Supplementary Information

Visualization and Control of Single Electron Charging in Bilayer Graphene Quantum Dots

Jairo Velasco Jr.[1,5,†,*], Juwon Lee[1,†], Dillon Wong[1†], Salman Kahn[1], Hsin-Zon Tsai[1], Joseph Costello[1], Torben Umeda[1], Takashi Taniguchi[4], Kenji Watanabe[4], Alex Zettl[1,2,3], Feng Wang[1,2,3] and Michael F. Crommie[1,2,3,*]

[1]*Department of Physics, University of California, Berkeley, California 94720, USA*

[2]*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

[3]*Kavli Energy NanoSciences Institute at the University of California, Berkeley and the Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

[4]*National Institute for Materials Science, 1-1 Namiki, Tsukuba, 305-0044, Japan*

[5]*Department of Physics, University of California, Santa Cruz, California 95064, USA*

[†]*These authors contribute equally to this manuscript.*

**Email: jvelasc5@ucsc.edu & crommie@berkeley.edu*

1. Tip pulsing procedure

Here we present the procedure for creating a bilayer graphene (BLG) pn junction through a tip pulse:

1. Set -1 V $\leq V_S \leq$ 1 V and $V_G = V_G^*$ under closed loop conditions, where V_G^* is the desired shift of the local charge neutrality point.
2. Open the STM feedback loop.
3. Withdraw the STM tip by $0.5 \leq \Delta z \leq 2.5$ nm. This tip pulse procedure also works in the tunneling regime (i.e. $\Delta z = 0$ nm), but can potentially damage the graphene or tip at such close distances.
4. Increase V_S to +5 V.
5. Wait Δt seconds. The wait-time Δt depends on the tip geometry and tip-to-sample distance,

   but needs to be significantly long to fully shift the local charge neutrality point E_{CNP} at $V_G = V_G^*$ to E_{CNP} ($V_G = 0$ V). Usually, $\Delta t = 30$ sec. is sufficient for $\Delta z = 1$ nm (although Δt can be significantly shorter for $\Delta z < 1$ nm and longer for $\Delta z > 1$ nm).
6. Decrease V_S back to the original value.
7. Close the STM feedback loop.
8. Set V_G =0V. The charge neutrality point at V_G =0V is now at E_{CNP} ($V_G = -V_G^*$) before the tip pulse procedure.

2. Analysis of diamond-like features

Figure S1a shows a measurement of dI/dV_S as a function of both V_G and V_S at the point where a tip pulse (with $V_G^* = 60$ V) was applied. A bias of -100 mV < V_S < 100 mV and an

STM tunnel current of 1.0 nA was used for tunneling parameters. Notably, by using these parameters it is possible to resolve spectroscopic features within the "phonon gap" bias regime. One prominent feature in this dataset is the set of bright (i.e., high dI/dV_S signal) diagonal lines that shift in energy as a function of V_G. Four prominent lines with similar spacing appear near the middle of the plot and numerous lines that are slightly fainter and with smaller spacing and similar slope appear to the left. Another prominent feature in the data is a narrow dark horizontal band around V_S = 20 mV that shifts to V_S = -20 mV with increasing V_G. Between V_G = 36V and V_G = 45V the narrow dark band and the bright diagonal lines intersect to form diamond-like features. Figure S1b is an enlargement of a region in Fig. S1a that is indicated by a box with white dashed lines. This enhancement reveals the structure of the diamond-like features: dark interiors corresponding to low dI/dV_S signal; and ends where dI/dV_S signal transitions to increasing values. The latter features are denoted by white arrows.

The diamond-like features in Fig. S1b must be related to Coulomb diamonds that have been previously studied in double barrier quantum dot systems [1]. This assignment, however, is complicated by the fact that the electric field induced gap must also reside in this low-bias region. Still, from this low-bias data we can extract a quantity that should be related to the addition energy (E_{add}). Our procedure involves summing the absolute value of the sample bias at the top and bottom of each diamond-like feature and dividing this sum by two [1, 2]. Through this analysis we find an average energy of ~27meV from Fig. S1b. This value is consistent with the estimate for the charging energy (~ 45 meV) and level spacing (< 5 meV) discussed in the main text.

