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Correlation between resistance fluctuations and temperature dependence of conductivity in graphene

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It is known that monolayer graphene has an extraordinarily high intrinsic mobility of the charge carriers. An important complication is the presence of strong mesoscopic resistance fluctuations (MRFs) that, in graphene, persist to relatively high temperatures. These reproducible fluctuations are seen as a function of gate voltage but they are not expected to be seen as a function of temperature T. However, we propose that the monotonic increases or decreases in resistance $R(T)$ that we observe in graphene flakes as T increases from 4.2 K to around 70 K arise from the decay of the magnitude of the MRFs due to progressive dephasing of the interfering scattered electron waves. Using the field effect transistor configuration, we demonstrate that this explanation is correct by measurements of $R(T)$ at different constant gate voltages V_G tuned to different features of the MRFs observed in $R(V_G)$ at constant temperature. We find that the T dependence of the MRF magnitude is, surprisingly, best fitted by an exponential decay.

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The extraordinary properties of graphene sheets of single-atom thickness have led to great interest both for new physics and for potential use in applications.[1,2] Investigations of suspended graphene flakes have yielded mobilities approaching 200 000 cm^2/Vs at low temperatures[3] and 120 000 cm^2/Vs at 240 K.[4] The resistivity increased linearly with temperature from 50 to 240 K (Ref. 4) suggesting that scattering by longitudinal-acoustic phonons was the main source of T dependence above 50 K in this case. Generally it is agreed that for low charge-carrier densities near the charge neutrality point (NP), the resistivity is governed[1,5,6] by carrier-density inhomogeneity induced by the potential of charged impurities ("puddles" of electrons and holes, as imaged using a scanning single-electron transistor[7]). However, toward low T, in some cases, a reversal of sign in the temperature dependence is observed,[8] an effect which lacks a clear explanation. It is on this low T behavior that we focus in this Brief Report. We find that in some cases $R(T)$ increases with T but in others it decreases and this opposite-sign behavior below around 70 K is present not only in the low charge-carrier density but also in the high charge-density regions.

Weak localization effects are suppressed in graphene[9] owing to intravalley elastic scattering that breaks the chirality of the charge carriers,[10] especially in the low carrier-density regime. On the other hand, mesoscopic resistance fluctuations (MRFs) effects[11] are strongly seen in graphene[9,12,13] as predicted[14] and are observed to persist to much higher temperatures (>50 K) (Refs. 9 and 12) than in conventional two-dimensional materials.[15] MRFs are clearly seen as magnetic field or gate voltage is changed. However, increasing temperature does not change the constructive or destructive nature of the interference but leads to a decay of the MRF amplitudes which is predicted to follow weak inverse power laws.[11]

We demonstrate in this Brief Report that the anomalous monotonic changes in $R(T)$ that we observe at low T in our graphene samples over a wide range of V_G are in fact due to this decay of the MRFs. This is done by making measurements of the resistance of monolayer graphene samples as a function of T at gate voltages tuned to specific maxima and minima of the mesoscopic fluctuation pattern. These measurements clearly show the correctness of our scenario, which explains the different signs of the resistance anomaly for similar gate voltages. Surprisingly, our fits to the measured $R(T)$ curves suggested an *exponential decay* with temperature rather than inverse power laws. We confirmed this by independent measurements of $R(V_G)$ at different constant T: the root-mean-square (rms) MRF amplitude as a function of T showed a remarkably accurate exponential decay.

The graphene samples were prepared by depositing graphene flakes from a crystal of highly oriented pyrolytic graphite on top of the 300-nm-thick SiO$_2$ layer formed on the surface of a heavily doped Si substrate. With the help of a marker coordinate system, monolayer graphene flakes were identified[16] from the optical contrast of digitized images. A monolayer strip of 10 μm in length and ~1.45 μm in width was chosen for the present measurements (Fig. 1). A set of parallel Cr/Ar (3 nm/40 nm) electrodes was fabricated by the electron-beam lithography technique. Electrical measurements were performed in the four-probe configuration where electrodes 1 and 7 [inset in Fig. 1(b)] were connected to a source-meter Keithley 2400 of constant current (100 nA was used in all measurements), and the voltage drop was recorded by a Keithley 2000 voltmeter (most extensively between electrodes 5 and 6 with the largest separation of 1200 nm and sample width 1450 nm). Similar effects were seen for other electrode pairs and other samples. The bulk silicon substrate, electrically separated from the graphene sample by the SiO$_2$ layer, was connected to a voltage source Keithley

