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Gate electrostatics and quantum capacitance in ballistic graphene devices

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We experimentally investigate the charge induction mechanism across gated, narrow, ballistic graphene devices with different degrees of edge disorder. By using magnetoconductance measurements as the probing technique, we demonstrate that devices with large edge disorder exhibit a nearly homogeneous capacitance profile across the device channel, close to the case of an infinitely large graphene sheet. In contrast, devices with lower edge disorder (<1 nm roughness) are strongly influenced by the fringing electrostatic field at graphene boundaries, in quantitative agreement with theoretical calculations for pristine systems. Specifically, devices with low edge disorder present a large effective capacitance variation across the device channel with a nontrivial, inhomogeneous profile due not only to classical electrostatics but also to quantum mechanical effects. We show that such quantum capacitance contribution, occurring due to the low density of states across the device in the presence of an external magnetic field, is considerably altered as a result of the gate electrostatics in the ballistic graphene device. Our conclusions can be extended to any two-dimensional (2D) electronic system confined by a hard-wall potential and are important for understanding the electronic structure and device applications of conducting 2D materials.

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I. INTRODUCTION

An accurate picture of the electrostatic charge induction mechanism in field effect devices made of graphene or other two-dimensional (2D) crystals [1,2] is necessary to understand the physical properties of these novel nanomaterials [1–18] as well as to guide their exploitation in novel electronic [18], spintronic [19], and optoelectronic [20] applications. Yet, the precise details of the gate electrostatics are far from being understood in these atomically thin and finite-size systems [4–10]. In general, the relation between the gate voltage V_g and the induced average carrier density n is complicated in 2D materials, arising not only from classical electrostatic interactions but also from quantum mechanical effects related to modifications in their band structure under gating [7–10]. The relation $V_g(n)$ is a function of the total capacitance per area of the system C_{tot} and the elementary charge e and can be expressed in terms of classical (electrostatic) C_c and quantum capacitance C_q contributions as [8,9]

$$V_g = en[C_c^{-1} + C_q^{-1}(n)] = enC_{\text{tot}}^{-1}(n) \quad (1)$$

where the dependence of C_q on n makes the capacitance-voltage relationship nonlinear in these systems [8,9]. Thus, C_{tot} is a carrier density dependent property, sensitive to the device geometry, edge morphology and external measurement conditions such as the presence of a magnetic field [9,10]. Furthermore, it exhibits local variations since the charge carrier density [$n(\vec{r})$] is not uniform in these finite-size systems:

$C_{\text{tot}}(\vec{r})/e \equiv \alpha_{\text{tot}}(\vec{r}) = n(\vec{r}) \cdot V_g^{-1}$ [5–8]. Here, \vec{r} represents the coordinate(s) in which the carrier density varies and α_{tot} represents the so-called lever arm of the system.

We focus on the capacitance-voltage characteristics of high-quality, ballistic devices made of graphene, the frontrunner of the 2D materials [1,2]. While the injection of charge carriers in graphene field effect devices is often approximated by the infinite parallel-plate capacitor model [1,2] where $n^\infty = \alpha_c^\infty V_g = (C_{\text{ox}}/e)V_g$ and C_{ox} is the gate oxide capacitance per unit area, the presence of hard-wall boundaries leads to accumulation of charges close to the device edges as a consequence of the fringing electrostatic field [7,8] [Fig. 1(a)]. This results first in a generic, inhomogeneous, and position dependent effective capacitance profile of classical origin $\alpha_c(x) = C_c(x)/e$ across the device channel (x direction) which qualitatively follows a divergent $1/\sqrt{x}$ dependence towards edges. Quantitatively, $\alpha_c(x)$ will depend on the entire device geometry, with the dominating factor being the ratio W/b between sample width W and the distance to the gate electrode b : $\alpha_c(x)$ increases for smaller W/b , especially for $W/b \leq 1$ [4,7–9]. Second, the presence of this varying $\alpha_c(x)$ results in a nontrivial potential profile $U(x)$ across these devices, a fact that may lead to the presence (or even dominance) of a quantum contribution $\alpha_q(x) = C_q(x)/e$ within the total effective capacitance $\alpha_{\text{tot}}(x) = C_{\text{tot}}(x)/e$ of the system [7–10].