3. Cross section schematic of bilayer graphene p-n junction

Figures S2a and S2b depict the most likely confinement mechanism for the massive Dirac fermions in our circular BLG p-n junctions. Red and green parabolic bands in Fig. S2a depict the conduction and valence bands of BLG. Due to the trapped negative charges in the hBN layer (resulting from a tip pulse), a spatially varying E_{CNP} emerges (black line) and the BLG bands shift vertically from left to right. Additionally, an out-of-plane electric field from the trapped negative charges in hBN and the back gate V_G induce a suppression of states between the conduction and valence bands [3]. The solid yellow line that spans from left to right denotes the chemical potential (μ_g) of BLG.

Figure S2b depicts our experimental setup which consists of a BLG/hBN heterostructure placed on an SiO_2/Si substrate. The BLG is electrically grounded with a gold contact and a V_G is applied to the heavily doped Si and can be used to tune the μ_g of the entire BLG flake. Notably, because of the location of the μ_g in Fig. S2a, BLG is n-doped where E_{CNP} is flat and p-doped at the apex of E_{CNP}. At the boundary of these two regions (the p-n junction) BLG's μ_g lies within the bandgap. As a result, tunnel barriers emerge at the p-n junction that confine massive Dirac fermions.

Figure S1. Scanning tunneling spectroscopy (STS) of locally doped bilayer graphene (BLG) within the "phonon gap" bias regime. (a) $dI/dV_S(V_G,V_S)$ plot consisting of dI/dV_S spectra measured at the center of a locally p-doped region of the BLG. The bright diagonal lines are peaks in dI/dV_S spectra that shift in energy as a function of V_G. (b) A zoom-in of the boxed region in (a). White arrows denote the ends of diamond-like features where dI/dV_S signal begins to increase. Initial tunneling parameters for all dI/dV_S spectra shown here: V_S= 0.1 V, I=1.0nA, V_{ac}=5mV.

Figure S2. Cross section schematic of BLG p-n junction and corresponding device configuration. (a) Parabolic conduction (red) and valence (green) bands and spatially varying charge neutrality point E_{CNP} (black line) of a BLG p-n junction. A band gap is present between the conduction and valence bands because of the out-of-plane electric field from the trapped negative charges in hexagonal boron nitride (hBN) and back gate (V_G). The solid yellow line denotes the BLG chemical potential (μ_g). (b) Device schematic for BLG p-n junction from (a). BLG is supported by hBN that rests upon a SiO_2/Si substrate.