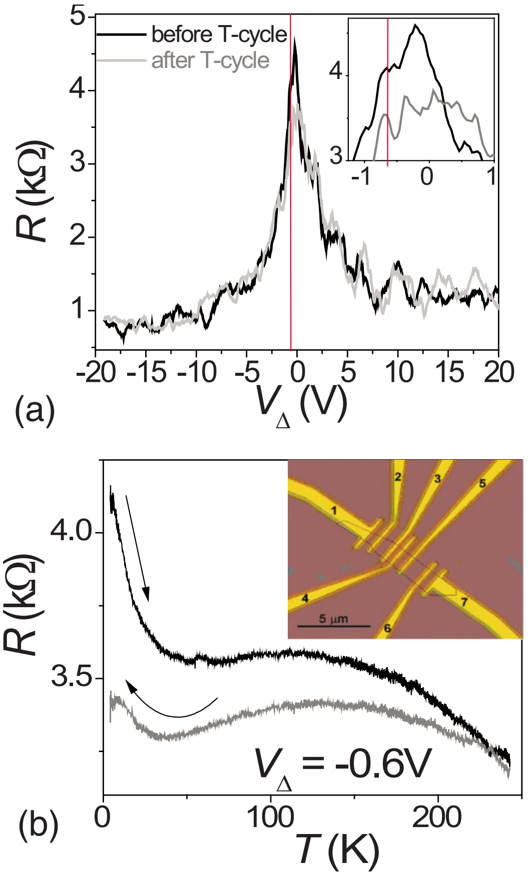

FIG. 1. (Color online) (a) Resistance with MRFs of the graphene flake as a function of gate voltage at 4.2 K before (black) and after (gray) a T cycle up to 245 K and back down again to 4.2 K; the inset shows the peak on an expanded scale. (b) Resistance during the T cycle carried at a constant gate voltage $V_\Delta = -0.6$ V [indicated by vertical lines in (a)]. Inset: optical image of the pattern of gold electrodes on top of a monolayer graphene sample (indicated by the dotted lines).

2400 and was used as a back gate for the field effect measurements. The sample was placed into the sample chamber and annealed at 120 °C in vacuum (10^{-7} mbar) for 2 days. Then helium gas was introduced into the sample chamber from the helium line, thus ensuring that the pressure of helium gas remain relatively stable in the whole temperature range.

A typical resistance as a function of gate voltage at $T = 4.2$ K is presented in Fig. 1(a) showing the usual maximum in resistance (at the NP) with charge-carrier density increasing for V_Δ on either side of the maximum (we use the gate voltage $V_\Delta = V_G - V_{NP}$ measured from the NP). Calculating the carrier density n from the formula $n = \alpha V_G$, where $\alpha = 7.3\ 10^{10}\ \mathrm{cm}^{-2}\ \mathrm{V}^{-1}$ for a plate capacitor made of a 300 nm SiO_2 layer,[17,18] we find that as the carrier density increases from 4×10^{11} to $14 \times 10^{11}\ \mathrm{cm}^{-2}$ the mobility decreases from 15 000 to 6000 $\mathrm{cm}^2/\mathrm{Vs}$. The strong irregular fluctuations that decorate $R(V_G)$ at low temperature are MRFs that originate from the interference of electrons scattered from a particular distribution of scattering centers as the phases of electron waves are changed by varying V_G. The interference pattern is reproducible on cycling V_G at low T where the scattering centers are immobile.

The resistance was found to be reproducible on cycling from 4 up to 50 K and back but at higher temperatures, some scattering centers may become mobile as has been observed for adsorbed molecules on graphene sheets[19]) and rearrange along the sample. The MRF interference pattern would then be changed when the sample is cooled down to low T again. A change occurred at high temperatures during the T cycle in Fig. 1(b) where the temperature was cycled up to 245 K and back to 4.2 K at constant $V_\Delta = -0.6$ V since $R(T)$ was different on the cooling part of the cycle. This is confirmed by the remeasurement of the MRF pattern at 4.2 K after the cycle [Fig. 1(a)], which shows that the interference pattern is significantly changed. Zooming close to the maximum in the gate voltage dependence [inset of Fig. 1(a)], the value of the resistance at $V_\Delta = -0.6$ V on the black curve corresponds to the value of the resistance at 4.2 K in the $R(T)$ curve at the *beginning* of the T cycle [black curve in Fig. 1(b)] and the value of the resistance at $V_\Delta = -0.6$ V on the gray curve matches the value of the resistance at 4.2 K in the $R(T)$ curve at the *end* of the T cycle (gray curve in Fig. 1).

