Unraveling Quantum Hall Breakdown in Bilayer Graphene with Scanning Gate Microscopy


ABSTRACT: Investigating the structure of quantized plateaus in the Hall conductance of graphene is a powerful way of probing its crystalline and electronic structure and will also help to establish whether graphene can be used as a robust standard of resistance for quantum metrology. We use low-temperature scanning gate microscopy to image the interplateau breakdown of the quantum Hall effect in an exfoliated bilayer graphene flake. Scanning gate images captured during breakdown exhibit intricate patterns where the conductance is strongly affected by the presence of the scanning probe tip. The maximum density and intensity of the tip-induced conductance perturbations occur at half-integer filling factors, midway between consecutive quantum Hall plateaus, while the intensity of individual sites shows a strong dependence on tip-voltage. Our results are well-described by a model based on quantum percolation which relates the points of high responsivity to tip-induced scattering in a network of saddle points separating localized states.

KEYWORDS: Graphene, scanning gate microscopy, quantum Hall effect, resistance metrology, quantum percolation

The presence of nanometer-sized potential fluctuations has a profound impact on the critical exponents and thresholds for electronic percolation in quantum nanodevices. Close to the classical threshold for percolation, for instance, quantum mechanical tunnelling enhances classically forbidden connections between nodes, while interference and scattering can suppress the formation of extended states. Quantum percolation is thus key to understanding the conducting properties of a wide range of two-dimensional systems, such as perovskite manganite films, dilute weakly disordered silicon channels, and chemically functionalized sheets of graphene.

Quantum percolation theory has been used to model the quantum Hall effect (QHE), or the appearance of quantized plateaus in the Hall conductance of a two-dimensional electron system (2DES), whenever the Fermi level is in the energy gap between two Landau levels (LL). Within the framework of quantum percolation theory, plateaus occur because electrons in the bulk follow closed and thus localized paths, while current-carrying extended states run along the free edges where they are protected from backscattering and dissipation. The QHE breaks down in between plateaus because electrons percolate through a network of bulk states, leading to backscattering between edge states and a nonintegral contribution to the Hall voltage. Several experiments reporting on the regularity of fluctuations in the two-terminal conductance and electronic compressibility suggest that many-body effects must also be considered for a complete explanation of transport in this regime. Electron–electron interactions introduce nonlinear screening effects due to the lack of available electron states at the Landau band edges, leading to the formation of localized compressible islands residing in an incompressible background. Transport via these states is only possible when the local potential is sufficient to overcome the Coulomb blockade, and the experimentally detected fluctuations are attributed to the periodic charging and discharging of these islands. As the linear screening regime is approached, the compressible regions delocalize, and a metallic–insulator transition occurs.

Interest in the QHE has been reignited recently by the discovery of the anomalous integer and fractional QHE in graphene. The plateau structure of single- and few-layer graphene, for instance, has revealed that valley, spin, and sublattice degeneracies can be broken by changing the morphology and topography of the crystal lattice, the length scale and strength of the potential disorder landscape, and the type of layer stacking order. Since quantum percolation...
and interaction effects govern the resolution, length, and quantization accuracy of the plateaus, understanding their role in graphene will be key to implementing graphene as a metrological standard of resistance that can be operated at lower magnetic fields and higher temperatures. \(^{14-17}\)

In this Letter we use scanning gate microscopy (SGM) to unravel the paths of electrons during QHE breakdown of a graphene bilayer and show that transport is well-described by quantum percolation between localized states. Although the nature of quantum Hall localization in graphene has enjoyed much attention recently,\(^{8,18-21}\) the topological origin of the QHE breakdown has previously only been examined using SGM in GaAs subsurface 2DESs.\(^{22-25}\) Equilibration between the bulk and edge states, for instance, was shown to occur at specific points in GaAs Hall bars\(^{22,26}\) and in the constrictions of InGaAs/InAlAs quantum rings.\(^{27}\) In similar devices, such tipsensitive regions were also observed away from the edges, and their pattern was found to repeat at the same value of half-filling of each Landau level.\(^{24,25}\) Both SGM results and those obtained by less invasive techniques such as scanning force microscopy\(^{28}\) and scanning tunnelling microscopy\(^ {29}\) were well-described within a single-particle framework.\(^{30}\) While the images we obtain also display features which are reminiscent of structures observed in other 2DESs via SGM, the higher spatial resolution and presence of pronounced potential fluctuations of SiO\(_2\) supported graphene lead to intricate patterns not observed in previous studies.

