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Quenching of the Quantum Hall Effect in Graphene with Scrolled Edges

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Edge nanoscrolls are shown to strongly influence transport properties of suspended graphene in the quantum Hall regime. The relatively long arclength of the scrolls in combination with their compact transverse size results in formation of many nonchiral transport channels in the scrolls. They short circuit the bulk current paths and inhibit the observation of the quantized two-terminal resistance. Unlike competing theoretical proposals, this mechanism of disrupting the Hall quantization in suspended graphene is not caused by ill-chosen placement of the contacts, singular elastic strains, or a small sample size.

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Graphene is a material that combines a highly tunable metallic conduction [1,2] with a flexibility of a two-dimensional (2D) membrane [3,4]. The hallmark of a 2D metal is the quantum Hall effect (QHE), which can be observed in graphene on SiO₂ substrates at room temperature. A more fragile fractional QHE is robust in graphene on boron nitride (BN) substrates [5]. Yet the demonstration [6,7] of the QHE in graphene suspended off a substrate has been difficult in most suspended samples, where the quantization is usually very poor [2,3,7,8]. Here, we propose a general explanation for this surprising result.

Being a topological property, the quantization of the Hall resistance ρ_{xy} can normally be destroyed only if external perturbations exceed the Landau gap. If so, the system can split into regions with different local ρ_{xy} while the observed Hall resistance can have some average non-quantized value. However, suspended graphene is believed to have very low disorder [9–13]. Another possible reason for deviation from the ideal QHE is backscattering of the chiral edge channels across the sample due to impurities [14] or elastic strains [15]. Thanks to the improvement of the fabrication methods, samples can reach a width of several microns, for which these sources of backscattering can be ruled out. Also the issue of *hot spots* [14], important for small samples in the four-terminal configuration, is becoming progressively irrelevant for the same reason. The only other known mechanism of disrupting the QHE is an edge reconstruction (ER), which can generate counterpropagating channels at the same edge. However, previously discussed ERs [16–19] are not unique to suspended graphene. They originate from a generic tendency of a 2D metal to have a nonuniform density near the edge.

In this Letter, we examine a novel type of ER—edge nanoscrolls—that is specific and indeed common to

freestanding and suspended graphene [9,20–22]. As shown hereafter, edge nanoscrolls are at the origin of the poor quantization in both two-terminal measurements, beyond geometry effects [23], and four-terminal measurements, where scrolls are even more likely to occur. Our result does not rule out the possible contribution of other back-scattering sources in specific samples nor entails that QHE is not observable in suspended samples. It just provides a simple relation between two very common experimental observations [8]. Our basic idea is that the magnetic field $B = 2\text{--}20$ T in typical experiments has only a weak effect on the edge scrolls (if the Fermi energy is large enough). In this respect, the scrolls are similar to multiwall carbon nanotubes (CNT) where the QHE is seen only at very high fields [24]. The reason is that electrons respond primarily to the component B_{\perp} of the total field along the local normal to the graphene sheet. Inside the scrolls, which look like spirals in cross section (Fig. 1), B_{\perp} oscillates in sign and largely averages out. In contrast, the flat portion of the sample is expected to be in the QHE regime and thus has a low dissipative conductivity. The current injected from the contacts may prefer to avoid the flat

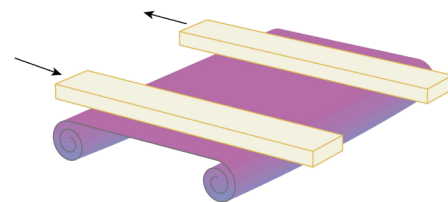


FIG. 1 (color online). Sketch of a system under study in which a suspended graphene ribbon (purple) has scrolled edges. The ribbon is attached to the source and drain contacts (gold). The arrows symbolize the external current.

region and instead diffuse into the more conducting scrolls. The lack of ρ_{xy} quantization therein would then significantly influence the net two-terminal resistance of the device.

Below we support this qualitative picture by several calculations, which are carried out at the progressively more microscopic level. The possible presence of strain in the regions of the scrolls is not taken into account. Actually, strain might be absent and in fact graphene can scroll just to release strain. Scrolls could be also strained, depending on the specific graphene device geometry, but this additional complication will induce nonuniform supplementary pseudomagnetic fields [15,25], of which impact on the QHE should be averaged out. We start with continuum models suitable for large scrolls. Subsequently, we consider a few nm-wide scrolls using the discrete lattice representation.

