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Large low-frequency resistance noise in chemical vapor deposited graphene

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We report a detailed investigation of resistance noise in single layer graphene films on Si/SiO_2 substrates obtained by chemical vapor deposition (CVD) on copper foils. We find that noise in these systems to be rather large, and when expressed in the form of phenomenological Hooge equation, it corresponds to Hooge parameter as large as 0.1–0.5. We also find the variation in the noise magnitude with the gate voltage (or carrier density) and temperature to be surprisingly weak, which is also unlike the behavior of noise in other forms of graphene, in particular those from exfoliation. © *2010 American Institute of Physics*. [doi:10.1063/1.3493655]

The electronic properties of graphene has recently been the subject of intense research for both fundamental science and technological applications. Mechanically exfoliated graphene offers the cleanest devices with mobility in the range of $\sim 200\,000\ \mathrm{cm^2/V\,s}$,[1,2] forming the backbone of fundamental phenomena such as the fractional quantum Hall effect,[3,4] or ultrahigh frequency transistors.[5] The exfoliation process is however statistical, and for regular large scale production, several new methods have been suggested including epitaxial growth of graphene on SiC wafers,[6,7] reduction of graphene oxide,[8] and thermally grown graphene from decomposition of hydrocarbon (methane) on transition metal (copper, nickel, iridium, etc.) surfaces.[9–17] The latter metal-based chemical vapor deposition (CVD) technique of realizing large area graphene is of particular interest as it displays excellent electrical (high mobility,[12] low resistance/square, half-integer quantum Hall effect[17]), mechanical (large gauge factor and electromechanical stability[11]) and optical (high transmittance[14]) properties. Moreover, recent developments in transferring large films of single-layer CVD-graphene onto insulating substrates offer great promise in nanoelectronics, transparent electrodes in solar photovoltaics,[14] or flexible/stretchable electronic applications.[16] An important aspect of such applications is the intrinsic electrical noise in CVD-graphene films, which has not been explored so far. A study of electrical noise may also be crucial in understanding the nature of disorder in these materials which can be significantly different from the other forms of graphene.[18,19] In this letter we report the first experimental investigation of low-frequency fluctuations of electrical resistance, often known as the $1/f$-noise or flicker noise, in large-area films of single layer graphene (SLG) grown on Cu-foil and subsequently transferred onto Si/SiO_2 substrate. We find the noise in CVD-graphene to be significantly larger than typical exfoliated graphene devices, along with several surprising features that separates the kinetics of disorder in CVD graphene from other graphene systems.

Recent studies of carrier mobility (μ) (Ref. 20) and $1/f$ noise[18,19,21] in exfoliated graphene on insulating substrates indicates that both static (that gives rise to average resistivity) and time varying (resulting in noise) components of disorder are dominated by the trapped charges at the graphene-substrate interface. This is particularly true at low carrier density (n) where scattering off the Coulomb potential from the trapped charges leads to a linear dependence of graphene conductivity ($\sigma \propto n$).[22] Short range scattering, involving for examples lattice defects or neutral impurities etc., become important only at large n where the Coulomb potentials are largely screened. In CVD-graphene the situation can be very different. The process of etching of the host metal, mechanical stressing during the transfer process etc., have been shown to lead to considerable additional disorder, which manifests in lower μ, and often a clearly visible D-peak in Raman spectroscopy.[15] Indeed, low temperature magnetoresistance measurements in CVD-graphene reveal a short elastic intravalley mean free path, indicating presence of spatially extended defects, such as line defects, dislocations, and ripples.[17] Whether this additional disorder can also cause higher noise in CVD-graphene is not known.

The CVD-graphene used in our experiments was grown by decomposition of ethylene on Cu foils at 875 °C as described in Ref. 13. Then a polymethyl methacrylate (PMMA) layer was spun on top of the graphene layer formed on the Cu foil, and the Cu foil was then dissolved in 1 M iron chloride. The remaining graphene/PMMA layer was thoroughly washed with deionized water and transferred to a Si/SiO_2 substrate. Subsequently, the PMMA was dissolved in hot acetone (80 °C) for one hour. The heavily doped silicon was used as backgate. Following transfer to the Si/SiO_2 substrate a detailed Raman spectroscopy was carried out on all our systems. Figure 1 shows a map of the D-peak ($\sim 1350\ \mathrm{cm^{-1}}$) intensity from a typical section of our CVD-graphene [Fig. 1(a)], and two representative spectra [Figs. 1(c) and 1(d)], which indicates a spatially varying I_{2D}/I_G ratio (I_{2D} and I_G are intensities of the 2D and G bands, respectively). Both features can arise from a spatially nonuniform adhesion/interaction of graphene with the underlying substrate, and associated ripples/local ruptures/line defects/residual byproducts of Cu etching process etc., highlighting significant disorder of non-Coulombic origin.[17] However, the 2D peaks could be described with a single Lorentzian lineshape, confirming single-layer graphene. An electrically contacted (with Au metal pads) device is shown in Fig. 1(d), where a five-probe geometry was used to measure the

