Giant Magnetoresistance in a Chemical Vapor Deposition Graphene Constriction

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ABSTRACT: Magnetic field-driven insulating states in graphene are associated with samples of very high quality. Here, this state is shown to exist in monolayer graphene grown by chemical vapor deposition (CVD) and wet transferred on Al₂O₃ without encapsulation with hexagonal boron nitride (h-BN) or other specialized fabrication techniques associated with superior devices. Two-terminal measurements are performed at low temperature using a GaAs-based multiplexer. During high-throughput testing, insulating properties are found in a 10 μm long graphene device which is 10 μm wide at one contact with an ≈440 nm wide constriction at the other. The low magnetic field mobility is ≈6000 cm² V⁻¹ s⁻¹. An energy gap induced by the magnetic field opens at charge neutrality, leading to diverging resistance and current switching on the order of 10⁴ with DC bias voltage at an approximate electric field strength of ≈0.04 V μm⁻¹ at high magnetic field. DC source–drain bias measurements show behavior associated with tunneling through a potential barrier and a transition between direct tunneling at low bias to Fowler-Nordheim tunneling at high bias from which the tunneling region is estimated to be on the order of ≈100 nm. Transport becomes activated with temperature from which the gap size is estimated to be 2.4 to 2.8 meV at B = 10 T. Results suggest that a local electronically high quality region exists within the constriction, which dominates transport at high B, causing the device to become insulating and act as a tunnel junction. The use of wet transfer fabrication techniques of CVD material without encapsulation with h-BN and the combination with multiplexing illustrates the convenience of these scalable and reasonably simple methods to find high quality devices for fundamental physics research and with functional properties.

KEYWORDS: multiplexed device arrays, graphene, CVD, giant magnetoresistance, magnetotransport, Fowler-Nordheim tunneling, direct tunneling

Insulating phases in graphene at high magnetic fields are typically observed only in exceptionally high quality devices, characterized by high carrier mobility and low residual carrier density. Demonstrations have been almost exclusively limited to exfoliated graphene flakes¹−⁶ and, recently, ultrahigh mobility graphene⁷ grown by chemical vapor deposition (CVD) and encapsulated with hexagonal boron nitride (h-BN).⁹ These devices are fabricated from monolayer material using sophisticated methods that limit scalability such as the use of exfoliated material, suspension of the graphene,¹⁰,¹¹ fabrication on h-BN substrates,¹²,¹³ or encapsulation with h-BN.⁹,¹⁴,¹⁵ At a high perpendicular magnetic field, the 2D density of state becomes quantized in Landau levels with energy E_{LL} = \frac{\hbar c v_F}{\sqrt{2} \hbar e B N}, where N is the Landau level index, v_F is the Fermi velocity, \hbar is the reduced Planck constant, and \epsilon is the electron charge, and an insulating phase can exist at charge neutrality⁵,¹⁴ characterized by a resistance that diverges with B as a magnetic field-dependent energy gap opens in the N = 0 Landau level. Here, we find such an insulating state can exist in conventionally wet transferred monolayer CVD graphene devices fabricated on amorphous oxides.
The device is contained within an array of CVD graphene devices nominally patterned as 10 μm-wide squares measured using a multiplexer to cryogenic temperatures. Multiplexing is a means for high-throughput testing and characterization of many devices, and the large number tested can lead to the discovery of individual devices with exceptional qualities that can be opportunistically studied. Here, a narrow constriction of ≈440 nm is created at one end of the graphene square, apparently by graphene tearing during fabrication. The resistance at charge neutrality diverges with B showing insulating behavior and an energy gap that increases with B. The conductivity appears to be reasonably temperature-independent below ∼2 K, and an increase in T shows activated behavior from which the energy gap is estimated to be ∼2.4−2.8 meV at B = 10 T. This behavior suggests the existence of a region of high electronic quality within the narrow channel, which dominates transport at high B. The analysis of charge transport mechanisms using DC source−drain bias measurements and quantum tunneling models at low T shows a transition from direct tunneling to Fowler−Nordheim tunneling with increasing DC bias from which the length of the tunneling region is estimated to be on the order of ≈100 nm. The device shows the current switching of order 106 with DC source−drain bias for an approximate electric field strength of ∼0.04 V μm⁻¹.

Here, although the narrow constriction is unintentionally created, it is straightforward to create the same geometry deliberately. A second device is fabricated with a similar contact configuration and a 600 nm-wide constriction defined next to one contact by electron−beam lithography. Similar behavior is observed in terms of an insulating state although being impacted by lithographic patterning and doping, potentially indicating the quality of the constriction edge as an important consideration. We speculate that less disorder is present in the unintentional constriction due to its formation as a result of tearing, where edges are never exposed to plasma etching, which can cause greater edge disorder and contamination compared to cleaner alternatives thus motivating further studies toward developing clean processes for constriction edges such as h-BN etch masks. The observation of the insulating phase at charge neutrality in wet transferred CVD graphene on amorphous oxide suggests the intriguing potential of developing systems to investigate exotic quantum phenomena using conventional and scalable fabrication techniques. A primary application of multiplexing is the statistical analysis of large data sets and identifying trends in data ensembles. Statistical analysis identifies general behavior, allowing outliers to be selected and studied in detail. Our present work highlights the capability of our multiplexing system for an in-depth study of the physics of such channels using numerous techniques including magnetic field, source−drain bias, and temperature from which one can obtain a detailed understanding of the fundamental phenomena. The identification of optimal devices has been a bottleneck in studies of fundamental physics, since multiple devices usually must be measured before a suitable one is found. The multiplexer expedits this process in a much more efficient manner. Our study also highlights the ability of the multiplexer to drive device development, since the inherent inhomogeneity in nanostructure devices leads to these outliers that can have desirable properties. The study of the outliers reveals a particular set of device conditions creating the observed behavior, and the identification of differences from other devices in the array provides information needed to reproduce the behavior deterministically.

