Supporting information for
New Generation of Moiré Superlattices in Doubly Aligned
hBN/Graphene/hBN Heterostructures

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FABRICATION

We align hBN to graphene by aligning the straight edges of each layer. The probability for each alignment is 50%, because the straight edges can be along either zigzag or armchair direction. Since we need to align both hBN layers to graphene, the probability drops to 25%. We think that this is why only two out of five samples exhibit 3L-MSL features in the experiment. The hBN/G/hBN stack is directly placed on the metallic gate, so the bottom hBN acts as the dielectric layer which is usually about 20-40nm thick in our case, resulting in a high gating efficiency.

MOIRÉ PERIOD AND ORIENTATION FOR 2L-MSL

The moiré period $\lambda$ and orientation $\theta$ with respect to the graphene lattice for a graphene/hBN system, as described by Eq. (1)&(2) in the main text, are plotted as a function of the twist angle $\phi$ in Fig. S1. The green dashed lines mark the twist angles at which the orientation $\theta$ of the 2L-MSL with respect to the graphene lattice is equal to $\pm 30^\circ$ and $\pm 60^\circ$, which gives rise to the two maxima in $\lambda_3$ for $\phi_1 = -\phi_2$ due to the $60^\circ$ rotational symmetry.

FIG. S1. The moiré period $\lambda$ (black) and the orientation $\theta$ (red) as a function of the twist angle $\phi$ for the graphene/hBN MSL. The green dashed lines mark the twist angles ($\phi_1 = -\phi_2$) which give rise to the two maxima of $\lambda_3$ in Fig. 3 of the main text.
SAMPLE A

A flake with both monolayer and bilayer graphene as shown in Fig. S2(a) was chosen for sample a. We fabricated six fully encapsulated devices out of this flake, with three monolayer devices and three bilayer devices. The different device geometries are designed for other experiments. The device discussed in the main text is device a2 in Fig. S2(b). In the following we show the gate traces of all devices and the Landau fan diagram of monolayer device a3. Unfortunately, we do not have the complete data set for other devices due to a gate leak that appeared during the measurements.

FIG. S2. (a) Micrograph of the graphene flake used for sample a. (b) Micrograph of all six finished fully encapsulated devices. a1-a3 are monolayer and a4-a6 are bilayer.

In Fig. S3, two-terminal differential conductance is plotted as a function of gate voltage for all devices. Each curve is offset by an individual $V_0$ in gate voltage in order to shift the MDP to zero gate voltage. All six devices spread over 50µm show extra conductance minima in addition to the MDP at roughly the same gate voltage, suggesting an intrinsic lattice related origin of these features.
FIG. S3. Two-terminal differential conductance $G$ as a function of gate voltage $V_g$ measured at 4.2 K for all three monolayer devices (a) and all three bilayer devices (b). $V_0$ is around 250 mV for all devices. Curves are shifted by $5e^2/h$ sequentially in y direction for clarity.

Device a3

The two-terminal differential conductance of monolayer device a3 is plotted as a function of charge carrier density $n$ in Fig. S4(a). In addition to the MDP, two pairs of conductance minima occur symmetrically at $n \approx \pm 2.4 \times 10^{12} \text{cm}^{-2}$ and $n \approx \pm 1.4 \times 10^{12} \text{cm}^{-2}$, respectively, exactly the same as in device a2 in the main text. The Landau fan diagram (see Fig. S4(b)) also looks very similar as that of device a2.
FIG. S4. Electronic transport of device a3 at 4.2 K. (a) Two-terminal differential conductance $G$ as a function of charge carrier density $n$. (b) $dG/dn$ as a function of $n$ and $B$ of the same device. Filling factors are indicated on top of the diagram. The dashed lines mark the indications of filling factors fanning out from higher densities.

SAMPLE B

The graphene flake used for sample b is shown in Fig. S5(a). Ten fully encapsulated devices were fabricated out of this flake as depicted in Fig. S5(b), with five monolayer devices, four bilayer devices and one trilayer device. We show the gate traces of all devices in Fig. S6 and the Landau fan diagram of one monolayer device b2 in Fig. S7. Unfortunately, the gate started to leak during the measurements as one can see for example in Fig. S7(b).
FIG. S5. (a) Micrograph of the graphene flake used for sample b. (b) Micrograph of all ten finished fully encapsulated devices. b1-b5 are monolayer, b6, b7, b9, b10 are bilayer and b8 is trilayer.

Two-terminal differential conductance of all ten devices are plotted as a function of gate voltage in Fig. S6. The additional DPs occur at slightly different gate voltages for different devices. One reason for that might be the gate leak, resulting in different lever arms for different devices. The measuring sequence is the same as the labelling, with device b1 measured first and device b10 measured last. The second reason might be a tiny relative rotation of any of the three layers at different locations due to bubbles or ripples formed in the stack during fabrication [1], which leads to effectively slightly different MSLs for different devices. In Fig. S6(c), it seems the 3L-MSL DP is almost absent in the trilayer device, which is expected due to the further separation of the top 2L-MSL and the bottom 2L-MSL.
FIG. S6. Two-terminal differential conductance $G$ as a function of gate voltage $V_g$ measured at 4.2 K for all five monolayer devices (a), all four bilayer devices (b) and one trilayer device (c). $V_0$ varies from 20 mV to 220 mV for different devices. Curves are shifted by $10e^2/h$ sequentially in $y$ direction for clarity.
Device b2

The two-terminal differential conductance of monolayer device b2 is plotted as a function of $n$ in Fig. S7(a). In addition to the MDP, one pair of SDPs appear symmetrically at $n \approx \pm 2.9 \times 10^{12} \text{cm}^{-2}$, resulting from a graphene/hBN MSL with $\lambda \approx 12 \text{ nm}$ and $\phi \approx 0.6^\circ$. Another SDP appears at $n \approx -0.48 \times 10^{12} \text{cm}^{-2}$. There are also filling factors fanning out from this SDP as shown in Fig. S7(b). This density corresponds to a superlattice with $\lambda \approx 29.6 \text{ nm}$, which we attribute to the 3L-MSL. With these parameters and the Fig. 3(c) of the main text, we can deduce back the parameters of the other graphene/hBN MSL to be $\phi \approx 1.1^\circ$ or $-1.1^\circ$ and $\lambda \approx 9.5 \text{ nm}$, corresponding to a density of $n \approx \pm 4.6 \times 10^{12} \text{cm}^{-2}$. This twist angle is in good agreement with our alignment precision.

FIG. S7. Electronic transport of device b2 at 4.2 K. (a) Two-terminal differential conductance $G$ as a function of $n$. (b) $dG/dn$ as a function of $n$ and $B$ of the same device. Filling factors are indicated on top of the diagram. The bending and instability/noise of the filling factors at higher densities is due to the gate leak.

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