Supporting Information

Gate-Tunable Resonant Tunneling in Double Bilayer Graphene Heterostructures

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Device fabrication

The fabrication starts with exfoliation of hBN on a silicon wafer covered with 285 nm-thick thermally grown SiO_2. Topography and thickness of the exfoliated hBN flakes are measured with atomic force microscopy (AFM), and flakes with minimum surface roughness and surface contamination are selected. On a separate silicon wafer covered with water soluble Polyvinyl Alchohol (PVA) and Poly(Methyl Methacrylate) (PMMA), bilayer graphene is mechanically exfoliated from natural graphite and identified using optical contrast and Raman spectroscopy. The PVA is dissolved in water, and the PMMA/bilayer graphene stack is transferred onto hBN flake using a thin glass slide. The PMMA film is then dissolved in acetone and the bilayer graphene is trimmed using EBL and O_2 plasma etching. Similarly, a thin hBN (t_{hBN} = 1.2-1.8 nm) flake exfoliated on a PMMA/PVA/Si substrate is transferred onto the existing bilayer graphene. A second bilayer graphene is transferred onto the stack, and trimmed on top of the bottom bilayer graphene using EBL and O_2 plasma etching. Finally, metal contacts to both top and bottom bilayer graphene are defined through EBL, electron-beam evaporation of Ni and Au, and lift-off.

Device #2 is fabricated using the dry transfer method described in ref. [S1]. The device fabrication starts with mechanical exfoliation of bilayer graphene and hBN on SiO_2/Si substrate. Then, we spin coat poly-propylene carbonate (PPC) on a 1 mm-thick Polydimethylsiloxane (PDMS) film bonded to a thin glass slide. The glass/PDMS/PPC stack is used to pick up the top bilayer graphene, the thin interlayer hBN (t_{hBN} = 1.2 nm), and the bottom bilayer graphene consecutively from SiO_2/Si substrates using the Van der Waals force between the two-dimensional crystals. The entire stack is transferred onto an hBN flake previously exfoliated on SiO_2/Si substrate. Figure 1(b) shows the transferred stack on top of bottom hBN/SiO_2/Si

substrate. After dissolving the PPC, a sequence of EBL, O_2 and CHF_3 plasma etching is used to define the active area. Finally, the metal contacts are defined by EBL, e-beam evaporation of Ti-Au, and lift-off.

Transverse electric field across the individual bilayers

The momentum-conserving tunneling between two bilayer graphene depends on their energy-momentum dispersion, and density of states. The band structure of bilayer graphene, particularly close to the CNP, can be tuned by an applied transverse electric (*E*) field, as a result of the applied V_{BG} and V_{TL}. It is therefore instructive to examine the *E*-field value for the two bilayers in a double bilayer graphene heterostructure. The general expressions for transverse *E*-field across the top (E_T) and bottom (E_B) bilayers in a double bilayer graphene device are:

$$E_B = \frac{en_B}{2\varepsilon_0} + \frac{en_T}{\varepsilon_0} + E_{B0} \quad (S1)$$

$$E_T = \frac{en_T}{2\varepsilon_0} + E_{T0} \quad (S2)$$

Here n_T and n_B are the top and bottom layer densities, respectively, and ε_0 is the vacuum permittivity. E_{T0} and E_{B0} are the transverse *E*-fields across the top and bottom bilayer at the DNP, as a result of unintentional layer doping. At a given V_{BG} and V_{TL}, the n_B and n_T values can be calculated from eqs. 1 and 2. The E_{B0} value can be calculated as following. We first determine $E_B = 0$ point, marked by minimum ρ_B along the CNL of the bottom bilayer resistivity contour plot (Fig. S1a). At $E_B = 0$, eq. 1 and S1 yield:

$$E_{B0} = \frac{C_{BG}\Delta V_{BG}}{\varepsilon_0} \quad (S3)$$

Here $\Delta V_{BG} = V_{BG-DNP} - V_{BG-E_B=0}$.

