In thermodynamic equilibrium, current in metallic systems is carried by electronic states near the Fermi energy, whereas the filled bands underneath contribute little to conduction. Here, we describe a very different regime in which carrier distribution in graphene and its superlattices is shifted so far from equilibrium that the filled bands start playing an essential role, leading to a critical-current behavior. The criticalities develop upon the velocity of electron flow reaching the Fermi velocity. Key signatures of the out-of-equilibrium state are current-voltage characteristics that resemble those of superconductors, sharp peaks in differential resistance, sign reversal of the Hall effect, and a marked anomaly caused by the Schwinger-like production of hot electron-hole plasma. The observed behavior is expected to be common to all graphene-based superlattices.

A rare exception is semimetallic graphene. At high carrier densities \( n \), the drift velocity in graphene is limited by phonon emission (6, 7), similar to other metallic systems. However, at low \( n \), thermal excitations can create a relativistic plasma of massless electrons and holes, the “Dirac fluid.” Its properties in thermodynamic equilibrium were in the focus of recent research (8–12), but the behavior at high biases represents an uncharted territory. Yet close to the Dirac point, even a small \( E \) can shift the entire Fermi surface and tap into a supply of carriers from another band (13, 14). This can trigger processes analogous to the vacuum breakdown and Schwinger particle-antiparticle production in quantum electrodynamics, in which they are predicted to occur at enormous fields of \( \sim 10^{15} \) V m\(^{-1} \) (15). Because such \( E \) is inaccessible, it is enticing to mimic the Schwinger effect and access the resulting out-of-equilibrium plasma in a condensed matter experiment (13, 14, 16). Certain nonlinearities observed near graphene’s neutrality point (NP) were previously attributed to the creation of electron-hole (e-h) pairs by means of a Schwinger-like mechanism (13, 14), but the expected intrinsic behavior was obscured by low mobility, charge inhomogeneity, and self-gating effects (6, 17).

We used graphene-based superlattices to identify an out-of-equilibrium state that sharply develops above a well-defined critical current \( J_c \). The current marks an onset of the Schwinger pair production and a transition from a weakly dissipative fluid-like flow to a strongly dissipative e-h plasma regime. The out-of-equilibrium Dirac fluid is realized at surprisingly small \( E \), thanks to the narrow electronic bands and low \( v_F \) characteristic of graphene superlattices (18, 19). The resulting dual-band transport leads to striking anomalies in longitudinal and Hall resistivities. Counterintuitively, an apparent drift velocity in this regime exceeds \( v_F \). With hindsight, we show that the current-induced critical state can be reached even in standard graphene, by using extra-high currents allowed by the point contact geometry.

The studied superlattices were of two types: graphene crystallographically aligned on top of hexagonal boron nitride (G/hBN) (20–23) and small-angle twisted bilayer graphene (TBG) (24–28). The superlattices were encapsulated in hBN, to ensure high electronic quality, and shaped into multiterminal Hall bar devices by using the standard fabrication procedures (29). The devices were first characterized by measuring their longitudinal resistivity \( \rho \) as a function of \( n \) as shown in Fig. 1, A to C, for three representative devices. The twist angles \( \theta \) were determined from measurements of Brown-Zak oscillations (30); for TBG, \( \theta \) was intentionally chosen away from the magic angle to avoid many-body states (27, 28). Aside from the familiar peak in \( \rho \) at zero doping, satellite peaks indicating secondary NPs were observed at \( n \) that agreed well with the \( \theta \) values (20–22, 26). For G/hBN superlattices, the low-energy electronic spectrum is practically identical to that of monolayer graphene (18), and the spectral reconstruction occurs only near and above the edge of the first miniband (Fig. 1D, top). By contrast, all minibands in TBG are strongly reconstructed (Fig. 1D, bottom) (19). At low biases (Fig. 1, A to C, and fig. S1), our devices exhibited transport characteristics similar to those reported previously for G/hBN and TBG superlattices (20–22, 26).