Figure S1

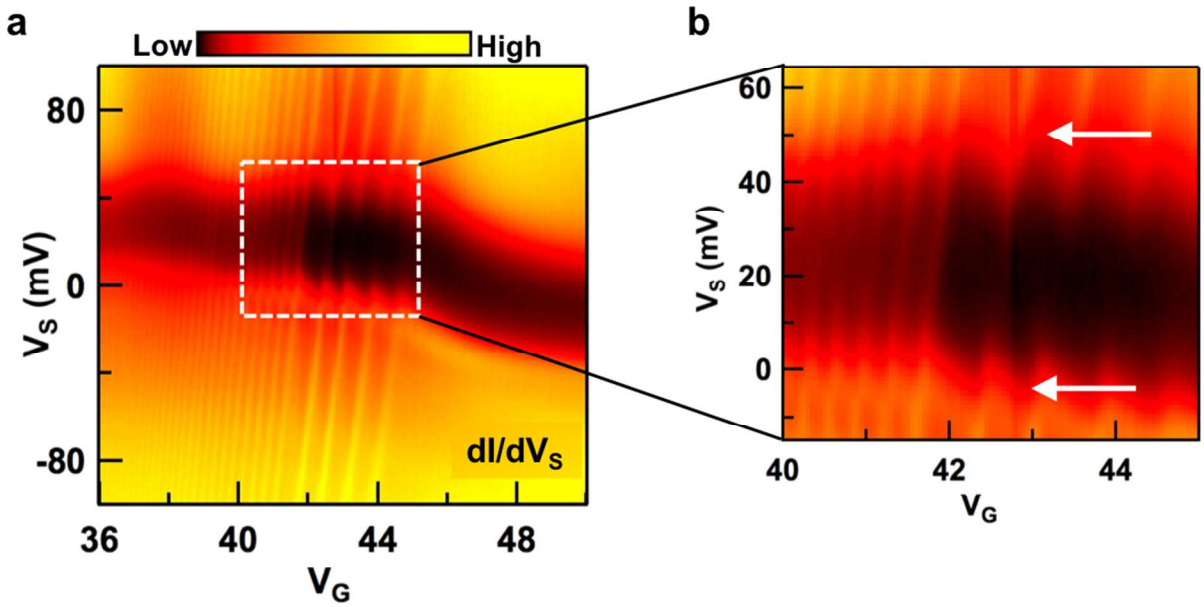
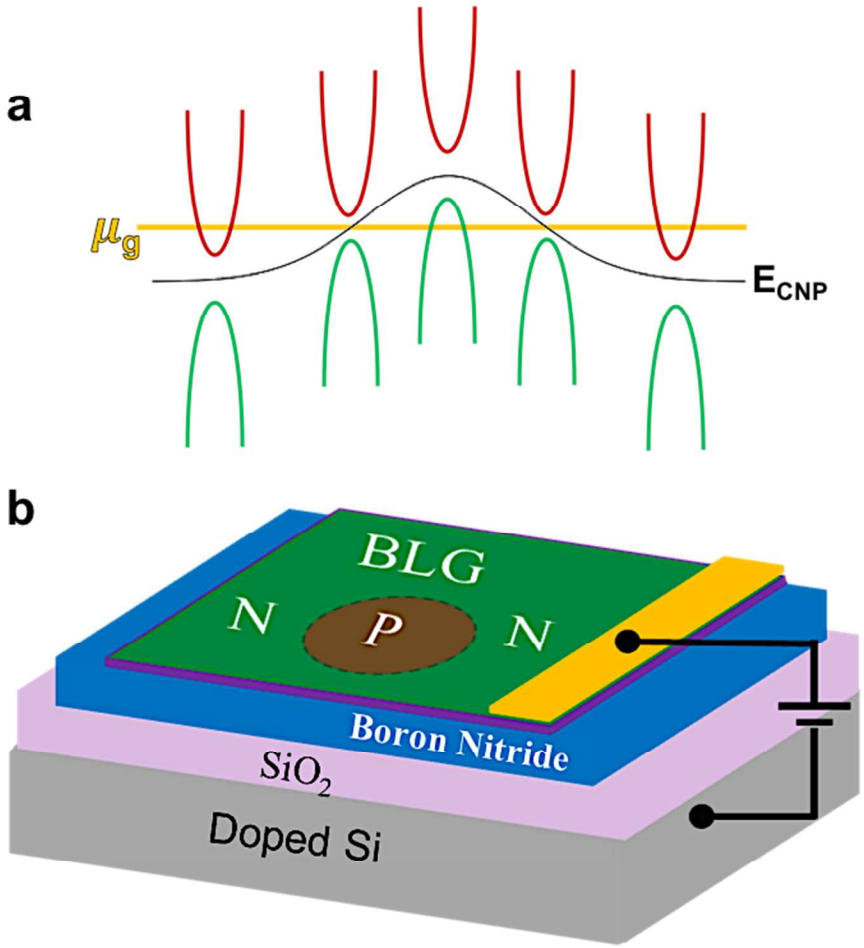

a
Low
High
80
0
-80
V_S (mV)
36
40
44
48
V_G
dI/dV_S
b
60
40
20
0
V_S (mV)
40
42
44
V_G

Figure S2



References

(1) Kouwenhoven, L. P.; Marcus, C. M.; McEuen, P. L.; Tarucha, S.; Westervelt, R. M.; Wingreen, S., *Electron Transport in Quantum Dots*. Springer: Kluwer, 1997.

(2) Park, J. Electron Transport in Single Molecule Transistor. UNIVERSITY OF CALIFORNIA, BERKELEY, 2003.

(3) McCann, E. *Phys. Rev. B* **2006,** 74, 161403.