Conductivity of graphene on boron nitride substrates

S. Das Sarma and E. H. Hwang
Condensed Matter Theory Center, Department of Physics,
University of Maryland, College Park, MD 20742-4111

We calculate theoretically the disorder-limited conductivity of monolayer and bilayer graphene on hexagonal boron nitride (h-BN) substrates, comparing our theoretical results with the recent experimental results. The comparison leads to a direct quantitative estimate of the underlying disorder strength for both short-range and long-range disorder in the graphene on h-BN system. We find that the good interface quality between graphene and h-BN leads to strongly suppressed charged impurity scattering compared with the corresponding SiO$_2$ substrate case, thus producing very high mobility for the graphene on h-BN system.

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An important recent development in the physics and materials science of graphene$^1, ^2$ is the successful fabrication of gated graphene layers on hexagonal boron nitride (h-BN) substrates$^3, ^4$. Since h-BN has the same hexagonal honeycomb lattice structure as graphene itself with an almost matching lattice constant, the expectation has been that graphene on h-BN would have much lower disorder than the generic graphene on SiO$_2$ substrates that has almost universally been studied so far experimentally. This expectation has, in fact, been spectacularly borne out by the recent experiments$^3, ^4$ at Columbia University where both monolayer graphene (MLG) and bilayer graphene (BLG) on h-BN substrates have been shown to have substantially higher (by roughly one order of magnitude or more) carrier mobility than graphene samples on the standard SiO$_2$ substrates$^1, ^2$. In fact, the quality of graphene on h-BN, as measured by transport experiments, appears to be comparable to that of annealed suspended graphene$^2, ^6$, with both systems exhibiting clear fractional quantum Hall effects attesting to their very high mobility.

In the current work, we consider theoretically electronic transport properties of graphene/BN MLG and BLG systems, using the highly successful Boltzmann-Kubo-RPA formalism which has earlier been used$^1$ to study graphene transport on SiO$_2$ substrates, both for MLG$^7, ^8, ^9$ and BLG$^{10}$ systems, as well as for the suspended graphene$^{11}$ system. Our goal is a thorough quantitative understanding of the specific operational features of resistive scattering mechanisms limiting carrier mobility in graphene on h-BN. By demanding quantitative agreement between our calculated graphene (on h-BN) transport properties with the corresponding experimental data$^3, ^4$ for both MLG and BLG systems, we establish the precise role of long-range (e.g. charged Coulomb impurities in the environment) versus short-range (e.g. point defects, neutral scatterers, vacancies) disorder in graphene on h-BN systems. We obtain excellent agreement with the experimental data$^3, ^4$ using very reasonable disorder parameters, establishing that the good interface quality between graphene and h-BN (e.g. lack of dangling bonds) leads to strongly suppressed charged impurity scattering compared with the corresponding SiO$_2$ substrate situation$^7, ^8, ^9$, thus providing very high mobility for the graphene on h-BN system. The relative suppression of long-range scattering compared with the short-range scattering also leads to rather nonlinear-looking MLG conductivity as a function of gate voltage (i.e. carrier density) for graphene on h-BN substrates compared with the SiO$_2$ substrates, thus explaining the peculiar experimental finding that the observed BLG (MLG) conductivity on h-BN substrates$^3, ^4$ manifests linear (nonlinear) conductivity as a function of the gate voltage. Our theory also naturally explains the weaker observed temperature dependence of MLG conductivity than the BLG conductivity for graphene on h-BN substrate. The actual conductivity of MLG/BN or BLG/BN$^3, ^4$ is determined by the detailed interplay between the long-range and the short-range disorder in the relevant system along with the distinct screening properties of the graphene carriers as it is in the usual graphene on SiO$_2$ substrates$^1, ^7, ^8, ^9, ^10, ^12$.

