The Curious Case of the Gate that Doesn't Work

1. **Unconventional ferroelectricity in moiré heterostructures**
   Authors: Zhiren Zheng, Qiong Ma, Zhen Bi, Sergio de la Barrera, Ming-Hao Liu, Nannan Mao, Yang Zhang, Natasha Kiper, Kenji Watanabe, Takashi Taniguchi, Jing Kong, William A. Tisdale, Ray Ashoori, Nuh Gedik, Liang Fu, Su-Yang Xu, and Pablo Jarillo-Herrero
   Nature **588**, 71–76 (2020)

2. **Giant ferroelectric polarization in a bilayer graphene heterostructure**
   Authors: Ruirui Niu, Zhuoxian Li, Xiangyan Han, Zhuangzhuang Qu, Dongdong Ding, Zhiyu Wang, Qianling Liu, Tianyao Liu, Chunrui Han, Kenji Watanabe, Takashi Taniguchi, Menghao Wu, Qi Ren, Xueyun Wang, Jiawang Hong, Jinhai Mao, Zheng Han, Kaihui Liu, Zizhao Gan, and Jianming Lu
   Nature Communications **13**, 6241 (2022)

3. **Electronic ratchet effect in a moiré system: signatures of excitonic ferroelectricity**
   Authors: Zhiren Zheng, Xueqiao Wang, Ziyan Zhu, Stephen Carr, Trithep Devakul, Sergio de la Barrera, Nisarga Paul, Zumeng Huang, Anyuan Gao, Yang Zhang, Damien Bérubé, Kathryn Natasha Evancho, Kenji Watanabe, Takashi Taniguchi, Liang Fu, Yao Wang, Su-Yang Xu, Efthimios Kaxiras, Pablo Jarillo-Herrero, and Qiong Ma
   arXiv: 2306.03922 (2023)

Recommended with a Commentary by Brian Skinner (Ohio State University) and Matthew Yankowitz (University of Washington)

Two-dimensional electron physics is being plagued by an ongoing series of high profile robberies: someone is stealing electrons that were supposed to have been injected into graphene by a gate electrode.

In the standard electrostatic gating setup, varying the value of a gate voltage allows one to inject or remove electrons from a 2D electron system and thereby change its chemical potential. In graphene (including most of its multilayer incarnations), one can infer the position of the chemical potential by measuring the electrical resistance as a function of the gate voltage, which produces a sharp peak when the chemical potential passes through the charge neutral point (CNP) (see Fig. 1a). In studies of graphene multilayers it has become common to connect the graphene system to *two* gate electrodes, one above and one below. In this setup both electrodes can inject or remove electrons from the graphene, and the CNP

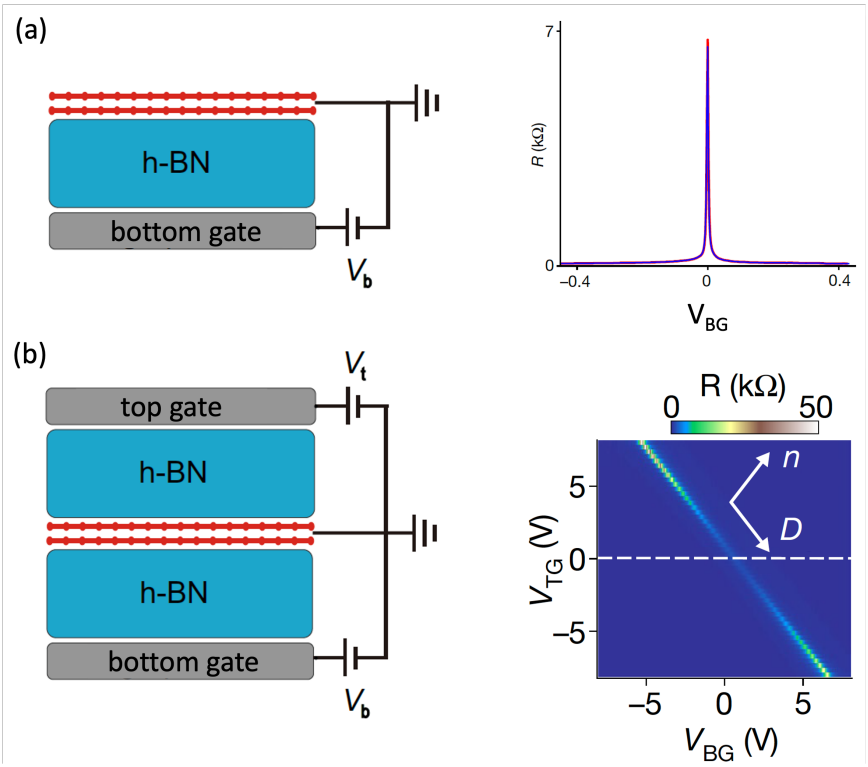

Figure 1: Electrostatic gating. (a) Single-gate setup, for which a peak in the resistance R versus gate voltage indicates the location of the CNP. (b) Dual-gate setup, for which both top and bottom gate voltages can inject electrons and the CNP occupies a diagonal line in the plane of $V_{\rm BG}$ and $V_{\rm TG}$. The figure is adapted from the first and second recommended papers.