By large scrolls, we mean those with outer radius $r_o \sim 10$ nm. Such scrolls may contain many turns and their total arclength can measure in hundreds or thousands of nm [21,22,26,27]. Adjacent graphene layers of the scrolls may have commensurate [28,29] or incommensurate lattice structure. In the latter, more typical, case the scrolls are held together by van der Waals attraction [30,31] and the separation between the adjacent layers should be about the same as in graphite, 0.34 nm. Assuming that incommensurability makes interlayer electron tunneling negligible, we can switch from the three-dimensional (3D) Fig. 1 to the equivalent 2D Fig. 2 (inset) of the unwrapped sheet. Discarding the part directly attached (and thus electrically shunted) by the contacts, we obtain the I-shaped silhouette of the unwrapped sample. (Existence of the scrolls underneath the contacts is suggested by published micrographs of suspended graphene [32]). This is the system we want to study the transport through.

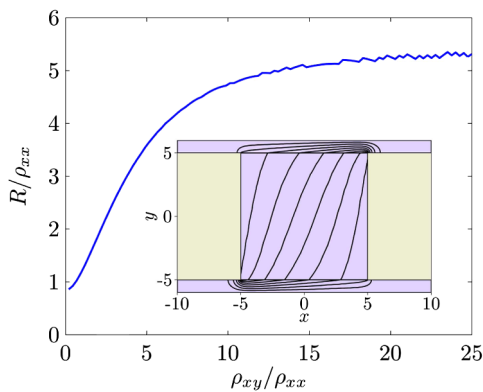


FIG. 2 (color online). Inset: Lines of current in the graphene ribbon (purple) in the semiclassical approximation. For clarity, the edge scrolls are unwrapped and only the parts of the electrodes in contact with the ribbon are shown (gold). Main panel: Two-terminal resistance R as a function of ρ_{xy} , both normalized to the constant diagonal resistivity ρ_{xx} .

We begin with a simple semiclassical approximation in which the transport in graphene is described by local diagonal $\rho_{xx}(x, y)$ and Hall $\rho_{xy}(x, y)$ resistivities. Assuming that magnetic field has no effect on electron states inside the scrolls, we set $\rho_{xy}(x, y)$ to zero in the scrolls and to a nonzero constant in the flat part of the sample. We take ρ_{xx} to be coordinate independent. The current distribution in this model can be easily computed numerically. The lines of the current for some representative parameters are shown in Fig. 2 (inset). The chosen arclength of each scroll ℓ is a small fraction of the width $L = 10\ell$ of the flat part (the central square). Yet a significant portion of the current flows inside the scrolls.

The results for the two-terminal resistance R are plotted in the main panel of Fig. 2. They imply that our system acts as three resistors in parallel. One resistor represents the flat region. In the QHE regime, where $\rho_{xy} \gg \rho_{xx}$, its magnitude is approximately equal to $R_f = \rho_{xy}$. The other two resistors, of magnitude $R_s = \rho_{xx}L/\ell$ each, represent the scrolls. If $1 \ll \rho_{xy}/\rho_{xx} \ll L/\ell$, which corresponds to weak or modestly strong magnetic fields, the scrolls are not important, so that we have $R \approx R_f \approx \rho_{xy}$. At larger ρ_{xy}/ρ_{xx} , the scrolls short circuit the transport, so that $R \approx R_s$, which is ρ_{xy} independent in this model.

One can extend the same approach to other situations, including scrolls clamped and tapered near the contacts [9,31]. In general, R_s would be augmented by a model-dependent contact resistance, which would suppress the portion of the current going into the scrolls.

Let us now address whether it is justifiable to neglect the effect of the magnetic field on the scrolls. This issue is separate from the role of disorder and contacts. Hence, we can assume for the time that the system is disorder-free and infinite in the x -direction. The electron energies $E(k_x)$ can then be labeled by the conserved momentum k_x . The low-energy part of the spectrum is easily computed by diagonalizing the effective Dirac Hamiltonian (suitable for zigzag edges)

$$\mathcal{H} = \hbar v[\sigma_x(k_x - l_B^{-2}Y) - i\sigma_y\partial_y], \quad (1)$$

where v is the Fermi velocity, Y is the y -coordinate in the original 3D space, $l_B = \sqrt{\hbar c/eB}$ is the magnetic length, σ_ν are the Pauli matrices [3]. In Fig. 3 we show a representative spectrum for the scroll of outer radius $r_o = 11.1$ nm and arclength $\ell = 100$ nm connected to the flat region of the same width. The discrete energy plateaus come from the flat region of the sample. The rest of the spectrum very much resembles that of a carbon nanotube (CNT) [33] except the dispersion curves appear in bunches. The literature on the Landau problem for CNT, e.g., [24,33–35] and other curved 2D systems [36] readily furnishes an interpretation of these spectral features. The spectrum consists of four regions demarcated approximately along the lines $E = \hbar v(k_x \pm l_B^{-2}r_o)$. These regions correspond to