Finding the value of the E_{T0} in a back-gated double bilayer device requires an assumption about the dopant position that cause the device DNP to shift from $V_{BG} = V_{TL} = 0$ V. To

calculate the E_{T0} in our devices assume the dopants are placed on the top bilayer graphene, an assumption most plausible when the top bilayer is uncapped, as in Device #1. Equation 1 combined with the Gauss law yield:

$$E_{T0} = \frac{C_{BG} V_{BG-DNP}}{\varepsilon_0}$$

Figures S1b and S1c show the calculated E_T and E_B in Device #1 and #2 along the locus of aligned neutrality points in the two bilayers, i.e. at the tunneling resonance, as a function of V_{BG}. At the tunneling resonance E_B shows a linear dependence on V_{BG}, while E_T remains constant. For Device #1, the condition $E_T = E_B$, desirable for identical energy-momentum dispersion in the two bilayers occurs at $V_{BG} = 24\,V$, and a finite E-field. For Device #2, $E_T = E_B$ closer to zero, and at $V_{BG} = -7\,V$. Figures 4a and S1b data combined suggest the tunneling resonance in Device #1 is strongest in the vicinity of the $E_T = E_B$ point, where the band structures are closely similar for both top and bottom bilayers. The tunneling resonance in Device #2 occurs over a wider range of V_{BG} where the difference between the E_T and E_B can be as large as 0.34 V/nm.

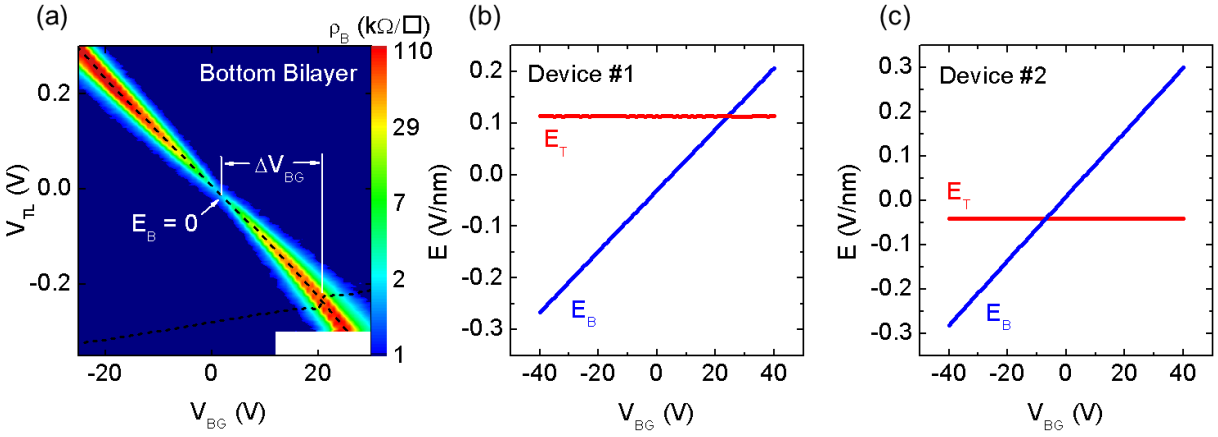

Figure S1. Transverse *E*-fields across the top and bottom bilayers. (a) Device #1 ρ_B contour plot vs. V_{BG} and V_{TL}, measured at T = 1.4 K. The CNL of the top bilayer graphene is added to

mark the DNP. E_T and E_B in (b) Device #1, and (c) Device #2, calculated at the tunneling resonance.

References

(S1) Wang, L.; Meric, I.; Huang, P. Y.; Gao, Q.; Gao, Y.; Tran, H.; Taniguchi, T.; Watanabe, K.; Campos, L. M.; Muller, D. A.; Guo, J. ; Kim, P.; Hone, J.; Shepard, K. L.; Dean, C. R. *Science*, **2013**, 342, 614.