occupies a line in the space of the top and bottom gate voltages, $V_{\rm TG}$ and $V_{\rm BG}$, as shown in Fig. 1(b). (If the two gates are equidistant from the graphene, then the CNP corresponds simply to $V_{\rm BG} + V_{\rm TG} = 0$.) The virtue of this dual-gating approach is that it allows one to independently tune the electron density n and the electric displacement field D passing through the graphene, which for multilayers enables independent coupling to the layer degree of freedom.

The dramatic effect demonstrated in the three recommended papers is that, sometimes, one of the gate electrodes mysteriously stops working. In the three recommended papers, the authors study dual-gated, Bernal-stacked bilayer graphene, with hexagonal boron nitride (hBN) used as the insulating dielectric on both sides. What they find is that, over certain voltage ranges of one of the two gate electrodes, the electrical resistance becomes independent of that gate voltage. Consider, for example, the data shown in Fig. 2(a) below, which is taken from the first recommended paper by Zheng *et al.* As the bottom gate voltage is scanned from large negative to large positive values, the position of the CNP first shifts toward more negative top gate voltages, as one would expect. But then at some value of $V_{\rm BG}$ the position of the CNP abruptly freezes in place, as if the bottom gate is no longer injecting any electrons at all into the graphene. This is what we call the "gate doesn't work" (GDW) effect. Depending on the specific device, it can be either the top gate or bottom gate that

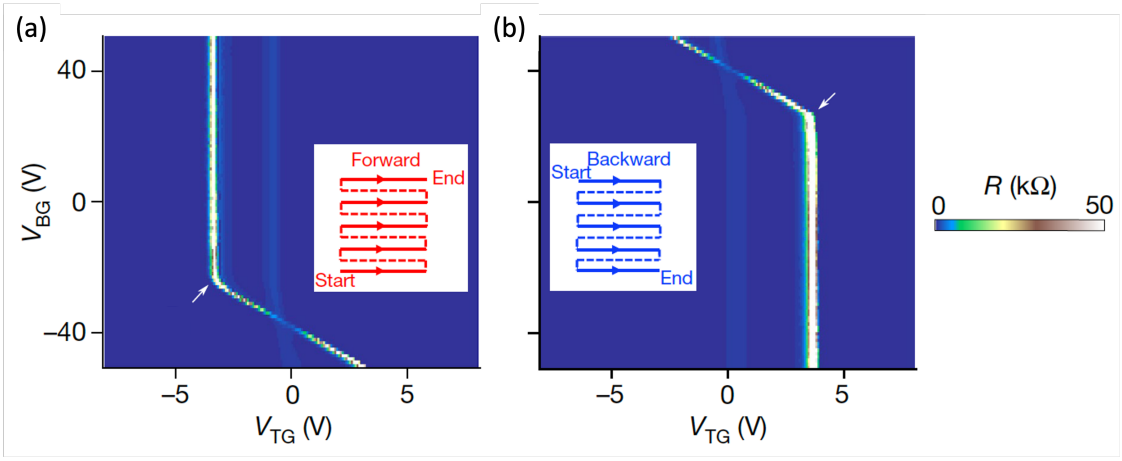

Figure 2: The Gate Doesn't Work effect in dual-gated Bernal-stacked bilayer graphene. (a) As the bottom gate voltage V_{BG} is swept from negative to positive, the CNP "get stuck" at a particular (negative) value of the top gate voltage V_{TG} denoted by the white arrow, as if further increase in V_{BG} is no longer able to inject carriers into the graphene. (b) When V_{BG} is swept from positive to negative, the CNP becomes stuck at a different, positive value of V_{TG}. Figure adapted from the first recommended paper.

becomes unexpectedly non-functional, but devices from all three recommended papers show the same general phenomenon.

Even more intriguing is that the GDW effect is strongly hysteretic. For example, Fig. 2(b) shows that if the bottom gate is instead swept from a positive value to a negative value, the CNP gets "stuck" at a positive value of V_{TG}, as opposed to the negative value that occurs when V_{BG} is swept in the opposite direction in Fig. 2(a). In other words, a system at $V_{BG} = 0$ (say) retains a "memory" of its most recent excursion toward large back gate voltage. This hysteretic behavior is described by the three papers as a type of ferroelectricity, presumed to arise from switchable layer polarization of electrons within the two graphene layers. The associated memory effect is already being considered as a possible route to building a "synaptic transistor" needed for some novel computing schemes [1].