We conclude that, in the heating curve, the dramatic initial drop of resistance as temperature is increased from 4.2 K represents the decrease in the MRF maximum as the phase coherence decreases with rising temperature,[11] leading to the eventual disappearance of the MRF. To confirm this scenario, we made a systematic study of the correlation between the features in the resistance fluctuations and the temperature dependence of the resistance:

First, we measured $R(V_G)$ at various constant temperatures [Fig. 2(a)] and calculated the root mean square of the resistance fluctuations $\Delta R_{\mathrm{rms}}(T)$ by subtracting the value of $R(V_G)$ at temperature 51 K, where the fluctuations are small, from that at a temperature T. A combination of power laws[11] was tried but the best fit as shown in Fig. 2(b) was to an exponential decay $\exp(-T/T_f)$ with values of the decay constant in the range $T_f = (16 \pm 1)$ K for four different electrode pairs. Interestingly, we find generally that the magnitude of the fluctuations in *resistance* does not vary greatly as gate voltage changes, which means that the *conductance* fluctuations are small near the NP (consistent with the suppression of MRFs near the NP noted earlier[9]). However, the MRFs do tend to vary faster as a function of gate voltage near the NP. The phase coherence length of the MRFs was very approximately estimated to be of order 0.3 μm as indicated by the weakening of $\Delta R_{\mathrm{rms}}(T)$ observed as electrode separation increased.

Second, we set constant values of the gate voltage corresponding either to destructive electron wave interference (a MRF peak) or constructive interference (a MRF valley) in $R(V_G)$ at 4.2 K and then measured $R(T)$ from 4.2 to 250 K. Values of V_Δ chosen included low charge density near the NP [e.g., Figs. 3(a) and 3(b)] and high carrier densities [Figs. 3(c) and 3(d)]. For all cases of V_G set at a MRF peak, there was a sharp decrease in $R(T)$ as T increases from 4.2 K and correspondingly for V_Δ set at a MRF minimum, there was an increase in $R(T)$. This is strong evidence in favor of our proposal that the varying sign of the resistance temperature dependence below 70 K is due to the presence of MRFs at

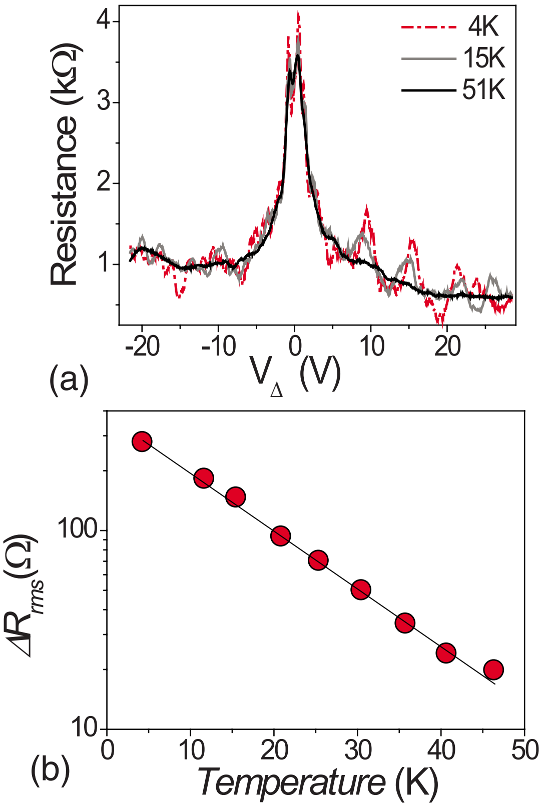

FIG. 2. (Color online) (a) Resistance as a function of gate voltage showing the decrease in magnitude of each extremum of the MRFs as temperature increases. (b) Rms magnitude of the MRFs as a function of temperature averaged over all gate voltages, showing the close agreement with an exponential decay.