We investigate a graphene flake (dimensions \(\approx 2.5 \times 6 \, \mu m^2\)) mechanically exfoliated from natural graphite onto a highly doped Si substrate capped with a 300 nm thick SiO\(_2\) layer. The flake was identified as a bilayer from its optical contrast,\(^ {31}\) and two (5 nm/30 nm) thick Ti/Au contacts were patterned using e-beam lithography, thermal evaporation, and standard PMMA lift-off processing. While bilayer graphene is not considered to be a prime candidate for quantum metrology, like monolayers it is susceptible to adsorbates, rippling, and potential fluctuations from the host substrate and should therefore reflect the features controlling QHE breakdown which are common to both. Figure 1b shows the numerical derivative of the two-terminal conductance of the device as a function of back-gate voltage \(V_{BG}\) and magnetic field \(B\) at a temperature \(T \approx 8 \, K\) (\(V_{SD} = 1 \, mV\)). As anticipated for two-terminal bilayer graphene devices, N-shaped conductance plateaus quantized in units of \(4e^2/h\) develop as a result of edge channel conduction and strong localization in the QH regime.\(^ {32,33}\)

To probe the QH state locally during breakdown, we tune the conductance of the device to a value between the first and the second quantized plateaus (\(V_{BG} = 17 \, V, B = 6.2 \, T\)), and image the device using SGM (see refs 25, 34-40 for more details). SGM involves scanning a sharp metallic tip over the surface of graphene while measuring its conductance, and a schematic of our SGM setup is shown in Figure 1a. A typical scanning gate micrograph is shown in Figure 1c. A striking feature of the image is a texture consisting of \(\approx 100 \, nm\) sized “hotspots” where the conductance is strongly modulated by the tip. Note that this fine structure appears against a broad background modulation, which probably stems from the long-range gating effect of the tip cone.\(^ {41}\) To examine just the fine pattern in more detail, the image in Figure 1c was flattened by subtracting a parabolic background with image analysis software.\(^ {42}\) To analyze features in the resulting image quantitatively, we calculate the two-dimensional autocorrelation function \(C(x,y)\) shown in the inset of Figure 1c. Owing to the roughly uniform density and size of the hotspots, \(C(x,y)\) exhibits oscillations with periodicity governed by the average hotspot spacing \((D)\) and a peak close to zero whose half-width reflects their size \((r)\).\(^ {43}\) Figure 1d shows a section of \(C(x,y)\) taken along the blue line in the inset of Figure 1c, allowing us to make estimates for \(r \approx 90 \, nm\) and \(D \approx 450 \, nm\).

As a first step toward understanding the origin of these hotspots and how they relate to the underlying electron trajectories, we employ numerical simulations based on the percolation of single-particle states near the center of disorder-broadened Landau bands. While our noninteracting model cannot account for the charge rearrangements in compressible dots, for the resulting Coulomb blockade effects, and in general, for the evolution of the potential due to screening as the filling factor is varied, it has been pointed out by some authors\(^ {35-48}\) that the predicted critical behavior at the transition between plateaus may exhibit no differences with respect to that obtained with self-consistent Hartree–Fock calculations. In particular, the localization behavior of the wave functions has been found to be similar to that characteristic of a single-particle model. In this paper we therefore restrict our analysis to the neighborhood of a single transition, attempting, as a simple approximation, a description of the Hall breakdown based on a semiclassical single-particle approach. We refer, in detail, to the phenomenological network model proposed by Chalker and Coddington.\(^ {5,44}\) This method has been successfully used to