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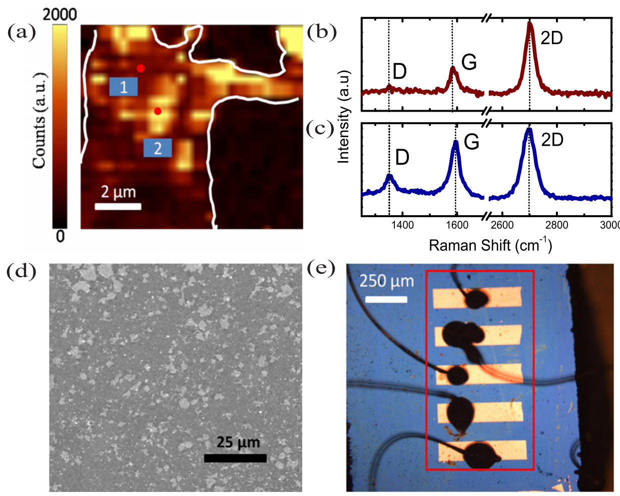

FIG. 1. (Color online) (a) D peak intensity map of a portion of graphene on SiO_2, where the graphene is outlined by thick white line. Raman spectra of graphene corresponding to the positions 1 and 2, in Fig. 1(a), are shown in (b) and (c), respectively. These indicate the spatial variation of D peak intensity, with varying I_{2D}/I_G ratio. (d) SEM image of CVD graphene indicating the ruptures. (e) Optical micrograph of a typical device outlined by the rectangle.

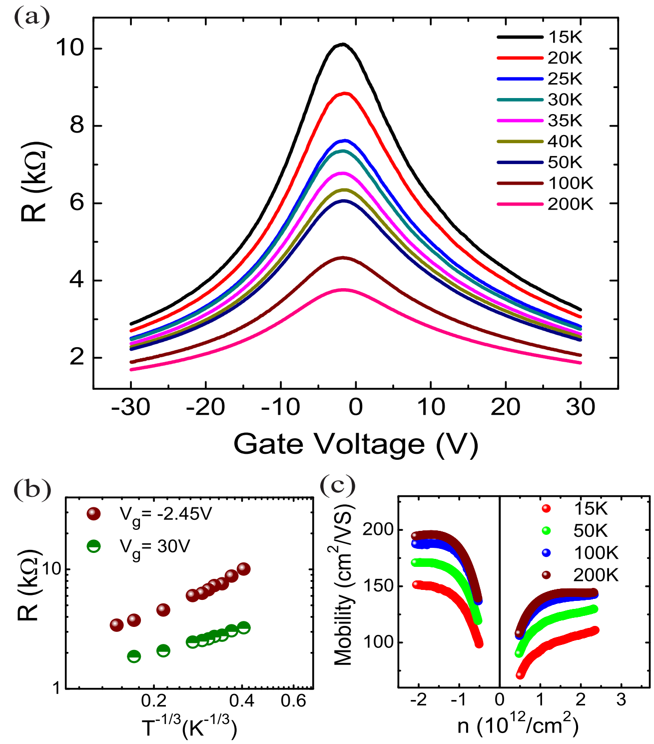

FIG. 2. (Color online) (a) Resistance vs gate voltage characteristics for temperature ranging from 15 to 200 K. (b) Resistance vs $T^{-1/3}$ are plotted for both V_g=−2.45 V (Dirac point) and V_g=30 V (far from the Dirac point), extracted from Fig. 2(a). (c) Mobility vs density (n) for various temperature are extracted from Fig. 2(a).

$1/f$-noise in a dynamically balanced Wheatstone bridge configuration. Both standard time-averaged resistance (utilizing four of the contacts) and noise were measured in low-frequency ac constant current mode. A detail description of the experimental methods are available elsewhere.[18,19] In order to avoid gate leakage-related problems we restricted most measurements at temperature $T \lesssim 200$ K.

The resistance (R)-backgate voltage (V_g) trace of the device in Fig. 1(d) is shown in Fig. 2(a) for various values of T ranging from 15 to 200 K. The sheet resistance was found to be ~680 Ω/sq at room temperature. The Dirac point was found to be low, which we believe to be a combined effect of substrate doping and surface adsorption. R was found to increase sharply with decreasing T at all V_g, reminiscent of the same in ozonization-damaged exfoliated graphene.[23] The T-dependence of R also seems to indicate a Mott-type variable range hopping with $\ln R \propto T^{-1/3}$ [see Fig. 2(b)], although limited range in T or R can make such an analysis relatively inaccurate.

