and 2d show the longitudinal resistivity ρ_{xx} as a function of magnetic field at various measurement temperatures (equivalent to the lattice temperature) T_L for Sample A and Sample B. The currents $I_A = 5$ µA and $I_B = 7$ µA were applied for these T-dependent measurements. At lower fields around zero magnetic field $B = 0$ weak localization dominates due to quantum interference and negative magneto-resistivity is observed in the sense that ρ_{xx} decreases with increasing magnetic field. Figures 2b and 2e then show the absolute value of the corresponding Hall resistivity for each sample, both of which are T-independent. From the Hall slope $\delta\rho_{xy}/\delta B$ defined as $\delta\rho_{xy}/\delta B \equiv B/(ne)$, the carrier densities of $n_H^A = 1.60 \times 10^{13}$ cm^{-2} and $p_H^B = 1.85 \times 10^{13}$ cm^{-2} are estimated for Sample A and Sample B. Although the Hall mobility of Sample A (≈ 890 cm^2/V/s) appears to be slightly higher than that for Sample B (≈ 770 cm^2/V/s), we shall show that in the field of magneto-transport, the quantum mobility, rather than the classical Hall mobility is a more important physical quantity regarding the disorder within the graphene systems. Figures 2c and 2f present the results of $\rho_{xx}(B)$ at various driving currents I with a fixed T_L, in which a clear dependence on I is observed.[36, 37] Hence by comparing $\rho_{xx}(B, I)$ with $\rho_{xx}(B, T)$, that is, by using the resistivity value as a self-thermometer as clearly demonstrated in samples which show insulating behavior in the literature,[29, 38, 39] we are able to determine the effective carrier temperature T_{cc}. The obtained results are presented in Fig. 3. We note that for the same currents, T_{cc} in sample B is higher than that in sample A, suggesting it requires less work to raise the mean kinetic energy of carriers in sample B than those in sample A.

In the seminal report of Wennberg *et al.*,[29] which deals with the energy transfer between charged carriers and the lattice by considering the two-bath model, it was proposed that

$$T_{cc} \propto I^{\alpha}, \tag{1}$$

where $\alpha = 2/(p + 2)$ and p is the exponent for the temperature dependence of inelastic scattering rate $\tau_{in}^{-1} \propto T^{p}$. It is found in Fig. 3 that $T_{cc}(I)$ of our samples follows the same trend as Eq. (1) with $p \approx 2$, that is, $\alpha \approx 0.5$. This result indicates the importance of carrier-phonon scattering.[40, 41] Given the relation $T_{cc}(I)$, one can further calculate the energy-loss rate P_{cc}, the average rate of energy loss per carrier, which is usually related to the carrier and lattice temperatures as

$$P_{cc} = A(T_{cc}^{\beta} - T_{L}^{\beta}), \tag{2}$$

where A and β are the parameters relevant to the scattering mechanism for energy relaxation.[42] In the model of Wennberg *et al.*, $\beta = p + 2$. Experimentally, P_e is determined as $P_{cc} = I^2 R_{xx} /(nwl)$. Moreover its relation with energy relaxation time τ_ε can be expressed by

$$P_{cc} = \frac{k_B (T_{cc} - T_L)}{\tau_\varepsilon}. \tag{3}$$

Figures 4a and 4d show that the energy-loss rate of both samples increases with T_e following the power-law behavior of Eq. (2). In the low-I region where $T_{cc} < 3$ K, P_{cc} deviates from the power-law dependence and thus Eq. (3) cannot be used for the determination of τ_ε. The underlying reason is that a low I cannot raise T_{cc} as presented in Fig. 3. The red lines denote the characteristic exponents which are obtained by averaging the power-law fitting results for each field. Such fits were thereby performed at current levels high enough to remove the equilibrium between carrier and phonon systems. The larger slope shown in Fig. 4d shows that the energy of Sample B increases faster than that of Sample A, however. For Sample B, the average exponent of $\overline{\beta} \approx 4.76$ is obtained whereas for Sample A it is $\overline{\beta} \approx 4.07$. The obtained values are consistent with the existing results on exfoliated and CVD-grown graphene, demonstrating that $\beta \approx 4$.[42-44] In addition, the magnitude of P_{cc} is about two orders of magnitude smaller for Sample B compared to Sample A for similar temperatures. Since P_{cc} is inversely proportional to τ_ε according to Eq. (3), the magnitude of τ_ε is larger for Sample B compared to Sample A as shown in Figs. 4b and 4e. We also plot τ_ε as a function of input current I in Figs. 4c and 4f. The power-law behavior can still be observed. Our charge heating measurements at various B show consistent results, suggesting the validity of using the resistivity value as a thermometer.