---

**Figure 1.** (a) Circuit used to perform scanning gate microscopy. The edges of the flake are indicated by white dashed lines, and superimposed over the surface is a raw SGM image taken at a lift height of 50 nm. (b) Landau level fan diagram showing the numerical derivative of the conductance as a function of back-gate voltage and magnetic field at \(T \approx 8 \, K\). The source-drain bias voltage is \(V_{SD} = 1 \, mV\). The white (solid) line is a trace of the conductance along the dashed line at \(B = 6.2 \, T\), showing plateaus at filling factors of 4 and 8. (c) Flattened SGM image obtained with the tip at \(\approx 50 \, nm\) lift height. The white dashed lines indicate the edge of the flake. Inset: Autocorrelation function taken over the flake. (d) Line profile through the peak of the 2D autocorrelation function. Labeled length scales \(r\) and \(D\) correspond to the radius of the hotspots and to the separation between them, respectively.
study percolative transport in QHE and, incidentally, has also been shown to map, with the proper positions, onto the two-dimensional Dirac equation, governing electron dynamics in monolayer graphene. This model assumes that conduction through the flake around the LL can only occur along a path of connected localized states. Here we employ the conventional picture that electrons perform cyclotron orbits while drifting along equipotential contours in the electrostatic landscape, and it is these orbits which we refer to as localized states. The tip can thus only affect the conductance by perturbing the potential of the saddle points where two such states approach each other and tunneling becomes possible (Figure 2a). For simplicity, our network consists of a regular array of saddle points, as shown in Figure 2b. As in ref 44, the transmission of each node of the network is parametrized by means of a dimensionless quantity \( \theta \), which is a nondecreasing function of the difference between the energy of the incident electron and the potential of the saddle point. Disorder is therefore introduced by randomizing the values of \( \theta \) characteristic of each saddle point, and keeping the variance of the distribution \( p(\theta) \) as a free parameter provides a way to extract the size of the potential fluctuations in a real sample. Further disorder is included by randomizing, as in the model of Chalker and Coddington, the phase shifts associated with each link, to take into account the random relative positions of the saddle points.

Figure 2c shows the transmission of a sample network as a function of \( V_{BG} \) with the tip voltage held at zero, showing a gradual transition from an insulating to a fully transmitting condition, in good agreement with the conductance versus \( V_{BG} \) measured experimentally in Figure 1b. At low values of \( V_{BG} \), corresponding, on average, to low values of \( \theta \), the probability of tunneling between localized states is small due to the large separation between neighboring localized states residing in the bottom of potential valleys, and the transmission is thus suppressed. At high values of \( V_{BG} \) corresponding, on average, to large values of \( \theta \), transmission is perfect owing to the formation of completely conducting links along the edge of the network. At intermediate values of \( V_{BG} \), transmission increases due to conduction through a series of localized states tunnel coupled at saddle points. Representative maps of the current density during these stages are shown in Figure 2d. The network model thus provides an ideal framework for simulating SGM images: we scan the probe over each node, thereby perturbing the \( \theta \) values, and we plot the change in conductance \( \Delta G \) as a function of the position of the center of the perturbation induced by the probe. The amplitude of the perturbation at each saddle point is obtained assuming a Lorentzian-shaped coupling between the probe and the flake. We performed numerical simulations to determine the effect of a realistic tip geometry on a graphene bilayer sheet and found that the induced charge density perturbation is well-described by the sum of two Lorentzians. We use the narrower of the two, which has a half-width at half-maximum of \( \approx \)50 nm, to describe the local extent of the tip perturbation. A hotspot implicates a particular node group as part of a current path that makes a substantial contribution to the conduction of the whole network, mimicking the experimental situation precisely.

To test this picture for quantum Hall breakdown, we experimentally monitored the evolution of the hotspots with back-gate voltage at a fixed magnetic field of 6.2 T. Figure 3a shows a sequence of SGM images captured at different values of \( V_{BG} \) as the conductance increases on the riser between the first and the second quantized plateaus. Inspection of the images shows that on the plateaus themselves (images 1 and 5) the image texture is characterized by weak long-range fluctuations in \( \Delta G \), while on the riser it becomes more intricate and the intensity of individual hotspots increases (see Supporting Information for a movie of the transition). The hotspot intensity also appears most pronounced when the LL is half-filled (image 3). This behavior is concisely represented, and more detail is revealed in Figure 3b, which shows the evolution of the normalized autocorrelation function with back-gate voltage. The data are extracted from a set of 50 images taken between back-gate voltages of 10 and 20 V in 0.2 V increments. The width of the peak around zero corresponding to 100 nm-sized hotspots (c.f. Figure 1c) remains constant on the riser, diverging to around 1 \( \mu \)m at either end where the flake enters the QH regime. This trend is clearly depicted in Figure 3b, where we show several line-profiles at different back-gate voltages at the edge of the plateau. To determine whether these features of our data are peculiar to the transition between the \( \nu = 4 \) and \( \nu = 8 \) states (where \( \nu \) indicates the filling factor), we captured a similar set of images between the Dirac point and the \( \nu = 4 \) plateau. The result is summarized in Figure 3c, which shows a \( \Delta G(x) \) profile across several hotspots (see image 2, Figure 3a) as a function of back-gate voltage spanning both the first two risers. The hotspots appear and reach their peak intensity at the same position along both risers, confirming that...
the observed image sequence is robust and is controlled by the filling factor of the top LL relative to half-filling. Note that the average intensity of all of the hotspots is also reflected by the amplitude of the central autocorrelation peak, which also reaches a maximum at the middle of the riser (see Figure 3b).