RESULTS AND DISCUSSION

The multiplexer is fabricated on a GaAs/AlGaAs heterostructure containing a two-dimensional electron gas (2DEG) with a conducting path from one input to 16 outputs. Individual graphene devices are connected to each output and selected using addressing gates. Multiplexer operation, fabrication, and layout are described in ref 16. The device measured here is different from that in ref 16 where the emphasis was on the versatility of the multiplexer for high-throughput testing and compatibility with diverse nanoelectronic devices via integration of arbitrary nanomaterials including 2D materials (both CVD-grown films and exfoliated flakes) and semiconductor nanowires. The multiplexing platform can also be adapted for devices fabricated within a 2DEG and controlled by surface gates.

Here, the array of graphene devices is patterned in a monolayer CVD graphene film after wet transfer to the multiplexer host chip, as described in the Methods. Scanning electron microscopy images of the measured device are shown in Figure 1a,b. The image shows a narrow constriction next to a contact, and the region around the constriction appears to have reasonably well-defined edges and angles, suggesting the graphene constriction was formed by tearing along clear crystallographic directions. Tearing has been observed to give armchair or zigzag termination, which could indicate the constriction edges are atomically clean and feature minimal...
The constriction is 440 nm at the narrowest point. Figure 1c shows a schematic cross section through the device (side profile). Source–drain contacts are created prior to graphene transfer, and 95 nm thick Al₂O₃ is deposited by atomic layer deposition above the back gate. The two-terminal differential conductance is measured in a 3He cryostat.

**Transport Characterization.** Figure 1d shows resistance (R) as a function of back gate voltage (Ṽ) at B = 0 T and T = 0.29 K. The black curve is a fit to the data using

\[
R = R_p + \frac{L}{W} \left( \frac{1}{\epsilon \mu \sqrt{\frac{n_0}{2} + n^2}} \right)
\]

where R_p is the parasitic resistance, L/W is the length to width ratio, \(\mu\) is the device mobility, n_p is the residual carrier density, and n is the back gate-dependent carrier density. The density is given by \(n = C_G(V_G - V_0)/\epsilon_0\) where \(C_G\) is the gate capacitance per unit area \(\epsilon_0\) is the free space permittivity, \(\epsilon\) is the oxide permittivity, and \(\epsilon_0\) is the oxide thickness.

The mobility and parasitic resistance are estimated separately for electrons and holes, i.e., for \(V < V_0\) (\(V \geq V_0\)), \(\mu = \mu_{\text{hole}}\) and \(R_p = R_{p\text{hole}}\) (\(\mu = \mu_{\text{electron}}\) and \(R_p = R_{p\text{electron}}\)). For this equation, \(V_0\) is obtained by initially fitting data with density-independent \(R_p\) and \(\mu\), giving 0.52 V. Fitted parameters are \(R_{p\text{hole}} = 17.4\ \Omega\), \(R_{p\text{electron}} = 19.0\ \Omega\), \(n_0 = 1.3 \times 10^{11} \text{ cm}^{-2}\), \(\mu_{\text{hole}} = 5350\), and \(\mu_{\text{electron}} = 6360\ \text{cm}^2 \text{V}^{-1} \text{s}^{-1}\) for \(L/W \approx 2.3\). L/W is estimated by dividing the conducting area into squares and analyzing the resulting resistor network to give an equivalent length-to-width ratio.

Figure 1e shows the Landau level splitting with the magnetic field with \(d\mathcal{R}/dV_G\) plotted as a function of \(B\) and \(V_G\). Local resistivity maxima corresponding to where \(+1\) Landau levels are half filled are indicated by dotted white lines, and regions between correspond to quantum Hall plateaus. The gate capacitance is estimated from the gate dependence of quantum Hall states \(\nu = \pm 2\), where \(\nu\) is the filling factor, using \(dn/dB = ne/h\) and \(dn/dV_G = C_G/\epsilon\), giving \(C_G \approx 60\ \text{nF cm}^{-2}\) and \(\epsilon = 6.4\). Landau level splitting also provides an alternative estimate of \(\mu\) using the approximation of \(\nu \approx 1/B\) at \(B\) where Landau levels can be resolved.\(^{40}\) This gives a possible mobility up to \(\mu \approx 8000\ \text{cm}^2 \text{V}^{-1} \text{s}^{-1}\), as discussed in Figure S3a, and mean free path \(l = \nu_p \tau \approx 17\ \text{nm}\) using scattering time \(\tau \approx 1/\omega_c\) cyclotron frequency \(\omega_c = \nu_p \sqrt{2eB/\hbar}\), and \(\nu_p = 1 \times 10^6\ \text{m} \text{s}^{-1}\).