The graphene conductivity $\sigma$ is given by$^1$

$$\sigma = \frac{e^2}{2} \int \! \! d\varepsilon D(\varepsilon) \nu_k^2 \tau(\varepsilon) \left( \frac{\partial f}{\partial \varepsilon} \right) , \quad (1)$$

where $f = f(\varepsilon_k)$ is the Fermi distribution function, $D(\varepsilon)$ is the density of states, $\nu_k = d\varepsilon_k/dk$ is the carrier velocity, and $\tau(\varepsilon)$ is the transport scattering (or relaxation) time which depends explicitly on the effective disorder scattering potential $'V'$:

$$\frac{1}{\tau(\varepsilon)} = \frac{2\pi}{\hbar} \sum_\alpha \eta^\alpha_i(z) \int \frac{d^2k'}{(2\pi)^2} |V_{kk'}(z)|^2 g(\theta_{kk'}) (1 - \cos \theta_{kk'}) \delta(\varepsilon_k - \varepsilon_{k'}) , \quad (2)$$
where $\varepsilon_k$ is the graphene carrier energy dispersion for 2D wave vector $k$, $\theta$ is the position of the impurity whose concentration is defined by $n_i^\alpha$ with $\alpha$ denoting the kind of impurity (e.g. long-range or short-range), $g(\theta)$ denotes a known chiral matrix element form factor determined by the band structure (and is therefore different for MLG and BLG), and $(k, k')$ are the incoming and outgoing carrier 2D wave vector due to the impurity scattering potential $V_{kk'}(z)$. Since the details of the transport theory for graphene have been discussed earlier in the literature [4, 5, 10], we only make a few comments on the calculational aspects of our theoretical results presented in this work: (i) the substrate h-BN is characterized by its static dielectric constant $\kappa_{BN} = 7.0$ [11], leading to an effective background dielectric constant $\kappa = 4.0$, which enters into the definition of the effective disorder potential; (ii) the effective disorder potential $'V'$ entering Eq. (2) is taken to be the screened disorder where the screening is by the static graphene (MLG or BLG) dielectric function $\epsilon(q, T)$ which has been calculated earlier in refs. [14, 15] respectively for MLG and BLG; (iii) we include two types of disorder in our theory, the long-range disorder characterized by randomly distributed charged impurity centers with 2D density of $n_i$ located at the graphene-BN interface and the short-range disorder characterized by an effective strength of $n_d V_0^2$ denoting a white-noise delta-correlated local disorder. (We emphasize that both long- and short-range disorder are necessary for quantitative and qualitative understanding of experimental data.)

The theory is characterized by two parameters, $n_i$ and $n_d V_0^2$, describing long-range and short-range disorder, respectively. In principle, the effective separation $d$ between the location of the charged impurity centers and the 2D graphene layer could also be an additional physically relevant parameter in the transport theory [4, 5, 8], but we put $d = 0$ throughout this paper, keeping the number of free parameters a minimum (only two) and assuming that the random charged impurity centers are located at the graphene/BN interface as consistent with the very high quality of the h-BN crystals used in refs. [3, 4]. For obtaining our theoretical transport results we have varied the parameters $n_i$ and $n_d V_0^2$ arbitrarily over a wide range, obtaining the best regression fit to the high-density data of refs. [3, 4] as shown in Figs. 1 and 2. (We have also used different values of $'d'$ using the charged impurity separation as a tuning parameters, but our results are qualitatively unaffected by an adjustable $d$.)

We first show our theoretical results valid at “high” carrier density ($n$) defined as $n \gtrsim n_i$ away from the minimum conductivity Dirac point regime, where the density fluctuations associated with the inhomogeneous puddle formation can be safely neglected. In Figs. 1 and 2 we show our calculated conductivity (at $T = 0$): $\sigma(n)$, as a function of the carrier density for a few different values of the disorder parameters choosing the parameters such that we get essentially exact quantitative agreement away from the Dirac point ($n > n_i$) with the experimental data of ref. [9] for MLG/BN (Fig.1) and BLG/BN (Fig.2) systems. In each figure, we present results for both MLG and BLG systems for a fixed set of values of the disorder parameters $n_i$ (long-range) and $n_d V_0^2$ (short-range) with the results of Fig. 1 (2) showing quantitative agreement with the corresponding experimental data for MLG (BLG) on h-BN in ref. [9]. In each figure we present the individual conductivity limited by long-range and short-range scattering as well as the total conductivity.