Such complex gating dependencies are, however, largely unexplored and remain experimentally inconclusive. In particular, the existence of inhomogeneous capacitance profiles of classical origin $\alpha_c(x) = C_c(x)/e$ has been experimentally verified in high-quality, narrow graphene strips up to a certain modulation $\alpha_{\text{tot}}(x)^{(\text{max})}/\alpha_c^\infty = C_{\text{tot}}(x)^{(\text{max})}/C_{\text{ox}}$

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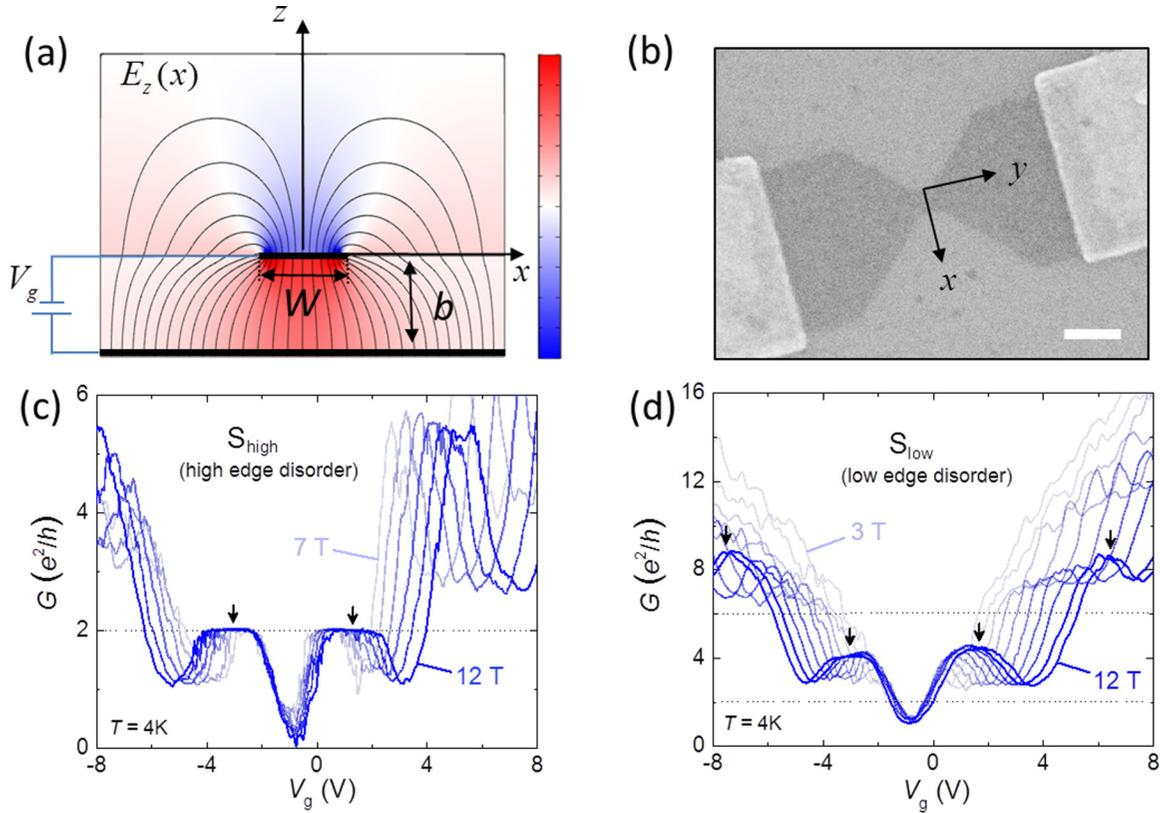


FIG. 1. Sample fabrication and electrical characterization. (a) Electric-field distribution $E_z(x)$ around a gated graphene device with $b = W = 100$ obtained by solving the Poisson equation in the device [27]. (b) Scanning electron micrograph of a graphene nanoconstriction device. Scale bar is 200 nm. (c) Conductance G vs gate voltage V_g in a nanoconstriction of type S_{high} at different magnetic fields (from 7 to 12 T in steps of 1 T) at $T = 4$ K. (d) Conductance G vs gate voltage V_g in a nanoconstriction of type S_{low} at different magnetic fields (from 3 to 12 T in steps of 1 T) at $T = 4$ K. Arrows in panels (c) and (d) indicate the experimental points (V_g, G) taken to calculate $\alpha_v(d_v)$ in Fig. 2 for $B = 12$ T, i.e., the center of the plateau for sample S_{high} [5,6] and G^{max} for S_{low} . Corresponding points are taken for other magnetic fields. Dotted lines indicate the expected quantization values for graphene.