**Magnetoresistance Measurements.** Figure 2a shows R as a function of \(V_G\) at \(B = 7, 8, 9, \) and 10 T and \(T = 0.29\ K\). The resistance at charge neutrality increases by almost 3 MΩ at \(B = 10\ T\). Figure 2b shows R at \(V_G = 0.31\ \text{V}\) as a function of \(T\), which diverges with \(B\) for the lowest \(T\). The estimated \(B\)-dependent resistance of the GaAs multiplexer is subtracted from the data in Figure 2 as described in Figure S1. Data in Figure 2b,c are measured at \(V_G = 0.31\ \text{V}\). Data for Figure 3 are obtained at \(V_G = 0.32\ \text{V}\) after reoptimization. Both are taken to represent the high \(B\) insulating state at \(\nu = 0\) when discussing each figure. Fitting transfer characteristics at \(B = 0\ T\) using eq 1 estimates a charge neutrality point voltage of 0.52 V. Differences may arise since the low \(B\) value is given by fitting methods over a broad range of \(V_G\), whereas values at \(B = 10\ T\) are experimentally determined to be as close as possible to the maximum \(R\). Additionally, differences may conceivably arise since the low \(B\) characteristics likely represent an average of the entire graphene area, whereas at high \(B\) a small region within the constrictions appears to dominate transport characteristics.

The conductance \(G = 0.31\ \text{V}\) is plotted as a function of \(T\) on a lin-log scale in Figure 2c for \(B = 7, 8, 9, \) and 10 T. This behavior becomes visible for \(B > 7\ \text{T}\) and above. Similar to that in ref 1, G appears to become relatively \(T\) independent at the lowest temperatures measured. Solid lines show fits to the data for activated transport using a Fermi–Dirac distribution\(^{15,41}\)

\[
G = \gamma + \beta/(1 + e^{(E - E_F)/k_BT})
\]

where \(E_F = 0.7, 1.0, 1.6, \) and 2.4 meV is the magnetic field-dependent energy gap at \(B = 7, 8, 9, \) and 10 T, respectively, \(k_B\) is the Boltzmann constant, \(\gamma\) is the zero \(T\) conductance, and \(\beta\) is a pre-exponential factor. Fits at each \(B\) are performed independently with \(E_F, \gamma, \) and \(\beta\) as fitting parameters. Data are first fit using the Arrhenius equation, which gives energy gaps comparable to the thermal energy \(k_BT\) at higher \(T\) values for which the measurement is performed (up to \(T = 25\ K\)). Therefore, the Fermi–Dirac function is used instead. Fitting using the Arrhenius equation is shown in Figure S2b. The energy gap is shown as a function of \(B\) in Figure 2d. If a linear dependence \(E_F = \Delta E_F - \Gamma\) is assumed,\(^{5,45}\) where \(\Delta E_F\) is Zeeman energy \(g\mu_BB\), \(\mu_B\) is the Bohr magneton, and \(\Gamma\) is the disorder broadening of Landau levels, parameters \(\Gamma = 39.2\ \text{K}\) and effective Lande g-factor \(g\approx 10\) are enhanced above the free-electron g-factor \(g = 2\), as seen previously for exfoliated graphene\(^{21,13}\) and attributed to exchange interactions.\(^{15,42}\) The quantum lifetime\(^{5,45}\) estimated from Landau level broadening
Figure 3. DC source–drain bias measurements in the $\nu = 0$ insulating state. Magnetic field ($B$) = 10 T for all data in (a)–(c). (a) Resistance as a function of DC source–drain bias at $\nu = 0$, $\Delta n_1 = 2.2 \times 10^{10}$ cm$^{-2}$, and $\Delta n_2 = 4.5 \times 10^{10}$ cm$^{-2}$, where $\Delta n$ represents the change in density from $\nu = 0$. Temperature $T = 0.29$ K. (b) Temperature dependence at $\nu = 0$. (c) Conductance $G$ at $\nu = 0$ for select $V_{DS}$ as a function of $T$ after subtracting series resistance. The dashed line is a fit at $V_{DS} = 0$ mV using eq 2. The inset shows $\Delta E = k_B \Delta T$ where $\Delta T$ is the temperature difference between data points at $T = 2$ K and the same conductance on the $V_{DS} = 0$ mV fit line. Dashed lines show linear fits with the same absolute value of gradient for positive and negative $V_{DS}$.