In the epitaxial graphene systems studied here the cooling of hot carriers is related to interactions with surface phonons of the SiC substrates,[45] giving rise to $\beta = 4$ as described in Ref. 42. At temperatures T_{cc} above the Bloch-Gruneisen temperature T_{BG}, the energy-lose rate is predicted to be enhanced due to disorder-assisted carrier-phonon scattering and the exponent of $\beta = 3$ would instead be found.[46] A graphene system with the presence of charge puddles near the Dirac point can also have an enhanced P_{cc} and $\beta = 3$ (Ref. 42). In our samples, $T_{BG}^{A} \approx 216$ K and $T_{BG}^{B} \approx 232$ K are estimated.[42] Since T_{cc} is well below T_{BG} and the carrier density is high for both samples, phonons of the SiC substrate would then play a dominant role in cooling hot carriers of the graphene sheet.

Figure 5 compares the energy relaxation time τ_ε between the two samples. We can observe clearly that τ_ε of Sample B with H-intercalation is about two orders of magnitude longer than that in Sample A without H-intercalation. We note that in the work of Somphonsane *et al.*[42] for the same density, τ_ε for electrons is about the same as that for holes in graphene. Therefore the observed vastly different τ_ε for electrons in Sample A compared with that for holes in Sample B cannot be ascribed to carrier type. Instead, we infer that the coupling strength between hot electrons or holes in the graphene sheet and the phonon heat reservoir of the SiC substrate is substantially different for these two samples. A deeper examination of the characteristics of these samples aids in understanding this difference. In Fig. 6a, Sample A and Sample B have similar Raman spectra with G bands located at ~ 1585 cm^{-1} and 2D bands at ~ 2680 cm^{-1}, characteristic of graphene.[20, 47] The absence of any significant D band contribution in the spectral region below the G band indicates that the graphene lattice is both relatively large in area and defect-free for the two samples. The 2D band is somewhat broadened for Sample A, suggesting that this sample may include some few-layer regions in addition to monolayer graphene. This is consistent with earlier low-energy electron microscopy (LEEM)[48] results for another sample produced under the exact same annealing recipe (see Fig. S1 in the Supporting Information) where the presence of bilayer regions was observed over about 15% of the surface area.

However in Fig. 6b we observe a significant difference in the high-field magneto-transport behavior. The background resistivity for Sample A increases strongly with magnetic field whereas it is weakly magnetic-field-dependent for Sample B. An analysis of the Shubnikov de Haas (SdH) oscillatory structure[49] shows that the filling factor is resolved up to about $v = 94$ for Sample B but only to about $v = 56$ for Sample A. These differences may reflect the relative (in)homogeneity of the samples, since the samples have similar carrier densities. As shown in Fig. S3 of the Supporting Information, from the slope of v versus $1/B$, which follows $n = veB/h$, carrier densities of $n_{SdH}^{A} \approx 1.60 \times 10^{13}\,\text{cm}^{-2}$ and $p_{SdH}^{B} \approx 1.76 \times 10^{13}\,\text{cm}^{-2}$ are estimated, consistent with the values from the Hall measurements. By analyzing the amplitudes of SdH oscillations $\Delta\rho_{xx}$ in Fig. S2 of the Supporting Information, we can obtain both the effective mass m^* and the quantum mobility μ_q according to

$$\Delta\rho_{xx}(B, T) = A\exp(-\pi/(\mu_q B))D(B, T) \quad , \tag{4}$$

where $D(B,T) = 2\pi^2 k_B m^* T/(\hbar eB)\sinh(2\pi^2 k_B m^* T/(\hbar eB))$.[50, 51] As shown in Fig. 6c, the fit of Eq. (4) to the experimental $\Delta\rho_{xx}(B, T)$ data for Sample B gives the effective mass of $m_B^* \approx 0.094 m_e$. However, the oscillations are so weak for Sample A that m_A^*

is difficult to determine. We can calculate the quantum motility, which includes both large-angle and small-angle scattering events, of Sample B to be $\mu_q^B \approx 386\,\mathrm{cm^2V^{-1}s^{-1}}$ by Eq. (4) as shown in Fig. 6d.

