Conductance fluctuations and field asymmetry of rectification in graphene

C. Ojeda-Aristizabal, M. Monteverde, R. Weil, F. Ferrier, S. Guéron, and H. Bouchiat
LPS, Univ. Paris-Sud, CNRS, UMR 8502, F-91405 Orsay Cedex, France

We investigate conductance fluctuations as a function of carrier density \( n \) and magnetic field in diffusive mesoscopic samples made from monolayer and bilayer graphene. We show that the fluctuations’ correlation energy and field, which are functions of the diffusion coefficient, have fundamentally different variations with \( n \), illustrating the contrast between massive and massless carriers. The field dependent fluctuations are nearly independent of \( n \), but the \( n \)-dependent fluctuations are not universal and are largest at the charge neutrality point. We also measure the second order conductance fluctuations (mesoscopic rectification). Its field asymmetry, due to electron-electron interaction, decays with conductance, as predicted for diffusive systems.

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Reproducible conductance fluctuations (CF) are one of the most striking signatures of phase coherent transport \[1\]. The conductance of a mesoscopic sample results from interference between all wave packets traversing the sample. This interference pattern is sensitive to variations in disorder configuration, Fermi energy or magnetic flux, leading to reproducible CF as one of these parameters is changed. In diffusive or chaotic systems the CF amplitude has been shown to be universal \[1–3\] and ergodic, i.e. independent of the mechanism of phase randomization (magnetic field, Fermi energy, configuration of impurities for diffusive systems, sample shape for ballistic systems). The CF amplitude is of the order of \( e^2/h \), with a coefficient which only depends on the symmetry class of the mesoscopic system. The typical correlation energies \( E_\varphi \) and fields \( B_\varphi \) of the fluctuations depend upon the typical time \( t_{\text{int}} \) and area \( A_{\text{int}} \) over which interference occur: \( E_\varphi = h/t_{\text{int}} \) and \( B_\varphi = \Phi_0/A_{\text{int}} \), with \( \Phi_0 = h/e \)[2]. CF have been extensively investigated in metallic and semiconducting systems \[1–3\]. The recently discovered graphene \[4\] provides a unique system in which the Fermi energy and diffusion constant can be tuned at will, over a broad carrier density range extending from hole to electron metallic conduction. Theoretical simulations of CF in graphene suggest a possible enhancement of the fluctuation amplitude with respect to standard mesoscopic samples, depending on the strength or nature of disorder (intervalley scattering) \[5–7\]. On the experimental side CF have been reported by several groups \[8–13\]. But to our knowledge the present work is the first complete investigation of their correlations and amplitudes as a function of Fermi energy and magnetic field, for both monolayer (ML) and bilayer (BL) graphene. The importance of the comparison lies in the fact that whereas ML and BL have similar resistivities and thus mean free paths (see Fig. 3), the (massless and massive) carriers have different velocities because of the different dispersion relations in these two materials. Thus the diffusion constants and therefore correlation energies and fields will have different carrier density dependences, providing a powerful test of the applicability of the theory of mesoscopic fluctuations in those systems. We find that the variations with carrier density \( n \) of the correlation field and energy are well related to those of the diffusion coefficient. We also find, in contrast with \[10\], that the amplitude of \( n \)-dependent fluctuations are largest near the charge neutrality point.

We also measure the second order conductance fluctuations \[21\] which, unlike linear conductance, are a probe of Coulomb interaction and screening, an important issue in graphene. Second order CF are inherent to systems lacking spatial inversion symmetry (because of random disorder in diffusive systems or geometry in ballistic systems). They stem from current-induced changes in the carrier density, which in turn, via Coulomb interaction, modify the electrostatic potential landscape, thereby inducing a current-dependent, or second order, change in the conductance of mesoscopic samples. Unlike the first order conductance which is even in field, the second order conductance has an odd part in field which was calculated for ballistic \[10\] and diffusive \[17\] systems. Whereas this mesoscopic rectification was experimentally investigated in ballistic mesoscopic systems (GaAs/GaAlAs quantum dots, Aharonov Bohm rings, carbon nanotubes \[19\] \[20\]), we provide in this letter the first measurement of second order CF in a diffusive system. We find, in qualitative agreement with theoretical predictions, that the odd in field rectification decreases with conductance.