$\tau_q = h/\Gamma \approx 195$ fs is reasonably comparable to scattering lifetimes estimated from mobility using the Drude formalism $\tau = (\hbar/2eV_F)(n/\pi)^{1/2} \approx 105$ fs at $n = 2 \times 10^{12}$ cm$^{-2}$. However, the range of data in Figure 2d is too small to give a true indication of the relationship between $E_a$ and $B$. An approximately linear relationship appears in some devices described in the literature$^{5,15}$ but not necessarily in others.$^{10}$ Reference 14 shows a linear relationship for a device engineered to minimize the Coulomb interaction and a square root dependence consistent with a Coulomb energy relationship in a separate device without this reduction. The estimated gap sizes here are much smaller than the Coulomb energy $E_C = e^2/4\pi\epsilon_0\epsilon_l l_0$ ($E_C = 28$ meV at $B = 10$ T), where $l_0$ is the magnetic length $l_0 = \sqrt{\hbar/eB}$. Previous studies show either $E_a$ to be small$^2$ compared to $E_C$ or that they are similar values.$^{13}$

Fitting parameters $\gamma$ and $\beta$ are shown in Figure 2e,f, respectively. The $\gamma$ value describes $G$ in the low $T$ limit and nears zero as $B$ increases. In a scenario of transport dominated by a region of gapped graphene at high $B$, this may represent hopping transport that diminishes as the energy gap $E_a$ increases and the size of the wave function reduces with $B$, leading to less hopping between sites. The pre-exponential factor $\beta$ is reasonably similar for $B = 8, 9, 10$ T for fits performed independently at each field and is reduced at lower $B$ as the gap becomes small. The $\beta$ term may be related to the geometry of the gapped region, becoming reasonably constant above a crossover field where the gap opens. The insulating behavior does not occur in other devices in the multiplexed array that do not contain a narrow constriction. The previous observation of insulating states in graphene associated with high electronic quality devices supports a scenario where a high quality region exists within the constriction and dominates the transport characteristics at high $B$.

**DC Source–Drain Bias Measurements.** The insulating state is further investigated by applying a DC source–drain bias across the graphene device ($V_{DS}$). Figure 3a shows $R$ as a function of $V_{DS}$ at select $V_G$ around charge neutrality for $T = 0.29$ K and $B = 10$ T. Data representing the high $B$ insulating state at $\nu = 0$ are obtained at $V_G = 0.32$ V; data at other $V_G$ are labeled according to $\Delta n = C_G(V_G - 0.32)/e$. Supporting Information describes how $V_{DS}$ across the graphene is extracted from the total source–drain bias applied to the circuit ($V_B$), accounting for series resistance. The temperature dependence at $\nu = 0$ is shown in Figure 3b. Energy scales associated with DC bias are reasonably similar to $E_a$ from Fermi–Dirac fitting, i.e., in Figure 3b, the full width at half-maximum (fwhm) of $\approx 3$ meV at $T = 0.29$ K and 2 K, accounting for a vertical offset from series resistance ($R_s$) of $\approx 219$ kΩ. Data in Figure 3a,b are plotted before subtracting $R_s$. There is a small effective gating of the graphene at high $V_{DS}$ due to the source–drain bias, discussed in Figure S4; however, the effect is negligible.

Figure 3c shows $G$ as a function of $T$ at select $V_{DS}$ after subtracting $R_s$. For clarity, data are shown for $V_{DS} \leq 0$, and data for $V_{DS} > 0$ are similar. The dashed line shows a fit to $V_{DS} = 0$ mV data using the Fermi–Dirac distribution. Parameters are as follows: $E_a = 2.8$ meV, $\tau = 0.02 e^2/h$, and $\beta = 1.4 e^2/h$. Differences from the parameters estimated for $B = 10$ T data in Figure 2c may arise from reoptimizing $V_G$ to find the maximum $R$. Data for finite $V_{DS}$ appear to fall along the $V_{DS} = 0$ mV fit line at high $T$ and diverge as $T$ is reduced, becoming reasonably $T$ independent for $T < 2$ K. The gap $\Delta T$ between $T = 2$ K data points for finite $V_{DS}$ and the $V_{DS} = 0$ mV fit line is shown in the inset converted to energy $\Delta E = k_B \Delta T$. Data at $T = 2$ K are chosen since $G$ becomes reasonably $T$ independent below this $T$. This may represent an equivalent thermal energy or an equivalent reduction in barrier height that would be required at $V_{DS} = 0$ mV to achieve the same effect. Dashed lines show linear fits with the same gradient (absolute value) for $V_{DS} > 0$ and $V_{DS} < 0$ mV, which cross the horizontal axis at $l_{DS} = 0.6$ meV similar to the AC excitation voltage $V_{AC} = 400 \mu V$ (rms). The relationship appears linear up to bias voltages similar to the activation energy. We do not observe a power law relationship between $\Delta T$ and applied power at each $V_{DS}$, which could be expected if $\Delta T$ represented a rise in electron temperature.$^{43}$ This suggests the main impact of $V_{DS}$ within this regime is to change the bias across the barrier. However, this is better investigated by analyzing the current–bias voltage characteristics.