Next, we studied high-bias transport using current densities \( j \) up to 0.1 mA \( \mu m^{-1} \), limited only to avoid device damage. Unless stated otherwise, all the reported measurements were carried out at the bath temperature \( T = 2 \) K. The superlattices exhibited qualitatively similar current-voltage (I-V) characteristics (Fig. 1, E to G), which were nearly linear at \( j < 0.01 \) mA \( \mu m^{-1} \) and then rapidly switched into a high-resistance state so that the differential resistivity \( d\rho/dV \) showed a pronounced peak at a certain critical current \( J_c \). The behavior was universal, found in all our devices (more than 10) (figs. S3 and S6), if the Fermi energy was tuned inside narrow minibands (that is, away from the main NP in the case of G/hBN). The I-V characteristics in Fig. 1, E to G, strongly resemble the superconducting response, despite electron transport being ballistic at low \( j \) and viscous at moderate currents (31); \( \rho \) always remained finite, although could be as low as <0.01 kilohms, a few orders of magnitude smaller than \( d\rho/dV \) above \( J_c \). Further details are provided in Fig. 2 by showing \( d\rho/dV \) as a function of \( n \), where the narrow white arcs indicate peaks in \( d\rho/dV \). Considerable similarities
are clearly seen across different superlattice types. One feature shared by all the maps was the rapidly decreasing \(j_c\) as \(n\) approached NPs (Fig. 2, A to C, and figs. S2 and S6). The only exception was the main NP in G/hBN superlattices, where the resistivity in its vicinity increased monotonically for all accessible \(j\) (fig. S2).

To gain more insight, we studied the Hall effect in small (nonquantizing) magnetic fields \(B\). An example of such measurements for G/hBN near the hole-side NP is shown in Fig. 2D. At small \(j\), the Hall voltage \(V_H\) increased linearly with \(j\), and \(dV_H/dj\) was positive, reflecting the hole doping. However, \(dV_H/dj\) abruptly turned negative above \(j_c\), revealing a change in the dominant-carrier type. \(dV_{xy}/dj\) maps for the G/hBN and TBG superlattices are shown in Fig. 2, E and G. There are clear correlations between the longitudinal and Hall maps that so the peaks in \(dV/dj\) and the Hall effect’s reversal occurred at same \(j_c\). The observed nonlinearities were robust against \(T\) up to ~50 K, above which the peaks in \(dV/dj\) became gradually smeared (fig. S4).

This shows that Ohmic heating—which is generally expected at high \(j\) (14, 31, 32)—was not the reason for the critical-current behavior (29).

The rapid decrease in \(j_c\) near all secondary NPs prompts the question why such a critical-current behavior was not observed in graphene (33, 14) or near the main NP of G/hBN (Fig. 2A) and whether it can be achieved at some higher \(j\). With this in mind, we used a point contact geometry that funneled the current through a short constriction, whereas wide adjacent regions provided a thermal bath for electron cooling. This allowed us to reach \(j\) an order of magnitude greater than those achievable in the standard geometry. At these \(j\), \(I-V\) characteristics near the main NP of G/hBN superlattices became similar to those near its secondary NPs (fig. S3), although they were more smeared because of Ohmic heating and, possibly, edge irregularities in the superlattice periodicity within narrow constrictions. To circumvent the latter problems and demonstrate the universality of the critical behavior at all NPs, we made constrictions from nonsuperlatticed graphene (monolayer graphene encapsulated in hBN but nonaligned). These devices also displayed a critical current behavior, although peaks at \(j_c\) were notably broader because of heating (Fig. 3A).