Three qualitative features of our theoretical results in Figs. 1 and 2 stand out: (i) for fixed disorder, MLG conductivity is always larger than BLG conductivity for all densities although they approach each other at very high density as expected; (ii) the quantitative values of the disorder parameters (i.e. $n_i$ and $n_d V_0^2$) necessary in Figs. 1 and 2 for obtaining agreement with the experimental data [3, 4] for graphene on h-BN substrates are typically much (by more than an order of magnitude) smaller than that needed for agreement between theory and experiment with the corresponding graphene on SiO$_2$ substrates (e.g. refs. [5, 8]) — this is particularly true for the charged impurity density $n_i$ which has the remarkably small value of $0.3 \times 10^{11} - 1.0 \times 10^{11}$ cm$^{-2}$ for graphene on h-BN substrates compared with $n_i > 10^{12}$ cm$^{-2}$ for graphene on SiO$_2$ substrates (we note that short-range disorder
characterized by \( n_dV_0^2 \) seems comparable in strength for h-BN and SiO\(_2\) substrates with h-BN having somewhat smaller values; (iii) the MLG conductivity results for h-BN substrates are much more sublinear than for the corresponding SiO\(_2\) substrate case clearly establishing the much weaker role of long-range charged impurity scattering in h-BN systems compared with SiO\(_2\) systems.

It is easy to show theoretically [1, 7, 8] using Eq. (2) that the charged impurity scattering limited MLG conductivity \( \sigma_{MLG} \) on h-BN substrates is given approximately by \( \sigma_{MLG}^i \approx 25.7(e^2/h)(n/n_i) \) whereas the short-range scattering limited conductivity is given by \( \sigma_{MLG}^s \approx 350(e^2/h)/(n_dV_0^2) \) where \( n_dV_0^2 \) is measured in \((eV)^2\) units. Our MLG numerical results for long-range scattering shown in Figs. 1 and 2 obey these analytical relations exactly with the net conductivity being given by \( \sigma = (\sigma_{i}^{-1} + \sigma_{d}^{-1})^{-1} \). For the BLG on h-BN substrates, a simple analytic relation can only be derived for the short-range scattering-limited conductivity \( \sigma_{BLG}^0 \approx 66.7(e^2/h)(n/n_dV_0^2) \). This is linear in carrier density, with \( n \) in units of \( 10^{12} \text{ cm}^{-2} \) and \( n_dV_0^2 \) in units of \((eV)^2\). The long-range disorder leads to \( \sigma_{BLG} \sim n^\alpha \) where \( \alpha \approx 1 - 1.3 \) depending on the parameter regime, and no simple analytic relationship can be derived except at very low BLG carrier density where the Coulomb disorder is effectively completely screened out since the BLG screening wave vector becomes much larger than the Fermi wave vector. For this very low carrier density regime (\( \ll 10^{12} \text{ cm}^{-2} \)), the charged impurity disorder limited BLG conductivity becomes linear in carrier density (i.e., \( \alpha = 1 \)) obeying the approximate relationship: \( \sigma_{BLG} \sim 15(e^2/h)(n/n_i) \). We mention, however, that this formula is not useful for \( n < n_i \) since density inhomogeneity effects associated with puddles would dominate close to the charge neutrality point.