The noise measurements were performed as function of V_g and T. In Fig. 3(a) we show the power spectral density (PSD), S_R, of noise over nearly three decades of frequency at various V_g for T=100 K. The PSD can be normalized with Hooge's phenomenological equation, as follows:

$$S_R = \frac{\gamma_H \langle R\rangle^2}{n A_G f^{\alpha}}, \quad (1)$$

where γ_H is the Hooge parameter, A_G is the area of graphene between the voltage probes, and α is the spectral exponent. In all cases we find $\alpha \approx 1-1.1$, indicating a $1/f$-type spectrum, and hence, a wide distribution of time scales in the kinetics of disorder. The noise amplitude, defined as γ_H/n, was found to be essentially independent of V_g (or n) in both electron and hole-doped regimes. Different devices showed identical behavior as illustrated in Fig. 3(b). This weak variation in noise, found for all T down to 15 K, is in contrast to the n-dependence of the noise amplitude in exfoliated single layer graphene, where γ_H/n decreases rapidly with increasing n on both sides of the Dirac point [see inset of Fig. 3(b)].[19]

Another crucial aspect to note is that the absolute magnitude of γ_H/n is nearly hundred times larger in CVD-graphene in comparison to the exfoliated devices in the same

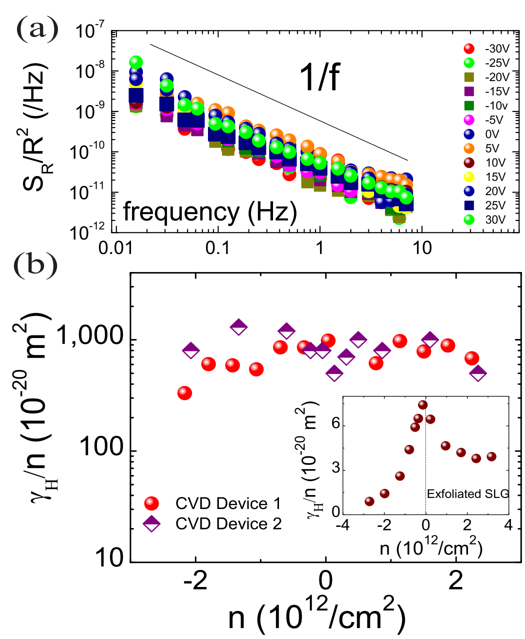

FIG. 3. (Color online) (a) The noise power spectra S_R/R^2 for various gate voltages are shown for T=100 K, showing $1/f$ type behavior. (b) Noise magnitude, γ_H/n are plotted as a function of density (n) for two devices at T=100 K, showing gate voltage independence of noise. The inset shows gate voltage dependence of noise magnitude for exfoliated single layer device which shows that noise decreases on both sides of the Dirac point.

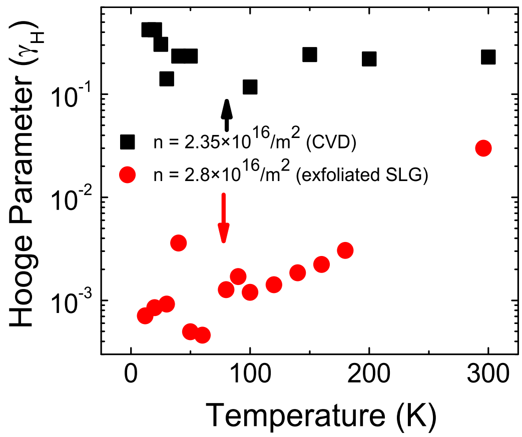

FIG. 4. (Color online) Comparison of Hooge parameter (γ_H) at similar carrier densities for both CVD made graphene and exfoliated graphene devices over a wide range of temperature (15–300 K). CVD graphene device shows two order of higher noise magnitude than exfoliated devices.

range of n. This is highlighted in Fig. 4, where we show the T-variation of γ_H (at $n \approx 2.5 \times 10^{12}$ cm^{-2}) for both graphene systems. For CVD-graphene γ_H is not only independent of T between ~15–300 K, but also displays a γ_H that is about one to two orders of magnitude higher than most graphene based systems.[18,19] The difference appears even larger at lower T, where noise level in exfoliated (or epitaxial) graphene are significantly reduced.

A possible explanation to the weak variation of noise in CVD-graphene can be through the mechanism of correlated number and mobility fluctuations due to the trap states at the graphene-substrate interface.[24] Such a mechanism predicts $S_R \propto 1/n^2$ due to number fluctuations, and $S_R \propto \mu^2$, when mobility fluctuation dominates. In Fig. 2(c), we show that the μ indeed varies weakly with n in our devices, possibly indicating mobility fluctuations to be the dominant source of noise. However, similar substrates have been used for exfoliated graphene that showed much lower noise magnitude.[19] In our CVD graphene, migration of surface adsorbates, such as those incurred during the transfer process, or relaxation of structural defects due to the in-built stress may lead to large noise magnitude. These processes lead to mobility fluctuations, which in an inhomogeneous charge distribution may lead to a gate voltage (as well as temperature)-independent noise. In the inhomogeneous regime, which can persist up to large $|n|$ in highly disordered CVD graphene, the gate voltage is likely to affect relative number of electron and hole puddles rather than the charge density within a particular puddle significantly.[25]

In conclusion, we report experimental investigation of resistance noise in single layer chemical vapor deposited graphene transferred onto a Si/SiO_2 substrate. We find the noise magnitude to be nearly two orders of magnitude larger than exfoliated single graphene, and largely independent of temperature and carrier density. A substrate or surface trap-mediated fluctuation model seems likely, although several details of the noise behavior remains to be understood quantitatively.

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