Both ML and BL graphene samples (the nature of which was confirmed by Raman spectroscopy) were exfoliated and deposited onto doped silicon substrates with a 285 nm thick oxide. The electrodes were fabricated by electron beam lithography and sputter deposition of 40 nm thick palladium, providing low contact resistances (200 \( \Omega \) for the ML and 20 \( \Omega \) for the BL). The dimensions are \( W = 2.7 \mu m, L = 0.8 \mu m \) and \( W = 4.8 \mu m, L = 0.7 \mu m \) for the ML and BL, respectively (Fig. 3). The carrier density \( n \) is controlled for both systems by the voltage \( V_g \) applied to the doped silicon back gate, via \( n = k_F^2/\pi = C(V_g - V_0) \), with \( C \) the gate capacitance per unit area, \( k_F \) the Fermi wave vector, and \( V_0 \) the gate voltage of the charge neutrality point. The transport pa-
rameters of both samples were determined via classical magnetoresistance measurement, described in [22]. We found that the transport mean free path $l_T$ of both ML and BL varies between 25 and 100 nm over the 60 V gate-voltage range probed. Thus $l_T$ is much smaller than the sample dimensions, so that both samples are in the diffusive regime. Two terminal resistance measurements were performed at 60 mK in a dilution refrigerator with resistive lines filtered at room temperature. The ac current amplitude $I_0$ was adjusted between 5 and 50 nA, depending on the sample resistance, to avoid heating while optimizing the signal to noise ratio. The first and second harmonics $V_1$ and $V_2$ of the ac voltage were measured with a low noise voltage preamplifier and a lock-in amplifier. The first and second order conductances were determined via $G_1 = I_0/V_1$ and $G_2 = 2V_2I_0/V_1^2$ [21]. The magnetic field was limited to less than 0.4 T, so that the contribution of Shubnikov de Haas oscillations is negligible. The CF of $G_1$ were measured as a function of B and $V_g$, for both the ML and BL (see Fig. 1). The $V_g$ dependent fluctuations were analyzed in 3 V-wide windows, over which the density $n$ can be considered practically constant. The fluctuations of $G_1$, even in B as expected in a two probe measurement, were characterized by their correlation functions and their amplitude $\delta G_1$, defined as the square root of their variance. The histogram of these fluctuations is Gaussian over the whole parameter range. The correlation gate voltage and field were determined in the following way. We calculated the Fourier power spectrum of the fluctuations as a function of $k$, the conjugate variable of $B$ or $V_g$, for each set of field or gate voltage data corresponding to a given average carrier density. This spectrum is the Fourier transform of the correlation function of the fluctuations. Each dataset yielded exponential-like functions which correspond to a lorentzian correlation function in the direct space (Fig. 2). The decay $\exp(-kx_c)$ at low $k$ yields the correlation gate voltage or field $x_c$ for each dataset. The correlation energy $E_c$ is related to the $V_g$ correlation scale $V_c$ by $E_c = V_c h v_F \sqrt{\pi C/(4eV_g)}$ for the ML and $h^2 \pi CV_c/(2em_{eff})$ for the BL, with $m_{eff} = 0.03m_e$ the (constant) low energy effective mass in the BL. These relations are incorrect near the charge neutrality point (CNP) because of density inhomogeneities, so that we deduced $E_c$ from $V_c$ via $k_F(V_g)$ determined in [22].

In Fig 2 we compare the variations of $E_c$ and $B_c$ to the predictions [2] $E_c = hD/L_{min}$ and $B_c = \Phi_0/(L_{min}W_{min})$, where $L_{min} = min(L,L_T,L_c)$ and $W_{min} = min(W,L_T,L_c)$ correspond to the typical longitudinal and transverse length of interfering trajectories; $L_T = \sqrt{hD/k_T}$ and $L_c$ are respectively the thermal and the phase coherence length. We first determine the diffusion coefficient $D = \sigma/(e^2\nu(E_F)) = v_F l_T/2$, where $\sigma$ is the conductivity and $\nu(E_F)$ is the density of states, $v_F$ is constant ($10^6$ m/s) for the ML and is $\hbar k_F/m_{eff}$ for the BL. Both $l_T$ and $k_F$ were extracted in [23]. We find that at 60 mK $L_T$ varies between 0.7 and 1.8 $\mu$m, so that $L < L_T < W$ for both samples over the entire density range investigated. Thus $W_{min} = L_T$ and $L_{min} = L$. Consequently $E_c$ is expected to vary like the Thouless energy $E_{Th} = h/T_D = hD/L^2$, and $B_c$ like $\Phi_0/(LL_T)$ [14]. As shown in Fig. 2, this is indeed what is found: the correlation energy deduced from the experimental data follows $E_c = 5 E_{Th}$, and the correlation field follows $B_c = 6 \Phi_0/(LL_T)$ for the ML and $B_c = 4 \Phi_0/(LL_T)$ for the BL. In particular, we find the expected linear dependence of $E_c$ with $D$, which itself varies like $V_g$ for the BL and $\sqrt{V_g}$ for the ML. This stems from the fact that in both samples the conductance varies linearly in $n \propto V_g$ within logarithmic corrections [22], and that $\nu(E_F)$ is independent of $V_g$ for the BL and varies like $\sqrt{V_g}$ for the ML. Similarly $B_c \propto \Phi_0/LL_T$ is expected to vary like $D^{-1/2} \propto V_g^{-1/2}$ and $V_g^{-1/4}$, as observed respectively for the BL and ML. The numerical factors may be explained by the samples’ aspect ratios, but more theory is needed to assess this point.