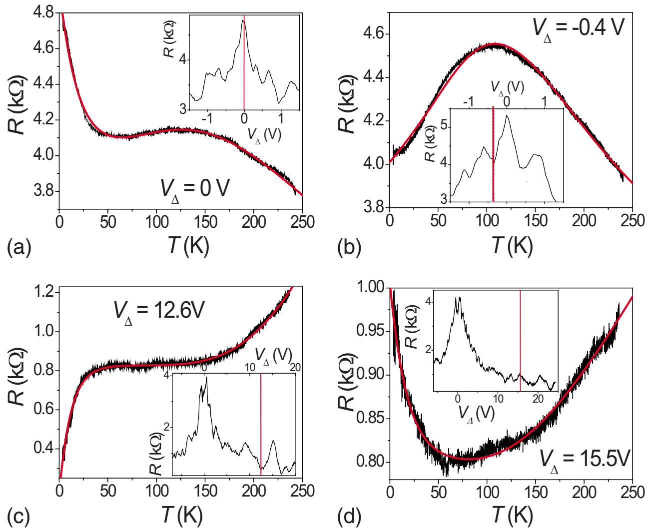

FIG. 3. (Color online) (a) Resistance $R(T)$ as a function of temperature at gate voltage $V_\Delta=0$ at an MRF maximum value at the NP, indicated by the vertical line in the inset. Inset: resistance $R(V_\Delta)$ showing strong resistance fluctuations as a function of gate voltage near the NP. (b) Resistance as a function of T at a MRF minimum value near the NP at $V_\Delta=-0.4$ V. The fit lines in (a) and (b) are fits of the $R(T)$ data for an exponentially decaying MRF term and other terms in Eq. (1), as discussed in the text. (c) Resistance $R(T)$ far from the NP (i.e., at large charge-carrier densities) at a minimum of the resistance fluctuation pattern as a function of gate voltage (as indicated in the inset). (d) Resistance $R(T)$ far from the NP at a maximum of the MRF pattern. The fit lines in (c) and (d) are fits to Eq. (1) without the activation term but with phonon-scattering terms as discussed in the text.

low T (of either sign) that disappear at higher temperatures. These experiments illustrated in Fig. 3 demonstrate that at low T we are observing the essentially monotonic decay of MRF amplitudes with T. It is clear from Figs. 3(a) and 3(b), and from Figs. 3(c) and 3(d), that there are dramatically opposite low-temperature anomalies for two quite similar values of gate voltage V_G. This initially surprising observation is easily explained by the correlation we have demonstrated between these anomalies in $R(T)$ and MRFs in $R(V_G)$ and provides evidence that the origin of the behavior is a fluctuation rather than a systematic effect. Our data in Fig. 3(b) have a similar T dependence to those of Morozov *et al.*[8] for monolayer graphene near the NP, so our scenario accounts for their data as well.

Since the rms magnitude of MRFs over a range of gate voltages decreases *exponentially* with temperature (Fig. 2), we expect that the individual fluctuation features at a fixed gate voltage should follow a similar exponential decay, albeit with some variation arising from the detailed interference pattern in $R(V_\Delta)$ around the chosen gate voltage V_Δ that will affect the thermal dephasing as T increases. Besides the low-T anomalies presented in Fig. 3, we have observed this phenomenon in 20 independent $R(T)$ measurements at different constant V_G in different samples. All the data were well fitted by an exponential decay function and the mean value of the decay constants for the total of 22 individual $R(T)$ measurements was evaluated with 95% confidence limits as $T_f=(15\pm3)$ K. This is very close to the value $T_f=(16\pm1)$ K for $\Delta R_{\mathrm{rms}}(T)$ averaged over V_G values in Fig. 2(b) (which is more accurate as each value is averaged over a very large number of fluctuations).

While the MRF term $R_f\exp(-T/T_f)$ we have proposed is usually a distinct anomaly below 70 K, for completeness we have included in our fits in Fig. 3 a residual resistance R_0 and a linear term aT ascribed to scattering by acoustic phonons.[4,8,20] Close to the NP [Figs. 3(a) and 3(b)], $R(T)$ at high temperatures decreases as T increases, in agreement with earlier work.[8] This suggests thermal excitation of carriers, although this effect in monolayer graphene (contributing a T^2 term to conductivity[21,22]) should be small for $T \ll T_F$, where $T_F \approx 700$ K is the Fermi temperature. We therefore model this increase in carrier density by the usual activation term $\exp(-E_c/k_BT)$, where E_c represents an activation energy (for example, between puddles),

$$R=[R_f\exp(-T/T_f)+R_0+aT]/[1+\exp(-E_c/k_BT)], \quad (1)$$

which is shown as the fit in Figs. 3(a) and 3(b) (with $E_c \sim 50$ meV).

At higher charge densities [Figs. 3(c) and 3(d)], the resistance is much smaller and increases with temperature, consistent with scattering by acoustic phonons (the aT term) and high-energy phonons (described by an additional bT^{-5} resistance term[8] in our fits, although a Bose-Einstein term for phonons of energy ~160 meV (Refs. 23 and 24) fits our data equally well).

In conclusion, we have demonstrated that the low-T anomalies observed in resistance below ~70 K for both low and high charge-carrier densities arise from the decay of resistance fluctuations with temperature. Whether $R(T)$ increases or decreases with T depends on whether scattered wave interference is constructive or destructive for the particular pattern of electron scatterers, so the sign and magnitude of the $R(T)$ anomaly varies considerably as a different constant gate voltage is set. An intriguing aspect of our work is that the decay of MRF features follows an exponential decay law with temperature rather than the weak power laws expected from existing theory.[11] The MRF exponential decay term in Eq. (1) provides a key component for understanding conduction at low temperature for both low and high charge-carrier densities.

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