The right panel of Figure 3a shows a representative sequence of simulated SGM images taken at the values of $V_{BG}$ marked in Figure 2c. Since the potential landscape for the simulations has been generated randomly, there cannot be a one-to-one correspondence with the experimental data (which are relative to a different random potential). The left and right panels of Figure 3a should therefore be compared from the point of view of the overall trend of increasing and decreasing hotspot intensity. Indeed, key experimental observations, such as the increasing intensity and intricacy of the texture toward the center of the LL, are well-captured by the simulations. In particular the inset of Figure 3b shows the simulated autocorrelation function, which displays the same divergence of hotspot size toward the ends of the risers, and the amplitudes of the conductance variations are also in good quantitative agreement. In our calculations we have considered different values for the mesh size, that is, the average separation $D$ between saddle points, from 50 to 100 nm, and we settled on the value of 60 nm, which appeared to yield the best agreement with the experimental results. The considered potential fluctuations are uniformly distributed around zero, as in ref 30 within an interval of amplitude $\approx 10$ meV. Both the spatial extent (60 nm) and the strength of the disorder which adequately reproduce the experimentally determined values for $r$ and $D$ are slightly larger than, though in reasonable agreement with, values existing in the literature.21,53 Thus our experimental observations are naturally explained within the proposed framework as arising from the increased likelihood of tip-enhanced tunneling due to the increased proximity of bulk states at the center of the LL, as illustrated in Figure 2d.

To obtain further insight into the properties of individual hotspots, Figure 4a illustrates a typical sequence of images captured with increasing tip voltage ($V_T$) at a fixed $V_{BG} = 7.4$ V, midway along the riser between the Dirac point and the $\nu = 4$ plateau where the hotspots are well-defined. The size of each hotspot tends to increase with increasing tip voltage, and they appear to merge into connected areas. This behavior is clearly
shifts the overall position of the riser in back-gate voltage (black dashed lines, Figure 4b). The peak in the conductance due to the hotspot moves through the riser with a steeper slope in the $(V_T, V_{BG})$ plane (white dashed line). As mentioned previously, the potential perturbation from the entire probe can be well-described by the sum of two Lorentzians, one broad and shallow and the other narrow and deep. Hence we ascribe the difference in these slopes to the smaller capacitive coupling between the entire sample and the tip-cone, and the large coupling between the saddle point and the tip-apex. The typical form of this peak in conductance, $G_{SP}(V_T)$, is shown superimposed in Figure 4b, where we have subtracted a linear function with slope $\beta$ from the raw data to eliminate the long-range gating effect of the tip-cone ($G_{SP}(V_T) = G(V_T) - \beta V_T$). The full-width of the peak in $V_T$ is $\approx 2$ V, and at its maximum the conductance increases by a few percent of $e^2/h$. We attribute this peaked form to the fact that transmission across the saddle point is likely to peak when the Fermi level is equal to the saddle point potential.