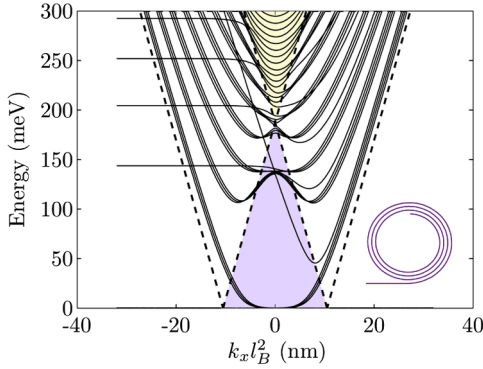


FIG. 3 (color online). Band structure of a scroll with the inner and outer radii, respectively, $r_i = 9.91$ and $r_o = 11.1$ nm at $B = 20$ T computed from the Dirac equation with the zigzag edge boundary condition. The dashed lines are defined by equations $E = \hbar v(k_x \pm l_B^{-2} r_o)$. The cross section of the system is sketched in the bottom right corner of the figure. For clarity, the flat region (full width 100 nm) is truncated and the layer separation inside the scroll is enlarged.

quasiclassical trajectories of four different types. The bottom region (shaded purple in Fig. 3) can be viewed as the Landau levels of electrons confined near the top of the bottom of the scroll and experiencing the local magnetic field $B_{\perp}(Y = k_x l_B^2)$. The left and the right regions correspond the snakelike states propagating along the eponymous sides of the scroll. The number of the lines in each dispersion bunch (either three or four in Fig. 3) represents the number of layers in the given region of the scroll (top, bottom, left, or right). The energy spacing within these bunches is due to the small difference in the layer radii. If we were to include effects of electron interaction, self-consistent screening of external electric fields, and/or disorder, the near-degeneracy of the bunches would be considerably lifted.

Our crude approximation of neglecting the B field is reasonably accurate inside the top region (shaded gold) of traversing [37], i.e., spiraling trajectories. Indeed, at $E \gg \hbar v l_B^{-2} r_o$ the cyclotron radius $R = cE/(evB_{\perp})$ greatly exceeds r_o , so that deflection of electrons by the Lorentz force is negligible. The spectrum of the top region is essentially the same as that of a graphene ribbon of width $\ell + \mathcal{O}(l_B)$ at $B = 0$. As r_o decreases, the region of traversing trajectories becomes progressively more dominant. Eventually, at $r_o < l_B$, neglecting the effect of B inside the scrolls is fully justified. In the remainder of the Letter we focus on exactly this case.

Dirac Hamiltonian (1) may no longer be accurate for very narrow scrolls (note that at, say, $B = 20$ T we have $l_B = 5.7$ nm). Therefore, we replace it by the tight-binding model $\mathcal{H} = \sum_j \epsilon_j |j\rangle\langle j| - \gamma_0 \sum_{\langle j,k \rangle} e^{-i\varphi_{jk}} |j\rangle\langle k|$ defined on a honeycomb lattice of sites $|j\rangle$. Here $\gamma_0 = 2.7$ eV is the coupling between nearest neighbors and φ_{jk} are the Peierls phases proportional to the magnetic flux through each hexagon of the lattice. This flux is constant in the flat

region but oscillates from positive to negative within each turn of the scroll.

For simplicity and numerical efficiency, we work with the rectangular rather than I -shaped sample. To be precise, we consider an armchair nanoribbon composed of 814 dimer lines, 814-aNR. Its width of 100 nm is significantly smaller than that of suspended graphene flakes studied experimentally. However, this does not qualitatively change any of our main conclusions.

We start with the disorder-free case where all the on-site energies ϵ_j are zero. Without the scrolls, we find the familiar sequence of Landau levels in the bulk (low k) and the dispersive channels at the edges (high k) at $B = 20$ T. Except for the narrow energy interval near the bulk levels, the edge channels are chiral. They have the same sign of velocity dE/dk , see Fig. 4(a). (The doublet structure of the edge states and the narrow region of non-monotonic dispersion is a nongeneric peculiarity of the armchair edge [3,38].)

The edge channels undergo a qualitative change when the $\ell = 15$ nm strips at both edges are wrapped into scrolls of two full turns. Both signs of velocity are now present at all E , see Fig. 4(b). The energy separation of the edge