We now discuss the CF amplitude. As is visible in figures [10] and [14], the $V_g$-dependent CF are greater near the CNP. This is quantified by the fluctuation amplitude $\delta G_1$ plotted as a function of $V_g$ in Fig. 3. In contrast, the $B$-dependent fluctuations do not significantly depend on the density $n$. This non ergodicity of the fluctuations in

![Fig. 1: CF as a function of magnetic field (left) and gate voltage (right). A linear fit was subtracted to remove the average conductance. Thick red (resp. thin blue) curves are measured around $V_g = V_0$, the charge neutrality point (CNP) (resp. at high carrier density, corresponding to $V_g - V_0 = 40.5$ V for the ML and $V_g - V_0 = 15$ V for the BL). The CNP is at $V_0 = 15.5$ V for the ML and $V_0 = -6$ V for the BL. It is clear (b,d) that the amplitude of $V_g$-dependent CF is larger at the CNP than at high density, both for the ML and the BL. Panel d also shows the reproducibility of the CF, via two nearly indistinguishable successive gate voltage sweeps (open dots and red line).]
graphene may be a consequence of the spatial inhomogeneities of n close to the CNP. Indeed, in a good conductor (large g), changing the Fermi energy is equivalent to changing the disorder configuration, and induces CF of order $e^2/h$. In graphene, on the other hand, it has been shown [23] that near the CNP the system breaks into conducting puddles of electrons and holes, and that transport takes place along an intricate percolating network of these n and p-type regions [6]. Thus, as pointed out in [5], the change in configuration induced by a change of $V_g$ in this region may induce a much larger variation of conductance than a change in the disorder configuration in a good conductor. In contrast, the magnetic field does not affect the network but only the phases of the wave functions, which explains why the fluctuations with B are independent of gate voltage [5]. Note that these results are in contradiction with those of [10] who found a smaller CF amplitude close to the CNP. This discrepancy could be due to a different nature of disorder in the samples [5]. For the ML, we find the B-dependent CF amplitude to be $0.4 \pm 0.1 \, e^2/h$, independent of $V_g$. Theory [2] predicts $\delta G = 0.7 \sqrt{W/L} \, e^2/h$ when the distance between electrodes L is smaller than $L_T$. This yields $\delta G = 1.2 \, e^2/h$ for the ML, which is three times the value measured in this work (Fig. 3).

We now turn to second-order non-linear CF in monolayer graphene. As mentioned in the introduction, current through a mesoscopic sample induces charge accumulation around impurities and sample edges. This bias induced change of electronic density $\delta n(r, V)$ modifies (via Coulomb interactions) the electrostatic potential landscape throughout the sample by $\delta U_{dis}(\gamma_{int}, V)$, where $\gamma_{int}$ quantifies e-e interactions [13]. As a result, CF acquire a non linear bias dependent contribution: $G = G[U_{eq}] + \delta G[\delta U_{dis}(\gamma_{int})]$, where $U_{eq}$ is the potential in zero bias. $G_1 = G[U_{eq}(r)]$ is the linear conductance, and $G_2 = (\partial G[\delta U_{dis}, \gamma_{int}]/\partial V)_{V=0}$ is the second order conductance. In a magnetic field B, $\delta n(r, V)$ and $\delta U_{dis}(\gamma_{int})$ contain a component which is even, as well as another which is odd in B, which can be viewed as a mesoscopic Hall voltage. Contrary to $G_1$ which is even in B, in the presence of interactions $G_2$ has a component which is odd in B, $G_2^{AS}$. The ratio between the even (symmetric) variance $\delta G_{int}^S$ and the odd (antisymmetric) variance $\delta G_{int}^{AS}$ of $G_2$ is predicted to be independent of conductance in ballistic systems, but in diffusive systems it should vary like $\delta G_{int}^{AS} = \gamma_{int} (\pi e^2/h)^2$ [17]. Figure 4 shows $G_2^S$ and $G_2^{AS}$ at two different carrier densities (close and far from the CNP), as well as the ratio of the odd to even conductance $r = \delta G_{int}^{AS}/\delta G_{int}^S$. We find that r significantly decreases as carrier density increases, in contrast to ballistic GaAs/GaAlAs rings [20], where r was nearly independent of the dimensionless conductance g. To compare to the predictions for diffusive systems, $r = \gamma_{int}/g$, we plot in Fig. 4 r($V_g$), and find that $r \approx (7 \pm 1)/g$. According to [10], the interaction constant $\gamma_{int}$ is related to the geometrical capacitance C and electro chemical capacitance $C_\mu = (1/(\nu e^2) + 1/C)^{-1}$ by $\gamma_{int} = C_\mu/C$. Thus $\gamma_{int} = 1/(1 + C/(\nu e^2))$ [10]. Perfect screening (in materials with large densities of states) corresponds to
conductance to find that its field asymmetry decreases with $g$, in agreement with theoretical predictions for diffusive systems. This indicates strongly screened electron-electron interactions in graphene.

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