[5], where $\alpha_{\text{tot}}(x)^{(\text{max})}$ is the maximum measured effective capacitance across the device. However, there are strong discrepancies between different experimental studies [3–6,11–17], and their correspondence with theoretical calculations, too [7–10]. Some studies exhibit capacitance profiles with modulation values across the device $\alpha_{\text{tot}}(x)^{(\text{max})}/\alpha_c^\infty \sim 2$, close to those given by classical electrostatic predictions [5]; others report much smaller capacitance modulations [$\alpha_{\text{tot}}(x)^{(\text{max})}/\alpha_c^\infty < 1.2$] across similar nanostructures [6], yet others observe no modulation at all [$\alpha(x) = \alpha_c^\infty$] [3], or simply neglect the effect by assuming a constant effective capacitance [11–13]. In a wider perspective, transport studies carried out in high-quality graphene devices display variations in how inhomogeneous gating is accounted for, whether these effects are included [4–6,14,15] or not at all [3,11–13,16,17] during the interpretation of the electrical measurements.

Here, we probe the total effective capacitance profile $\alpha_{\text{tot}}(x)$ across ballistic graphene nanoconstrictions [Fig. 1(b)] with different degrees of edge disorder in a perpendicular magnetic field via magnetoconductance measurements. It is noteworthy that the local capacitance is accessible with these measurements in graphene devices [5,6,21], similar to conventional semiconductor structures defined in a 2D electron

gas [5,22]. Indeed, magnetoconductance measurements are particularly relevant for the characterization of narrow (≤ 150 nm) confined channels [22], where alternative magnetocapacitance techniques are limited due to several reasons. For instance, apart from possessing spatial resolution comparable to or larger than the device size, scanning gate microscopy [23], single-electron transistor [24], or microwave impedance spectroscopy [14,25] measurements might affect or be affected by the actual device electrostatics. Specifically, by analyzing quantum Hall (QH) transport measurements [5,6] we show that devices with stronger edge disorder exhibit a nearly homogeneous effective capacitance profile, with $\alpha_{\text{tot}}(x)$ similar to α_c^∞ . In striking contrast and in quantitative agreement with theoretical predictions for disorder-free systems, devices with a lower degree of edge disorder (< 1 nm roughness) show an inhomogeneous effective capacitance consisting of both classical $\alpha_c(x)$ and quantum $\alpha_q(x)$ contributions. The presence of quantum capacitance effects in graphene devices is a direct consequence of the low density of states (DOS) of the system in a perpendicular magnetic field [10]. Also, $\alpha_q(x)$ is additionally influenced by band-structure modifications occurring in disorder-free, gated graphene devices—systems dominated by the electrostatic screening of the gate potential [4,10].

As shown below, the large $\alpha_c(x)$ diverging toward the edge is comparable in magnitude to or larger than $\alpha_q(x)$ at low carrier densities and certain positions x across the channel of these clean devices, so that $\alpha_q(x)$ dominates the total effective capacitance [Eq. (1)].

II. SAMPLE FABRICATION AND MAGNETOTRANSPORT MEASUREMENTS

We have fabricated graphene nanoconstrictions [Fig. 1(b)] with similar lengths L and widths W , $L = W \sim 100$ nm on hydrophobized SiO_2 substrates with thicknesses $b = 100$ nm and with different degrees of edge disorder [4]. Our initial graphene is exfoliated on hydrophobic SiO_2 , resulting in flakes with mobilities ~ 20000 cm^2/Vs [4,26] and mean free paths l larger than L, W at $T = 4$ K ($l \sim 200$ nm [4,27]). Therefore, the transport of carriers through these devices is limited by boundary scattering [4]. Furthermore, we note that the edge roughness in these samples can be precisely assessed via transmission electron microscopy [4], favoring their use in the present work with respect to nanoconstrictions [3] made from encapsulated graphene [28–30].