The agreement, using reasonable values of disorder parameters, between theoretical results presented in Figs. 1 and 2 with the experimental data [3, 4] of the Columbia group indicates that graphene on h-BN indeed has substantially lower long-range Coulomb disorder in its environment than graphene on SiO\(_2\) substrates, most likely due to the high-quality graphene/BN interface without any dangling bonds as already speculated in ref. [8]. A direct consequence of this reduced Coulomb scattering is the manifestly sublinear \( \sigma(n) \) observed in the MLG/BN system to be contrasted with the linear \( \sigma(n) \) in the MLG/SiO\(_2\) system [1, 7] except at very high densities. We note that our theory indicates a direct way of estimating the strength of both long- and short-range disorder from the high-density MLG/BN \( \sigma(n) \) data by obtaining the slope \( d\sigma/dn \) at high-density (which gives \( n_i \)) and by obtaining the intercept of the high-density \( \sigma(n) \) extrapolated to \( n \rightarrow 0 \), which gives \( n_dV_0^2 \).

In Fig. 3, we present our theoretical results for the temperature dependence of the MLG (Fig. 3) and BLG (Fig. 4) conductivity, \( \sigma(n, T) \), on h-BN substrates. These results are again valid (similar to those in Figs. 1 and 2) at high carrier density \( (n > n_i) \) where density inhomogeneity effects are negligible. All phonon effects [10, 17] are neglected here with the temperature dependence arising entirely from the temperature dependence of the screening function and the energy-averaging associated with the finite-temperature smearing of the Fermi surface [3]. The first effect (“screening”) produces weak metallic temperature dependence (i.e. \( \sigma \) decreasing with increasing \( T \)) since screening weakens at higher temperatures whereas the second effect (“thermal averaging”)...
produces weak insulating temperature dependence (i.e. $\sigma$ decreasing with increasing $T$). Although the temperature dependence is weak, as is obvious from Fig. 3, the theoretical behavior of $\sigma(T)$ is qualitatively consistent with the experimental observations \cite{18} away from the Dirac point: (i) MLG manifests weak metallic $T$-dependence, and (ii) BLG manifests weak insulating $T$-dependence. Experimentally, both systems manifest insulating $\sigma(T)$ at the Dirac point where density inhomogeneity effects dominate \cite{18}, but at higher density ($n > n_i$) our results are consistent with experimental finding of ref. \cite{3}.

Finally, we consider in Fig. 4 the low-density transport in graphene/BN systems, where the results (valid for $n > n_i$) shown in Figs. 1–3 do not apply. At low carrier density $n ( < n_i)$, which is very low ($\lesssim 10^{10}\text{cm}^{-2}$) for the graphene/BN system because of its extremely weak Coulomb disorder, the graphene layer is known \cite{1, 8, 19–23} to break up into inhomogeneous puddles due to the charged impurities. Microscopic self-consistent calculations \cite{10, 23} calculation of transport using $n_{rms} = n_i$, where $n_{rms}$ is the root-mean-square fluctuation in the carrier density due to the puddles induced by the charged impurities. Microscopic self-consistent calculations \cite{10, 23} show that $n_{rms} \approx n_i$ is a reasonable qualitative approximation for the density inhomogeneity around the Dirac point. Our $T = 0$ effective medium theory transport results (using the Boltzmann-Kubo-RPA transport formalism) are shown in Fig. 5 for both MLG/BN and BLG/BN systems. The most important features of Fig. 5 are: (i) the high density ($n \gtrsim n_i$) results shown in Figs. 1 and 2 remain valid; (ii) near the Dirac point (for $n < n_i$), $\sigma(n)$ saturates with a non-universal disorder-dependent minimum conductivity \cite{11, 18, 23} whose value is roughly given by $2 \sim 10 \left( e^2/\hbar \right)$ consistent with the experimental observations \cite{3, 4}.

We have also carried out our effective medium theory calculation at finite temperature to include the puddle effects on $\sigma(n, T)$. These results (not shown) agree with the results shown in Figs. 3 and 4 at high densities ($n > n_i$), but for low carrier densities ($n < n_i$) we qualitatively recover the experimentally observed strongly insulating $\sigma(T)$ induced by the puddles.

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