To understand the criticalities, we first discuss the conceptually simplest case of the Dirac spectrum, such as in nonsuperlatticed graphene. We consistently observed that the transition between the low- and high-resistance states occurred at \(j_c \approx nev_F\) (\(e\) is the electron charge)—that is, at \(v_F \approx \sqrt{\frac{2}{m}}\), independently of \(n\) (Fig. 3B). This condition means that the Fermi surface is shifted from equilibrium by the entire Fermi momentum, and all electrons in the conduction band move along \(E\) with a drift velocity of about \(v_F\) (Fig. 3C). If the spectrum were fully gapped, \(j\) could not increase any further because all available carriers already move at maximum speed. This should result in saturation of \(j\) as a function of \(V\), which is in agreement with the observations at \(j \leq j_c\). Simulations of this intraband-only transport corroborate our conclusions (Fig. 3A, dashed curves). To explain the supercritical behavior at \(j > j_c\) for a gapless spectrum \(E\) can move electrons up in energy from the valence band into the conduction band, leaving empty states (holes) behind (Fig. 3C, bottom). The extra electrons and holes created by the interband transitions allow the current to exceed \(j_c\). Accordingly, the apparent \(v_F = j/ne\) seemingly exceeds the maximum possible group velocity, \(v_F\) (because \(n\) is fixed by gate voltage, but the actual concentration of carriers increases by \(\Delta n\)). Quantitatively, the e-h production at \(j > j_c\) can be described by the Schwinger (or Zener-Klein tunneling) mechanism. It can generate interband carriers at a rate \(\approx E^{3/2}\) (13, 16), but at small biases, the production is forbidden by the Pauli exclusion principle. Above \(j_c\), the Fermi distribution is shifted sufficiently far from equilibrium so that \(E\) depletes the states near the NP, which eliminates the Pauli blocking and enables the e-h pair production (Fig. 3C).

The above analysis can also be applied to graphene superlattices. Their narrow minibands display low \(v_F\) and therefore, the onset of interband transitions is expected at small \(j\). The switching transition in our superlattices occurred at \(v_F\) typically >10 times smaller than in nonsuperlatticed graphene (fig. S5). This yields a characteristic \(v_F\) of several \(10^4\) m \(s^{-1}\), which translates into minibands’ widths...
Fig. 2. Switching into the high-bias regime. (A to C) dV/dI as a function of j and n for the superlattices in Fig. 1, A to C, respectively. Bright arcs appear at the critical current. Yellow arrows indicate NPs as found with low-bias measurements (29). (D) Hall voltage (red curve) and the corresponding differential resistivity (black curve) measured at n indicated by the dashed line in (E). (E to G) Maps of dV_{xy}/dI for the superlattices in (A) to (C), respectively. B = 30 mT; T = 2 K. The black arrows indicate positions of van Hove singularities.

of ~10 meV, as expected from band structure calculations (19). For the relatively small j_c, superlattices were much less affected by heating than graphene and, accordingly, exhibited sharper transitions (Figs. 2 and 3A). The experimental I-V curves are compared with the above predictions for Schwinger-like carrier generation in Fig. 1, E to G. Good agreement was found for j_c. Notable deviations seen at highest j are expected because Δn is no longer small compared with n, the assumption used to derive the plotted dependences (29). Furthermore, j_c in graphene evolved ≪ n as expected for the Dirac spectrum (Fig. 3B). By contrast, superlattices exhibited clear deviations from the linear dependence (Figs. 2, A to C). This is attributed to the group velocity of charge carriers rapidly decreasing away from secondary NPs, dropping to zero at van Hove singularities (VHSs). If non-equilibrium carriers reside near VHSs, they move at low speed and contribute little to the current (Fig. S5C), leading to the sub-linear j_c(n), as observed experimentally.