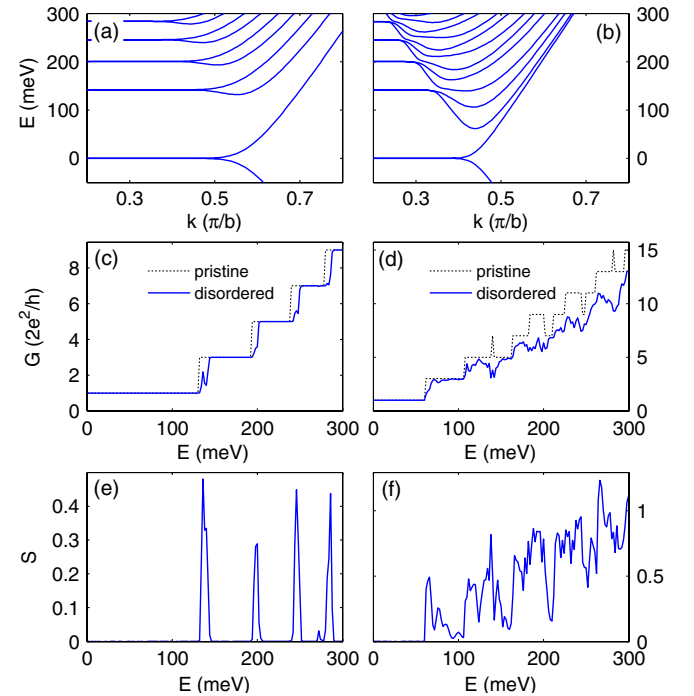


FIG. 4 (color online). (a) Band structure of a flat 814-aNR at $B = 20$ T. (b) The same as (a) in the presence of nanoscrolls of arclength $\ell = 15$ nm and two full turns. (c) Conductance as a function of the electron energy for pristine (black dashed lines) and disordered (blue solid lines) 814-aNRs at $B = 20$ T in the absence of scrolls. (d) The same as (c) in the presence of the scrolls. (e) and (f) Shot noise for a disordered system described in, respectively, (c) and (d).

states is about the same as in the $30 \text{ nm} \approx \ell + 3l_B$ wide 241-aNR in zero field. Evidently, it has little to do with the bulk Landau level gaps.

Next we compute differential conductance $G(E) = 1/R = (2e^2/h)\sum_m T_m$, where T_m are the transmission coefficients for the transmission channels at a given Fermi energy E [39]. Only occasionally these channels are of mixed origin. At most E , they can be attributed to either the flat or the scrolled regions. Therefore, the earlier formula for resistors connected in parallel, $G = R_f^{-1} + R_s^{-1}$, still applies. However, the magnitude of these resistors is now computed from the quantum expressions.

Since the energy spacing in the scrolled regions in Fig. 4(b) is considerably smaller than the Landau level spacing in the bulk, we deal with the case $R_s \ll R_f$ where the scrolls essentially dominate the transport. This is similar to the large ρ_{xy}/ρ_{xx} limit in Fig. 2 where the total resistance is B -independent. Indeed, let us compare the dashed traces in Figs. 4(c) and 4(d). Both are perfectly quantized. However, their average slopes differ almost twice. While Fig. 4(c) reflects the quantization of ρ_{xy} , Fig. 4(d) basically describes the subband quantization of the 241-aNR in zero field, with not much relation to the actual B . Some interplay between the two types of quantization is still present. It leads to, e.g., intriguing non-monotonic behavior of $G(E)$ in Fig. 4(d), noted previously for other scrolled 2D systems [40].

To make the model more realistic we introduce on-site disorder in a ribbon section of length 210 nm. It includes a short-range disorder generated by randomly varying the on-site energies ϵ_j within the range $[-25, 25]$ meV. We also add a finite-range disorder modeled by a sum of 50 Gaussians with range $\xi = 1$ nm centered at random positions and having random strengths in the range $[-500, 500]$ meV. The disorder causes scattering among counterpropagating channels, which is conveniently quantified by the shot noise $S = \sum_m T_m(1 - T_m)$.

When disorder is present, the scroll-free structure exhibits a robust quantized conductance and zero shot noise, except for energies nearby each onset of new subbands, see the solid line in Fig. 4(c). This situation is typical of the quantum Hall effect. (Some residual backscattering is due to the relatively small width of the system and also due to the aforementioned aNR-specific nonchiral edge states near the bulk Landau energies.) For the ribbon with scrolled edges, Fig. 4(d), the conductance quantization is considerably degraded with the exception of region corresponding to the first conductance plateau. The enhanced shot noise, Fig. 4(f), also indicates a stronger backscattering. This behavior is typical of narrow disordered ribbons in which spatial overlap of conduction channels causes significant backscattering even in the presence of a moderate disorder.

The backscattering of the counterpropagating channels would eventually lead to localization of the nonchiral

modes in the scrolls, thus recovering the quantum Hall effect in long ribbons. However, the localization length should scale with the number of channels, i.e., the arc-length ℓ of the scroll. Therefore, for large enough ℓ , the localization may not be very important. This can be contrasted with the case of multiwall CNT where the current is confined to the outer shell [24], which reduces the number of channels.

In conclusion, the edge scrolls in suspended graphene have been shown to short circuit the source-drain current paths in the quantum Hall regime inhibiting the observation of the Hall conductance quantization in such systems.

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