**I–V Characteristics.** Figure 4a shows the DC current $I_{DS}$ as a function of $V_{DS}$ for select $V_G$ at $T = 0.29$ K. The current is obtained by integrating the measured differential conductance $G = dI_{DS}/dV_{DS} over the total DC bias applied to the circuit $V_B$. The $I_{DS}$ remains near zero over the widest range of $|V_{DS}| \lesssim 3$ mV at $\nu = 0$, an energy scale similar to $E_a$. Figure 4b shows the temperature dependence for the $\nu = 0$ insulator. Above $T = 2$ K, data become much more linear, as transport becomes thermally activated. The $I_{DS}/V_{DS}$ characteristics are shown on a log–lin scale in Figure 4c,d, for different $n$ at $T = 0.29$ K and different $T$ at $\nu = 0$, respectively. Positive $V_{DS}$ data are plotted
in Figure 4c–h. Negative $V_{DS}$ data show similar results and are shown in Figure S5. The current changes by nearly 4 orders of magnitude for $\Delta V_{DS}$ from 0 to $\approx 4$ mV at $T = 0.29$ K and $\nu = 0$. If the barrier length is on the order of $\approx 100$ nm (see later analysis); this corresponds to an electric field strength of $\approx 0.04$ V $\mu$m$^{-1}$. The narrow constriction connects to the Ti/Au contact on one side and the wider graphene area on the other. The similarity of data for positive and negative $V_{DS}$, Figures 4 and S5, respectively, suggests that any asymmetry caused by different types of contacts to a tunneling region within the constriction is small.

Data at $T = 0.29$ and 2 K cross at high $V_{DS}$ likely caused by uncertainties in the series resistance correction. Therefore, only trends at separate $T$ will be considered. In this measurement, $R_s$ depends on $T$ and total bias $V_{app}$ applied to the circuit. The $V_B$ dependence of $R_s$ arises from a gating effect where the bias across the multiplexer changes the Fermi energy in the GaAs 2DEG relative to the addressing gates. The resulting change in resistance manifests as an approximately linear background with DC bias voltage, dependent on $T$, and is corrected for as described in the Supporting Information. The accumulation of estimates makes comparing data between different $T$’s less reliable when $V_{DS}$ is the highest and the graphene resistance has become small. It is straightforward to remove the $R_s$ dependence on DC bias with changes to the multiplexer operation during the experiment by maintaining a fixed potential difference between addressing gates and the 2DEG.\textsuperscript{45} This will allow easier characterization of $I$–$V$ curves in the future.

Tunneling mechanisms are investigated by analysis of the current–bias voltage characteristics using eqs 3 and 4.\textsuperscript{21–24} Equation 3 describes direct tunneling through a trapezoidal barrier at low bias

$$\ln \left( \frac{I}{V^2} \right) \propto \ln \left( \frac{1}{V} \right) - \frac{2d \sqrt{2m \phi}}{\hbar} \left( \frac{1}{V} \right)$$

where $\phi$ is the barrier height, $m$ is the effective mass, and $d$ is the length of the tunnel barrier. Plots of $\ln(1/V^2)$ as a function of $1/V$ show a linear relationship in this regime. At high bias ($V > \phi$), the barrier becomes triangular and transport can be described using

$$\ln \left( \frac{I}{V^2} \right) \propto - \frac{4d \sqrt{2m \phi^3}}{3e \hbar} \left( \frac{1}{V} \right)$$

known as Fowler-Nordheim (FN) tunneling or field emission. Plots of $\ln(1/V^2)$ as a function of $1/V$ show a linear relationship with a negative gradient. Figure 4e shows $\ln(I_{DS}/V_{DS}^2)$ as a function of $1/V_{DS}$ at different $V_{DS}$’s for $T = 0.29$ K. A linear trend is observed at low $V_{DS}$, consistent with direct tunneling. As $V_{DS}$ increases, an inflection point suggests a transition between tunneling regimes and a changing barrier shape between trapezoidal and triangular. The schematic diagrams indicate the barrier shape at different $V_{DS}$’s. A zoom-in of the high $V_{DS}$ regime is shown in Figure 4g on a $\ln(I_{DS}/V_{DS}^2)$ vs $1/V_{DS}$ scale. The vertical dotted line corresponds to the inflection point voltage ($V_f$) for the $\nu = 0$ state. The negative gradient at high bias is consistent with FN tunneling. The bias energy at the inflection point $eV_f \approx 1.8$ meV is lower than the barrier height $E_f$ from Fermi–Dirac fitting. Other studies have also observed $eV_f$ to be smaller than the estimated barrier height,\textsuperscript{21} attributed in part to the approximation of the barrier being simple trapezoidal/triangular shapes instead of being fully treated.