Extending the described physics onto the Hall effect, it is straightforward to understand the sign changes in Fig. 2, D to G. With reference to Fig. 3C, interband transitions result in extra holes near the NP plus extra electrons that effectively appear at higher energies in the out-of-equilibrium Fermi distribution (Fig. 3C). For superlattices, contributions of these e-h pairs into V_{xy} do not cancel each other because of the broken e-h symmetry, which results in different masses and mobilities of the extra carriers. The effect is particularly strong upon approaching a VHS. For example, if the dominant carriers are electrons, their distribution would be shifted by E towards a VHS (Fig. S5C), and they should have heavy masses. By contrast, the reciprocal holes generated near the NP should be light (Fig. S5C). These higher-mobility holes are expected to provide a dominant contribution into the Hall signal, and therefore, dV_{xy}/dI should change its sign from electron to hole near j ≈ j_c, as observed experimentally. If the asymmetry is sufficiently strong, even V_{xy} can reverse its sign (Fig. 2D). The observed changes in the Hall effect can qualitatively be described by using the two-carrier model with different mobilities of out-of-equilibrium electrons and holes (fig. S7).

Last, we discuss the interband carrier generation at the main NP in graphene (Fig. 3), which closely mimics the Schwinger effect in quantum electrodynamics. Consequences of the Schwinger-like effect at the Dirac point are qualitatively different from those described by Zener-Klein tunneling at finite doping (29). In contrast to the latter case, there is no low-to-high resistance switching at n = 0, and dV/dI rapidly drops with increasing j, reaches a minimum, and then gradually increases (Fig. 3D). This behavior was highly reproducible for all graphene constrictions (fig. S8) but, because of self-gating and heating effects, could not be observed in the standard geometry, where I-V curves were similar to those in the literature (6). The initial drop is attributed to e-h puddles present at NPs, in which small E starts generating interband carriers along puddles’ boundaries and enhances conductivity (23). Minima in dV/dI typically occurred at j_m ≈ 0.05 mA μm^{-1} (Fig. 3D), which translates into Δn = j_m/eν_F = 3 × 10^{10} cm^{-2}, which is in agreement with the charge inhomogeneity δ found in our devices. In principle, the initial dV/dI drop could be fitted again by j^{-1/3}, but such fits were inconclusive because of the involved inhomogeneity. For higher j so that Δn ≫ δ, the Schwinger production fills graphene with a plasma of electrons and holes in equal concentrations, n_e = n_h = Δn. Because...
and holes, respectively. The red arrow illustrates e-h pair production. (D) \(dV/dI\) at the NP for a 0.6-\(\mu\m\)-wide constrictions. The arrows indicate minima.

Fig. 3. Nonlinear transport in nonsuperlatticed graphene near the Dirac point. (A) Voltage and differential resistance (red and black curves, respectively) for a constricttion of 0.4 \(\mu\m\) in width; \(n = 0.4 \times 10^{12} \text{m}^{-2}\). (Inset) Optical micrograph of the graphene device and its measurement geometry. Scale bar, 2 \(\mu\m\). The small bump at zero bias is caused by electron-electron scattering (34). Dashed curves indicate I-V characteristics calculated for the Dirac spectrum at \(j < j_c\) (29).

The vertical arrows indicate \(j \text{ with } V_{\text{th}} = 1 \times 10^{10} \text{ m s}^{-1}\). (B) Example of \(dV/dI\) maps for graphene constrictions. Red lines indicate \(j = j_{\text{th}}\). (C) Schematic of graphene’s spectrum and its occupancy in (top) equilibrium and in out-of-equilibrium for (middle) \(j > j_{\text{th}}\) and (bottom) \(j > j_c\). Blue and red circles indicate electrons and holes, respectively. The red arrow illustrates e-h pair production. (D) \(dV/dI\) to the NP for a 0.6-\(\mu\m\)-wide constrictions. The arrows indicate minima.

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REFERENCES AND NOTES


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SUPPLEMENTARY MATERIALS

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Supplementary Text

Figs. 51 to 58

References (36–40)

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Out-of-equilibrium criticalities in graphene superlattices
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Displacing the Fermi surface
Electrons that contribute to electrical conduction in a metal typically occupy high energy levels near the Fermi level. To get electrons from lower bands to join the flow, extremely large electric fields would be needed. In graphene and its superlattices, Berdyugin et al. show that small, experimentally accessible fields are sufficient to achieve this regime. The researchers discerned the signatures of this highly nonequilibrium state in transport data. —JS

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