In particular, we study two types of nanoconstrictions, referred to as type S_{low} and type S_{high} made in an identical way except for the final etching step [4,31]. Sample type S_{low} was etched using reactive ion etching (40 sccm argon, 5 sccm oxygen), a procedure producing significantly less edge disorder than oxygen plasma ashing [4,31] technique used in sample type S_{high} . Further fabrication details of these devices can be found in [27] and in [4], where similar systems were studied. Measurements of differential magnetoconductance were performed at different magnetic fields using a Stanford SR830 lock-in amplifier with an excitation voltage of 100 μV at a frequency of 17.77 Hz in a cryostat at $T = 300$ and 4 K.

Figures 1(c) and 1(d) show the magnetoconductance G as a function of V_g for the two sample types at different perpendicular magnetic fields B . The effect of edge disorder reflects itself in the magnitude of the conductance $G(V_g)$, which is more than two times smaller in type S_{high} [Fig. 1(c)] compared to type S_{low} [Fig. 1(d)] at any B and V_g . Furthermore, sample type S_{high} shows a quantized conductance $G = G(V_g)$ at high B for the zeroth-order Landau level, LL0, at the corresponding filling factor for graphene: $\nu = 2$. This plateau is followed by a conductance dip due to the presence of a nonzero longitudinal conductivity in the two-terminal device, effect that depends on the device geometry [4,32]. For the higher LLs, G is smaller than the expected quantized value due to the pronounced effect of edge disorder [4], promoting backscattering between edge channels [33]. In contrast, as reported in our previous study [4], sample type S_{low} (edge roughness < 1 nm) shows a conductance the value of which is larger than the value expected for the single-electron picture and does not exhibit quantization. This effect, referred to as conductance quantization suppression (CQS) [4,34], is a manifestation of additional, overlapping, and counterpropagating conducting channels in the constriction. These channels emerge from a qualitative modification of the band structure of the system in the quantum Hall

regime due to fringe field effects occurring in ballistic systems with low edge disorder [4,34] (see [27]). As such, their appearance is symptomatic of and relies on the presence of a large and inhomogeneous capacitance profile across the nanoconstrictions [4,34].

III. EFFECTIVE CAPACITANCE PROFILE

We experimentally probe $\alpha_{\text{tot}}(x)$ in both types of samples S_{low} and S_{high} by using the evolution of the magnetotransport data shown in Figs. 1(c) and 1(d) with respect to different B [the selected positions of $G(V_g, B)$ are marked by arrows]. This is possible thanks to the extreme sensitivity of the QH transport to the carrier density and carrier density distributions [5,6]. Briefly [5,6], in the QH regime, when the Fermi level lies in-between two LLs and the conduction is governed by ν propagating edge states, each edge channel probes a different spatial region from the edge of the device [5], whereas the bulk is insulating [5,6]. On the one hand, the carrier density n_ν for a filling factor ν is given by $n_\nu = \nu e B h^{-1}$ with a corresponding effective capacitance $\alpha_\nu = n_\nu (V_g - V_{\text{CNP}})^{-1}$, where V_{CNP} is the charge neutrality point of the device. On the other hand, an estimate of the position of these edge channels is given by corresponding cyclotron diameter $d_\nu = (\nu h)^{1/2} (e \pi B)^{-1/2}$ for graphene [5,6]. As the capacitance increase is linked to the charge accumulation across the channel of finite-size graphene devices, edge states of different LLs probe different spatial regions of the device and are subjected to a different effective capacitance. Thus, by making the assignment $\alpha_\nu \equiv \alpha(x = d_\nu)$, the profile of $\alpha_{\text{tot}}(x)$ across the device can be experimentally assessed [5,6]. Such capacitance mapping is clear in the absence of backscattering characteristic of the QH regime.

We argue that this semiclassical method can be also used in our ballistic devices exhibiting the CQS effect [sample type S_{low} , Fig. 1(d)]. This is particularly valid at gate voltages where the (nonquantized) conductance shows a maximum [arrows in Fig. 1(d)] $G^{(\text{max})}$, places where, if existent, backscattering between the counterpropagating edge channels in the system is weakest [4,34] (see [27]). As such, the link between G and $n(x)$ is still valid by taking $\nu(x) \sim G^{(\text{max})}(e^2/h)$, and the capacitance profile $\alpha_{\text{tot}}(x)$ can then be extracted as demonstrated below when comparing the experimental values with the corresponding theoretical calculations.