The $T$ dependence of the $\nu = 0$ state is shown in Figure 4f,h. Results at $T = 0.29$ and 2 K are consistent with a transition between direct tunneling and FN tunneling. No inflection

![Figure 4. DC current–voltage characteristics. Magnetic field $B = 10$ T for all data in (a)–(h). The top row of (a), (c), (e), and (g) show data at $T = 0.29$, $\Delta n_1 = -2.2 \times 10^{10}$ cm$^{-2}$, and $\Delta n_2 = -4.5 \times 10^{10}$ cm$^{-2}$ for $T = 0.29$ K. Data representing the $\nu = 0$ insulating state are obtained at $T = 0.32$ V and $\Delta n = C_2(V_g - 0.32)$. The bottom row (b), (d), (f), and (h) show the $T$ dependence at $T = 0$. (a, b) Current $I_{DS}$ as a function of source–drain bias across the graphene device $V_{DS}$ (c, d) Log-\ln plots of $I_{DS}$–$V_{DS}$ (e, f) $\ln(I_{DS}/V_{DS}^2)$ as a function of $1/V_{DS}$. Schematic diagrams in (e) represent the barrier shape in different bias regimes. (g, h) Fowler-Nordheim plots of $\ln(I_{DS}/V_{DS}^2)$ as a function of $1/V_{DS}$. The vertical dotted line labeled $V_f$ in (g) corresponds to the inflection point voltage for $\nu = 0$, marking a transition between Fowler-Nordheim and direct tunneling. The diagonal dotted line shows a linear fit at high bias for Fowler-Nordheim tunneling. Data at positive $V_{DS}$ are shown in (c)–(h). Negative $V_{DS}$ shows similar results.
Figure 5. (a) Optical micrograph prior to Al₂O₃ deposition with false color graphene (blue) and source–drain contacts (yellow) illustrating the device geometry. The scale bar is 2 μm long. (b) Top down schematic of the lithographic pattern defining the constriction [dashed black box outlined in (a)]; colors are the same as (c). (c) Schematic cross section from source to drain. (d) Magnetic field dependence of transfer characteristics at T = 1.5 K. (e) Colormap of R as a function of V_G and B. Horizontal dashed lines and arrows indicate charge neutrality points for peaks a and b as labeled. (f) Temperature dependence of transfer characteristics at B = 12 T. (g) Maximum R of peak a and peak b resistance measured at V_G = 18 V as a function of B at T = 1.5 K. (h) Resistance R as a function of ln|B| at B = 12 T for peaks a and b.

point occurs at T ≥ 5 K, consistent with the T at which transport has become thermally activated (i.e., the G in Figures 2c and 3c does not change significantly with T until T > 2 K). Activated carriers are able to overcome the barrier leading to thermionic emission.22,23

Equation 4 is the linearized form21 of I² ∝ V²exp(−4d√(2mfi²/3heV)) from which the slope in the FN regime is given by −4d√(2mfi²/3he). A linear fit to ν = 0 data at high bias in Figure 2g is shown by a dotted line, which gives barrier length d on the order of ≈100 nm. The effective mass is estimated as26 m = e/ν = (h²n/4πe²)(1/2) using ν = 1 × 10⁶ s⁻¹ and n = n₀ (the residual carrier density), since data are obtained at charge neutrality. Barrier height φ = E₀ = 2.8 meV is used, provided by the Fermi–Dirac fitting of V_DS = 0 mV data in Figure 3c. The barrier length is consistent with the estimated Fermi wavelength λ_F = √(4π/n) = 97 nm at n = n₀. The barrier length of ≈100 nm is only given as an approximate estimate, since within the fitting range, the data are not yet truly linear, and it may be that V_DS is not yet sufficiently high. Further deviations in linearity can occur due to T and image effects20,21,47 that are not included in eq 4. The equation also assumes a simple triangular barrier shape and is an approximation in the high bias and zero T limit. These factors can contribute to uncertainties in the estimate of d. Additionally, uncertainties in R_S at high V_DS may also cause deviations from linearity. The transition between Fowler-Nordheim and direct tunneling has not previously been reported in this insulating regime, and to our knowledge, detailed I–V measurements and analysis including temperature dependence have not hitherto been performed. Where I–V sweeps were carried out at a single T, the nonlinear regime indicated an energy scale an order of magnitude larger than the activation energy, suggesting charge transport may occur over several connected insulating areas.10 In contrast, our results indicate energy scales similar to E₀ perhaps suggesting an individual insulating region.

An aspect that distinguishes this work is the use of bottom contacts. These have potential to induce strain in graphene next to the contact edge, creating the possibility for the physical origin of these effects to lie in the combination of narrow constriction and strain, where strain from metal contacts can affect device properties.7 The high quality region may also indicate cleaner interfaces and low defect density within the narrow constriction, and strict control of the interface quality may be necessary for high yields. We do not observe symmetry breaking and ν = ± states as have been reported in samples with much higher mobilities5,49 or measured at much higher magnetic fields.5 Similar to refs 2, 3, and 50, this may be attributed to comparatively higher disorder in our devices or insufficiently high B. For comparison, devices hosting the high B insulating state with very similar mobilities to ours, refs 2 and 3, did not detect ν = ±1 states for B up to 30 and 18 T, respectively. It is also likely that the amount of residual carrier density or disorder in our device precludes the existence of low B insulating transitions that appear within the global phase diagram for 2D systems in a magnetic field,51 where increasing density can reduce the critical field of the transition to B = 0 T.52 Instead, our results appear to be consistent with the phase diagram in ref 3.