The above observations, together with the rising background magneto-resistivity[52] of Sample A shown in Fig. 6b, indicate that disorder is stronger in Sample A than that in Sample B. Although our experimental findings do not give conclusive results on this point, some of this difference may be related to the greater coupling strength between the non-intercalated graphene sheet and the SiC substrate. As shown in Fig. S4, the resistivity of hydrogen-intercalated sample shows weak insulating behavior over the whole range of *T*. In contrast, graphene sample without hydrogen intercalation shows metallic behavior in the sense that the resisitivy increases with increasing T, characteristics of electron-phonon scattering. These results suggest that charge-phonon scattering is stronger in sample without hydrogen intercalation compared with that in a hydrogen-intercalated device. Other factors may be the greater substrate roughness due to terrace formation and the presence of bilayer regions. Our main experimental finding is to demonstrate that in the graphene sheet without H-intercalation the energy acquired from the input current is transferred to the substrate more quickly than in the sample with H-intercalation, i.e., the more strongly coupled carriers have a better ability to make energy transfer with the supporting substrate than the weakly-coupled ones. With H-intercalation, the carrier-phonon coupling strength is reduced and the energy relaxation time is shown to be longer (See Fig. S5). Such behavior is important especially when the device works at high frequencies.

4. Conclusion

We have investigated hot carrier effects caused by current heating in epitaxial graphene sheets with and without H-intercalation. It is shown that intercalation of hydrogen atoms can assist to decouple the graphene sheet and the SiC substrate and can thus reduce scattering and increase the mobility of the device. The energy relaxation time is shorter for the sample without H-intercalation compared to that with intercalation since the coupling at the interface plays an important role in the energy transfer. Therefore although the coupling between the graphene sheet and the substrate could be detrimental to the carrier mobility, it is beneficial for the removal of the heat from the sheet. Our work thereby shows that as-grown epitaxial graphene could be utilized and optimized for promising applications in hot-carrier and high-frequency devices.

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Figure Captions

Figure 1 AFM results including height and phase spatial images for the samples (a) without and (b) with H-intercalation. The bottom panel shows the line profile along the line denoted in the height image.

Figure 2 (a)&(b) Magneto-resistivity $\rho_{xx}(B)$ and Hall resistivity $|\rho_{xy}(B)|$ at various temperatures T_L for Sample A. From top to bottom in (a): T_L = 3 K, 5 K, 7 K, 9 K, 12 K, 15 K, and 21 K. (c) $\rho_{xx}(B)$ at different input currents I with T_L fixed at 2 K. From top to bottom: I = 20 μA, 50 μA, 70 μA, 85 μA, and 100 μA. (d)-(f) The corresponding results for Sample B. From top to bottom in (d): T_L = 2 K, 5 K, 8 K, 10 K, 15 K, and 20 K. The sequence of the input current in (f) is the same as in (c). The lattice temperature is also constantly fixed at $T_L \approx 2$ K.

Figure 3 The effective carrier temperature T_{cc} versus input current I on a log-log scale.

Figure 4 (a) The carrier energy-loss rate P_{cc} versus T_{cc} at various magnetic field on a log-log scale for Sample A. (b) The energy relaxation time τ_ε versus T_{cc} on a log-log scale. (c) The energy relaxation time τ_ε versus I on a log-log scale. (d)-(f) The corresponding results for Sample B. The arrows denote the lower bound for the fits.

Figure 5 Comparison of $\tau_\varepsilon(T_{cc})$ between Sample A and Sample B.

Figure 6 (a) Raman spectrum for Sample A, Sample B, and the SiC substrate. (b) $\rho_{xx}(B)$ for $0 < B < 15$ T. (c) The amplitudes of SdH oscillations $\Delta\rho_{xx}$ as a function of temperature T at B = 13.6 T for Sample B. (d) $\ln(\Delta\rho_{xx}/(X/\sinh X))$ as a function of $1/B$ for Sample B.