Figure 2 shows the extracted effective capacitance α_ν versus d_ν plotted for the different LLs in our samples with high and low edge disorder. Samples with high edge disorder [sample type S_{high} , Fig. 2(a)] show a slowly monotonically increasing capacitance $\alpha_\nu(d_\nu)$ towards edges, with quantitative values close to those of the infinite parallel-plate capacitor, α_c^∞ (green dotted line). Specifically, this capacitance profile is nearly homogeneous across the device channel (x direction), with only a small measured modulation up to $\alpha_{\text{tot}}(x)^{(\text{max})}/\alpha_c^\infty \sim 1.2$ for LL0. In contrast, devices with low edge disorder [sample type S_{low} , Fig. 2(b),] exhibit an inhomogeneous capacitance profile, with much larger modulations $[\alpha_{\text{tot}}(x)^{(\text{max})}/\alpha_c^\infty \sim 2.5$ for the case of LL0 and ~ 1.7 for LL1]. Moreover, we note how the capacitance profile is nonmonotonic in these samples for LL0.

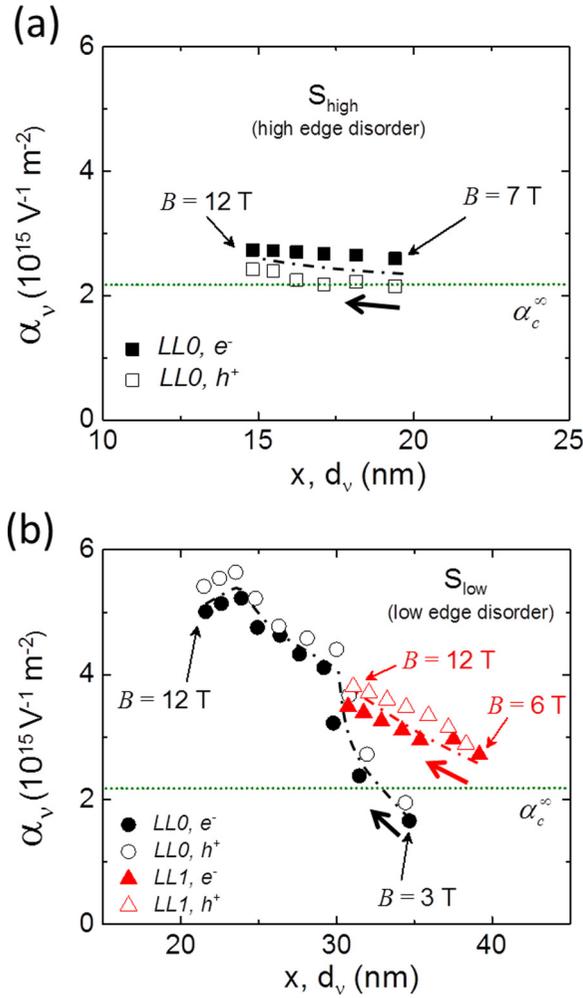


FIG. 2. Experimental effective capacitance α_v across the nanoconstrictions for different Landau levels (LLs) for electrons (e^-) and holes (h^+) at positions x corresponding to the cyclotron diameter d_v . (a) Sample type S_{high} . B varies from 7 to 12 T increasing in 1-T steps. $B = 7$ T is the minimum B to observe a conductance plateau for LL0 in samples S_{high} . (b) Sample type S_{low} . B varies from 3 to 12 T (LL0) and from $B = 6$ to 12 T (LL1) in 1-T steps. $B = 3$ and 6 T are the minimum fields to observe the CQS regime in these samples for LL0 and LL1, respectively. Green dotted lines show the capacitance value for an infinite parallel-plate capacitor $\alpha_c^\infty = \frac{C_{\text{ox}}}{e} = 2.19 \times 10^{15} \text{ V}^{-1} \text{ m}^{-2}$. Dash-dotted lines represent a guide to the eye.