The insulating state is often observed as B is increased,1–4,10,11,53 despite the fact that the ground state at ν = 0 is predicted to be a quantum Hall ferromagnet with counter-propagating edge modes, where each edge section contributes R = h/e² and total R is given by the combination of these edge sections.13,14,54–56 It has recently been shown that quantum Hall ferromagnetism can exist in devices engineered to screen the Coulomb interaction, whereas the insulating ν = 0 state is present in devices where this is not the case.53 This was attributed to the indirect modification of lattice–scale electron–electron interactions, which determine the ground state and can lead to either ferromagnetic or insulating phases.57

The observation of the insulating state, previously requiring the electronic quality given by exfoliation and encapsulation methods, in unencapsulated and wet transferred CVD graphene may be pertinent for the development of graphene-based devices to investigate physics phenomena due to the scalability and relative simplicity of these methods. To address this potential, the role of constriction width and possible strain at the contact edges should be investigated, since here, the behavior appears in a device with a narrow constriction and back contacts. Although the constriction is created unintentionally in the initial device, it is straightforward to determinately recreate with standard lithographic techniques. We deliberately create a similar geometry using electron–beam lithography and oxygen reactive ion etching to define a 10 μm-wide graphene channel with a 600 nm wide constriction next to one contact electrode in monolayer CVD graphene.
transferred on a doped Si wafer. The device layout is shown in Figure S5a–c, and fabrication and measurement details are given in the Supporting Information. An ~50 nm Al₂O₃ encapsulating layer is added by atomic layer deposition (ALD) after patterning,⁵⁸ and all data presented includes contact resistance.

Figure 5d shows R as a function of V_G at B = 0, 3, 6, 9, and 12 T and T = 1.5 K. Two clear resistance peaks are visible at B = 0 T (bold trace), labeled a and b. The colormap in Figure 5e reveals their evolution into two sets of Landau levels with charge neutrality points V_{CNP,a} = ~42 V and V_{CNP,b} = ~18 V for a and b, respectively, marked by the horizontal dashed lines and arrows. When one considers the graphene channel as two resistive components in series with distinct aspect ratios representing the constriction and main channel, there are indications that the behavior of peak a (peak b) is consistent with arising from the constriction (wide region). Specifically, the higher R of peak a at B = 0 T is consistent with a larger L/W ratio, which occurs in constriction, as supported by fitting the peaks separately using eq 1, shown in the Supporting Information. Additionally, nanopatterning graphene (the use of PMMA and planar etching) has been shown to lead to p-doping, which may explain the positive shift of V_{CNP} for peak a due to the enhanced impact of these doped etched edges on transport through the constriction relative to the wide section. Dual peak behavior is not observed in the unintentional constriction, possibly due to the tearing process resulting in cleaner constriction edges and reduced contamination such that there is no difference in V_{CNP} for graphene in the constriction and elsewhere in the device. We speculate that removing the larger graphene area, i.e., fabricating only narrow graphene channels between micron-spaced source–drain electrodes, may remove the secondary Dirac peak.

Figure 5g shows R at ν = 0 for peaks a and b as a function of B at T = 1.5 K. The resistance of peak a rises with B, whereas b plateaus at ~18.4 kΩ. The T dependence of peaks a and b at B = 12 T is shown in Figure 5h. Peak a shows insulating behavior, whereas peak b is relatively unchanged in comparison. The behavior of peak a is similar to the device with the unintentional constriction in terms of increasing R with B and insulating T dependence. The magnitude of the change in resistance for the insulating behavior is reduced compared to the unintentional constriction, which can be attributed to a combination of the higher base temperature (1.5 K vs 300 mK) and increased disorder and doping in the etched edges.⁷,²⁸ It is expected that devices with higher disorder will show a later onset of the insulating state such that R is lower at the same B and T.¹,¹¹,⁵³ For comparison, Figure 2b shows R ~860 kΩ at B = 10 T and T = 2 K. We speculate that less disorder is present in the unintentional constriction since it forms as a result of tearing,²⁵,₂⁶ such that the edges are never exposed to plasma etching. However, the trends of R with T and B in both devices are the same, and fitting data from Figure 5h using eq 2 estimates an energy gap E_g of ~0.9 meV at B = 12 T, compared to E_g of ~2.4–2.8 meV for the unintentional constriction.

We note that the qualitative difference in the B and T dependence of peaks a and b suggests behavior that is inherent to each charge neutrality point, and the effect of overlapping Landau levels is small. If this were not the case, both peaks might be expected to show the same behavior, since peak b overlaps a and vice versa. The strong T dependence at peak a in Figure 5f compared to the little discernible change around peak b and for V_G < 18 V (i.e., in the region of Landau levels from peak b) also suggests the T dependence around V_G = ~42 V (V_{CNP,b}) can be attributed to the ν = 0 state of peak a rather than overlapping Landau levels.

These data provide evidence that the insulating state at ν = 0 can be reproduced in deliberately fabricated CVD graphene devices and that the unintentional constriction may be benefiting from higher electronic quality due to the presumably torn edges. This suggests the importance of lithographic patterning processes that minimize edge disorder and contamination, such as using h-BN masks,²⁸,³⁸ when creating deterministically defined constrictions, and may imply the possibility for systems to investigate exotic physics using relatively simple and scalable fabrication methods with the implementation of such techniques. Our results highlight the use of multiplexing in driving device development, since by studying the device conditions leading to the initial observation of the insulating state, we are able to reproduce it deterministically and identify key factors for its existence. Additionally, the multiplexing platform combined with scalable fabrication such as wet transfer techniques of CVD material has demonstrated a convenient means by which devices can be identified for fundamental physics research or with functional properties.