We note that some of these observations contrast with other work investigating the role of localized states in 2DEGs, where images consisting of conductance halos revealed the presence of the Coulomb-blockaded localized states on either side of the saddle points, such as those in GaAs and graphene, stemming from Aharonov-Bohm oscillations of edge states in InGaAs/InAlAs quantum rings. As was recently highlighted by bulk transconductance measurements of graphene, at low bias it is likely that the extended single-particle states described by our model will be both capacitively and tunnel coupled to such localized states. The large bias ($\approx 10$ mV) used in our experiment probably exceeds the charging energy ($\approx 5$ meV) of tunnel-coupled localized states, however, in such a way that transport is dominated by percolation rather than Coulomb-blockade physics. The hotspots we observe may still be influenced by the charging and discharging of capacitively coupled states not directly involved in transport, and including nonlinear screening effects in our simulations is a fruitful direction for future work. Performing SGM at lower source-drain bias will also enable these interaction effects to be explored in greater detail.

In conclusion, we have examined the breakdown of the QHE using low temperature scanning gate microscopy and numerical simulations. In the quantum Hall regime, the position of the scanning probe tip has a weak influence on transport because conduction occurs at the edges while bulk localized states are well-isolated from each other. During quantum Hall breakdown we found that the conductance is strongly modulated by the tip at specific locations, and these conductance “hotspots” were found to repeat at the same relative filling factor. To understand our experimental observations we performed numerical simulations based on a network model for percolation between localized states. By comparing the evolution of the structure and intensity of the conductance modulations in the simulated and measured scanning gate images we were able to optimize the network parameters, yielding a 60 nm node separation and disorder fluctuation of $\approx 10$ meV, both in good agreement with previous studies. Finally, by imaging at different tip potentials we found strong evidence to suggest that conductance modulations at individual hotspots are dominated by the transmission probability of localized states across saddle points in the disorder potential. Our results demonstrate that SGM is a powerful tool for probing the quantum Hall state in graphene and supplies important information about the interaction

Figure 4. (a) Sequence of SGM images captured at a fixed back-gate voltage $V_{BG} = 7.4$ V with tip voltages of $-0.8$ V (image 1) to 0.8 V (image 5) in 0.4 V steps. (b) Numerical derivative of the conductance with respect to the tip voltage as a function of back-gate and tip voltage. The tip is panned over the hotspot indicated by a cross in image 2 of part (a). Black and white dashed outlines indicate the edges of the riser between quantum Hall plateaus and the conductance peak due to the hotspot, respectively. The superimposed solid line profile shows the saddle-point conductance as a function of tip voltage along the solid horizontal line ($V_{BG} = 14.5$ V). A linear slope has been subtracted to remove the background gating effect of the tip-cone. (c) Normalized autocorrelation profile as a function of tip voltage.
between potential disorder and magnetic field induced localization in graphene devices.

Methods. Our scanning probe microscope head (AttoAFM I) is mounted to the mixing chamber of a dilution refrigerator, and the oscillation of the cantilever is measured using standard interferometric detection with a fiber-based infrared laser. We use a Pt/Ir coated cantilever (NanoWorld ARROW-NCPt) with a nominal tip radius of 15 nm. To avoid any cross-contamination between the tip and the flake during SGM, once the flake is found using tapping mode, we switch to lift mode with the static tip at a lift height of ≈50 nm. As a precaution against drift when scanning close to the 100 nm-thick metallic contacts, we measure the conductance using an RF lock-in amplifier with an excitation frequency matched to the resonant frequency of the cantilever over the bare SiO$_2$. The stray field from the metallic contacts is sufficient to excite the cantilever into oscillation over the SiO$_2$, though over the graphene the cantilever is off-resonance and static. To obtain a good signal-to-noise ratio at these lift heights, we use an excitation voltage of 10 mV. While this is rather large for low-temperature transport measurements, it is still less than the energy separation (≈50 meV) between consecutive LLs at the magnetic fields and filling factors examined in our experiments.

ASSOCIATED CONTENT

Supporting Information

A movie showing the evolution of the conductance hotspots with back-gate voltage and further details of the Chalker–Coddington model. This material is available free of charge via the Internet at http://pubs.acs.org.

AUTHOR INFORMATION

Corresponding Author

*E-mail: mrc61@cam.ac.uk.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was financially supported by the European GRAND project (ICT/FET, Contract No. 215752).

REFERENCES

(38) Berezhovsky, J.; Westervelt, R. M. Nanotechnology 2010, 21, 274014.
NOTE ADDED AFTER ASAP PUBLICATION

The Table of Contents graphic and Abstract graphic were incorrect in the version published ASAP October 18, 2012. The corrected version was re-posted on November 1, 2012.