Three initial conclusions can be drawn from the experimental data shown in Fig. 2, all of them consistent with a more pronounced capacitance profile for sample type S_{low} as compared to S_{high} .

(i) As expected from an electrostatic point of view [6,35], there exists electron-hole ($e^- - h^+$) symmetry in both type of samples. The difference in capacitances observed for e^- and h^+ is lower than 10% with respect to the absolute capacitance at any B .

(ii) The lowest LL (LL0) is closest to the edge, where more charge is expected to accumulate [4–8]. Consequently, for a constant B , α_v exhibits a higher absolute value for LL0 than for LL1 in sample type S_{low} .

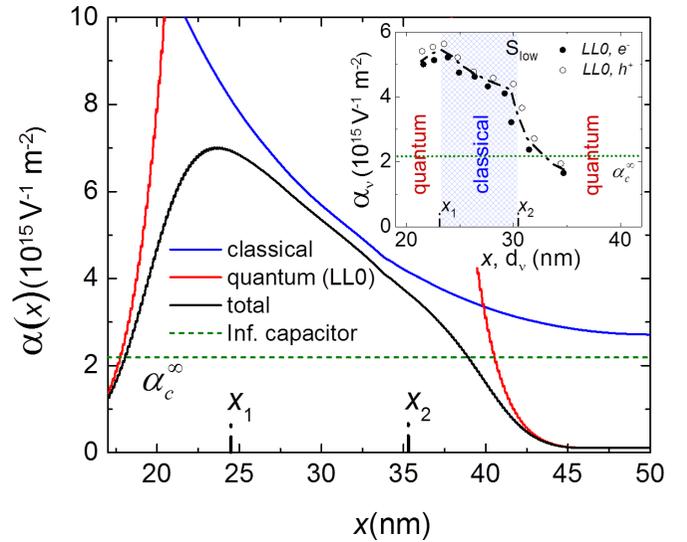


FIG. 3. Capacitance profile across the nanoconstrictions of type S_{low} for LL0. Calculated classical (blue curve), quantum (red curve), and total effective capacitance (black curve) profiles across a graphene nanoconstriction with $W = 100$ nm. The constant dashed green line represents α_c^∞ . The inset shows the experimental data for sample type S_{low} [Fig. 2(b)], separating zones where classical (blue) or quantum (red) contributions dominate according to the prediction shown in the main figure. Dash-dotted lines represent a guide to the eye.

(iii) For a varying B , the capacitance variation of LL0 is the largest as well.

Next, we analyze in more detail the capacitance profile $\alpha_v(d_v)$ in both types of devices. For LL0, the small modulation $\alpha_{\text{tot}}(x)^{(\text{max})}/\alpha_c^\infty \sim 1.2$ in devices of sample type S_{high} differs quantitatively from the electrostatic capacitance $\alpha_c(x)$ calculated for our constriction geometry, which is monotonic and diverges rapidly towards edges (Fig. 3, blue line, simulation details in [27]). Such behavior is attributed to scattering from edge defects, decreasing the charge accumulation in a similar manner as seen for low-quality devices with disorder in the channel [5,6]. This result explains the fact that some high-quality devices in literature exhibit effective capacitances resembling the one from an infinite parallel-plate capacitor α_c^∞ [3,4].

Then, we note that despite having a large modulation, the capacitance variation in sample type S_{low} does not completely follow the trend of the calculated classical electrostatic capacitance $\alpha_c(x)$ either: rather than following a divergent $1/\sqrt{x}$ dependence towards the edge [7,8] the measured capacitance α_v has a nonmonotonic profile, decreasing more rapidly than $\alpha_c(x)$ for distances close to the edge and in the middle of the channel. Moreover, α_v exhibits some values smaller than α_c^∞ at the central part of the constriction (distances > 35 nm from the edge) for LL0 but not for LL1. In addition, α_v shows a maximum $\alpha_v^{(\text{max})}$ at a position $d_{v,\text{max}} \sim 24$ nm from the edge, with α_v smaller than $\alpha_v^{(\text{max})}$ for $d_v < d_{v,\text{max}}$. We do not expect this extremal value to be caused by the breakdown of the classical capacitance $\alpha_c(x)$ at distances from edges given by the magnetic length $l_B = \hbar^{1/2}(eB)^{-1/2}$ [7]: $d_{v,\text{max}}$ is three times larger than l_B at the corresponding B (10 T)

at which the maximum is observed. This argument is additionally supported by the fact that no local maximum is experimentally observed in sample type S_{high} [Fig. 2(a)] despite the minimum measured d_v in such samples (15 nm) being smaller than $d_{v,\text{max}}$.