Future expansion of the multiplexing tool to achieve four-probe capabilities and incorporate h-BN substrates or h-BN encapsulation presents intriguing possibilities to explore other diverse phenomena, including those in graphene devices on patterned h-BN.⁶⁰ The N = 0 Landau level in hybrid graphene systems has also shown intriguing behavior and could be investigated.¹⁶ Expansion to other nanomaterials is straightforward since nanowires, exfoliated 2D materials, and CVD-grown 2D materials can be transferred to the multiplexer using compatible fabrication methods.¹⁶ The onset of the high resistance state studied here in graphene has been shown to occur at lower magnetic fields in devices with higher mobility and lower disorder.¹,¹¹,⁵³ The reduction of the magnetic field required may bring CVD-grown graphene devices into the realm of giant magnetoresistance devices and magnetic switches.¹⁶ and motivate the development of graphene-based magnetic field detection or magnetic texture mapping sensors with cryogenic readout.

**CONCLUSIONS**

These results show the existence of a magnetic field-induced insulating state in monolayer CVD graphene with resistance that diverges with B at charge neutrality. This state is usually associated with very high quality devices made using exfoliated graphene flakes and suspended, encapsulated, or fabricated on extremely smooth substrates such as hexagonal boron nitride. The device studied here uses CVD-grown graphene that is unencapsulated and fabricated on an Al₂O₃ substrate created by atomic layer deposition. The insulating state shows activated behavior with an energy gap that increases with B. DC source–drain bias measurements reveal a transition between direct tunneling and Fowler-Nordheim tunneling at T = 0.29 and 2 K as bias increases. The results support the existence of a gapped region with a length on the order of ~100 nm in an ~440 nm-wide constriction created within a larger area of graphene. This work demonstrates the opportunistic study of properties serendipitously created in a device within a multiplexed array. High-throughput testing via the multiplexing platform increases the opportunity to identify...
such devices with unusual properties and study them and is compatible with different nanoelectronic devices including 2D materials and semiconductor nanowires, measured from cryogenic to room temperatures.\(^\text{16}\)

**METHODS**

These methods concern the primary device on the multiplexing platform. Methods relating to the second device are provided in the Supporting Information. The multiplexer is fabricated in a GaAs high electron mobility transistor (HEMT) in which a 2DEG forms 90 nm below the surface (Cavendish wafer V832). The mean sheet density and electron mobility are \(n_{\text{GaAs}} = 1.53 \times 10^{11} \text{ cm}^{-2}\) and \(\mu_{\text{GaAs}} = 8.17 \times 10^5 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}\), respectively, measured on a separate Hall bar device. A conducting path is defined within the 2DEG from one input to 16 outputs, and individual outputs are chosen by surface addressing gates. Multiplexer layout, fabrication, and operation details are described in ref 16, although the device measured here is different from that in ref 16. Source–drain contacts (Ti/Au) are fabricated on the multiplexer before transferring a monolayer graphene film grown by chemical vapor deposition onto Cu. The graphene is coated with poly(methyl methacrylate) (PMMA), and the Cu is etched using a 1.2% solution of ammonium persulfate etchant (\(\text{NH}_4\text{J}_2\text{S}_2\text{O}_8\)). The PMMA–graphene stack is transferred to the multiplexer after rinsing in a DI water bath and dried in air overnight before baking at 125 °C for 30 min and soaking in acetone to remove the PMMA. Graphene channels (10 \(\mu\text{m}\) wide) are defined between source–drain contacts (10 \(\mu\text{m}\) separation) by photolithography and oxygen reactive ion etching. For the device measured here, Raman measurements show a large 2D to G peak ratio across the entirety of the graphene, consistent with a single layer, shown in Figure S6. The two-terminal differential conductance is measured using 77 Hz AC voltage, \(V_{\text{AC}} = 400 \text{ µV (rms)}\), applied to the multiplexer and graphene in series. At \(B = 10\text{T}\), the maximum power dissipated in the graphene and energy loss rate per carrier are estimated to be \(<0.2\text{ pW}\) and \(<1.5 \times 10^{-13}\text{ W}\), respectively, which suggests a carrier temperature of \(<0.6\text{ K}\), as discussed in the Supporting Information.

**ASSOCIATED CONTENT**

*Supporting Information*

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsnano.1c09815.

Series resistance, energy gaps estimated using an Arrhenius equation, carrier mobility estimated using the quantum Hall effect, DC source–drain bias across the graphene, effective gating of the graphene due to source–drain bias, DC current–voltage characteristics for negative \(V_{\text{DS}}\) energy loss rates and carrier temperature, Raman spectroscopy, and methods for the deliberately defined constriction (PDF)

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*Notes*

The authors declare no competing financial interest.

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