The GDW effect is not small in magnitude. In the second recommended paper by Niu et. al., for example, the density of "missing" electrons can exceed 5×10^{12} cm^{-2}, with hysteresis surviving to above room temperature. And the memory persists for a long time. Mostly it appears to be quasi-permanent, but in some cases, as reported by the second recommended paper, the position of the CNP drifts back toward zero gate voltage over a time scale of $\sim$ hours, with a significant asymmetry between positive and negative gate histories. (The authors report that if the most recent large excursion of V_{TG} was toward the positive side, the decay time is as long as 7 hours, while if the most recent large excursion of V_{TG} was toward the negative side, the decay time is only about 10 minutes.)

The third recommended paper by Zheng *et al.* revisits the GDW effect in more detail, emphasizing the role of alignment between the graphene and the hBN. In the third recommended paper the top hBN dielectric is crystallographically aligned with the graphene, producing a long-wavelength moiré pattern. The bottom hBN, on the other hand, is intentionally misaligned by either 15° or 30°. The authors observe that the top gate (the one

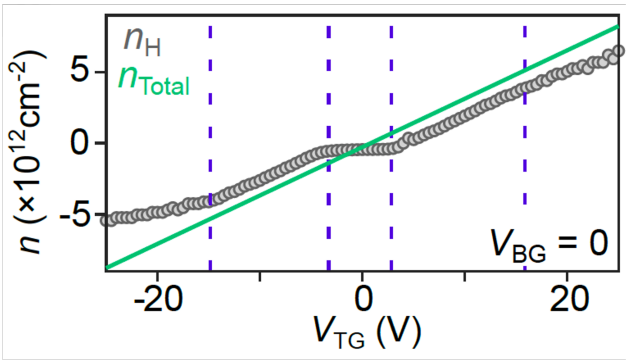

Figure 3: The disparity between the injected electron density and the apparent concentration of itinerant electrons, as measured in the third recommended paper. The green line shows the linear relation between V_{TG} and n that one would expect given the top gate capacitance, and the gray data points show values of n inferred by the Hall effect.

on the aligned side) has a range of voltage over which it "stops working". Measuring the electron concentration n by the Hall effect, for example, yields a value of n that always has fewer carriers than what should have been injected by the gate electrodes (see, e.g., Fig. 3). The authors interpret the GDW effect in terms of "localized" and "itinerant" electron states that coexist in the same bilayer graphene sample. The localized states are presumably living primarily in the moiré-influenced graphene layer. The corresponding GDW effect (which they deem an "electronic ratchet" effect) is explained in terms of interlayer excitons, in which itinerant electrons bind to localized holes (or itinerant holes bind to localized electrons).

But despite considerable effort, at this point it's fair to say that the theoretical origins of the GDW effect are not settled. All three of the recommended papers consider a bilayer graphene sample for which at least one of the two hBN layers is either presumed or known to be aligned, pointing to the potential importance of a moiré pattern. However, there are often ambiguities in determining whether an hBN flake is near 0° or 30° alignment with the bilayer graphene, as well as questions regarding the potential importance of the relative alignment between the two encapsulating hBN flakes. The second recommended paper further shows that the amount of "missing" charge can, in some cases, exceed the amount needed to fully fill the lowest moiré miniband.

There have also been other apparent instances of the GDW effect, including in magic-angle twisted bilayer graphene [2], as well as in a sample comprising two slighlty twisted monolayers of graphene separated by a monolayer of hBN and sandwiched by thick hBN spacers on either side [3]. In the latter case, there is no known alignment between any of the neighboring layers. Furthermore, despite the qualitative similarities in the GDW effect across different samples, the details can vary in rather fundamental ways. For instance, in the first recommended paper, the orientation of the hysteresis loop is opposite in two different devices. It is not yet clear what the minimal set of conditions are for deterministically achieving the GDW effect.

In closing, it is worth pointing out that a version of the GDW effect was observed in 2014 by Ref. [4] in single-gated monolayer graphene. The effect had a similar large magnitude and strong hysteretic behavior. But crucially, the effect only appeared while the sample was being illuminated by an incandescant lamp or laser light. The authors explained the

GDW effect in this case by invoking bulk defects in the hBN, from which trapped electrons can be ionized by light (photodoping). The authors provided some support for this picture by showing how the strength of the GDW effect varies from one sample to another (which seems to be common to all groups reporting it), and generally increases in magnitude when the hBN spacer is thicker (and therefore, presumably, has more defects). Subsequent work revealed that large electric fields generated by voltage pulses from an STM tip could also create local doping patterns in the graphene by ionizing hBN defects [5]. But whether bulk defects can somehow also explain a GDW effect that appears in the dark is still an open question.

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