IV. CAPACITANCE CALCULATION: CLASSICAL AND QUANTUM CONTRIBUTIONS

We demonstrate that the mismatch between the calculated electrostatic capacitance (Fig. 3, blue line) and the experimentally probed capacitance of sample type S_{low} at the LL0 [Fig. 2(b), black circular dots and Fig. 3, inset] can be fully accounted for by including quantum contributions to the total capacitance of the system. Such quantum contributions, arising due to the low local DOS [36–39] across the quasi-one-dimensional system [10], are not only dependent on the presence of external magnetic fields B but also heavily affected by the electrostatic screening of the gate potential [4,34] (see [27]).

We calculate the quantum capacitance contribution [27] $\alpha_q(x)$ across the ballistic, gated devices at $B = 10$ T and low carrier densities (LL0) (Fig. 3, red line). First, we show that both classical and quantum contributions are comparable in magnitude and coincide at two distances: ~ 25 nm far from the edge and at the central part of the nanodevice (~ 40 nm far from the edge). By using Eq. (1), we calculate the total effective capacitance $\alpha_{\text{tot}}(x)$ of these devices (Fig. 3, black curve), which is in quantitative agreement (below $\sim 30\%$ mismatch) with our experimental data $\alpha_v(d_v)$ for LL0 [Fig. 2(b) and Fig. 3, inset]. Specifically, $\alpha_{\text{tot}}(x)$ shows a clear maximum $\alpha_{\text{tot}}^{(\text{max})}(x_1)$ at a position $x_1 \sim 25$ nm, close to $d_{v,\text{max}}$. For closer distances ($x < x_1$) towards the edge at $x = 0$, $\alpha_{\text{tot}}(x)$ is dominated by quantum contributions since $\alpha_c(x) > \alpha_q(x)$. Instead, for distances $x_1 < x < x_2$ with $x_2 \sim 32$ nm, $\alpha_{\text{tot}}(x)$ is mostly dominated by the classical capacitance profile, since $\alpha_c(x) < \alpha_q(x)$ in this interval. Furthermore, for distances $x > x_2$ (central part of the nanoconstriction) $\alpha_{\text{tot}}(x)$ is again dominated by the quantum contribution due to the vanishing DOS at these positions. This argument explains the fact that, experimentally, α_v for LL0 is smaller than α_c^∞ at the central part of the constriction, too. Finally, for completeness, we note that the experimental capacitance profile α_v shown for LL1 in samples S_{low} [Fig. 2(b), red triangles] increases monotonically towards edges, similar to

the classical profile. This behavior is due to the fact that the quantum contribution to the capacitance for LL1 is larger than the classical one [27] for the probed distances from the edge $x \equiv d_v$ (between 30 and 40 nm). Thus, the classical contribution dominates the total effective capacitance profile at these higher carrier densities (LL1).

V. CONCLUSIONS

In summary, by analyzing quantum Hall transport measurements, we have extracted the total effective capacitance profile across ballistic graphene devices. Such profiles heavily depend on the edge disorder level in the device. In particular, similar to diffusive devices, ballistic samples with large edge roughness show a nearly homogeneous capacitance with small modulation and values close to the infinite parallel-plate capacitor. In contrast, in excellent agreement with calculations for pristine and gated systems, narrow ballistic devices with low edge disorder (< 1 nm roughness) show an effective capacitance profile with a much larger modulation due to an enhanced impact of the fringe field effects at graphene edges. This profile is nonmonotonic at low carrier densities due to the interplay between classical and quantum capacitance contributions across the gated device even when using relatively thick (100 nm) dielectric layers [8,9]. Despite being demonstrated in ballistic graphene nanoconstrictions, our conclusions can be extended to other systems with sharp edge potential [4,7,8,34], and, thus, our findings can help to understand the electronic properties of other types of gated nanostructures made from 2D materials [40,41] or other 2D systems [42].

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