Supplementary Information: Flat bands and gaps in twisted double bilayer graphene

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SI. BAND STRUCTURE AND SYMMETRIES

We have considered two different geometries of the unit cell with different symmetries. If the moiré supercell is obtained starting from an ABBA stacking with the rotation axis going through the A sites, such that at zero twist angle the inner layers are in direct stacking, then the crystal has a C_{3z} plus three C_2 symmetries (180° rotations through an axis parallel to the layers). However, starting from an ABAB stacking with the same axis of rotation, the C_2 symmetry is not present. In order to keep the C_2 symmetry in the ABAB stacking, the rotation axis should pass through the B atoms. Fig. S1 shows the two twisted cells starting from an ABAB (a) and ABBA (b). In Fig. S1a no C_2 symmetry can be found.

The bands of the two 21.8° structures (Fig. S1c) show some differences at the M point of the Brillouin zone at high energies. In the region of interest for the flat bands, i.e., at low energies, there are not perceptible differences. The crossings and anticrossings are due to the presence or absence of C_{2y} symmetry.

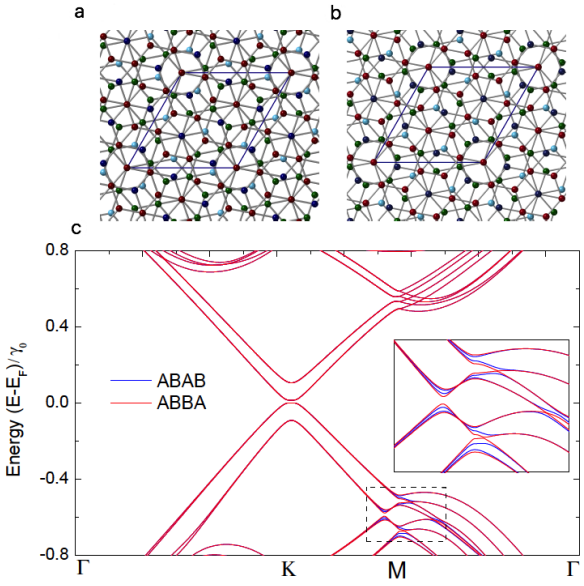

FIG. S1. Comparison of the band structures for two possible stackings (rotational angle=21.8°). (a) ABAB and (b) ABBA. The rotation axis goes through atoms AA of layers 1 and 3 in the ABAB case and through AA atoms of layers 1 and 4 in the ABBA case. (c) Band structures calculated with a TB model in both cases. The inset shows differences at high energies.

SII. FULL TB MODEL

The full TB Hamiltonian employed in the article, including the on-site terms, is an extension of a previous model [1]

$$H = \sum_i \Delta_i \left|i\right\rangle \left\langle i\right| - \sum_{i \neq j} t(\mathbf{R}_i - \mathbf{R}_j) \left|i\right\rangle \left\langle j\right|$$

where $\Delta_i = \pm$ 20 meV. The on-site term takes a positive or negative value depending on the layer, and we ultimately relate it to the charge transfer. The distance-dependent hopping parameter $t(\mathbf{d})$ is given by

$$-t(\mathbf{d}) = V_{pp\pi}(d)\left[1 - \left(\frac{\mathbf{d}\cdot\mathbf{e_z}}{d}\right)^2\right] + V_{pp\sigma}(d)\left(\frac{\mathbf{d}\cdot\mathbf{e}_z}{d}\right)^2,$$

$$V_{pp\pi}(d) = V^0_{pp\pi}\exp\left(-\frac{d-a_0}{r_0}\right),$$

$$V_{pp\sigma}(d) = V^0_{pp\sigma}\exp\left(-\frac{d-d_0}{r_0}\right),$$

where $\mathbf{d} = \mathbf{R}_i - \mathbf{R}_j$ is the distance between two atoms, $\mathbf{e}_z$ is the unit vector on the z axis, $V^0_{pp\pi} = -2.79$ eV

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is the intralayer nearest-neighbor hopping in graphene, a_0 =0.142 nm is the nearest-neighbor C-C distance, $V^0_{pp\sigma} = -0.14\, V^0_{pp\pi}$ is the interlayer nearest-neighbor hopping, and d_0 = 0.335 nm is the distance between consecutive layers. The decay length parameter r_0 is chosen to be 0.45 nm to control the value of the second nearest-neighbor hopping. Notice that the parameter ϵ_0 included in the low-energy Hamiltonian is not needed here, because the electron-hole symmetry is broken for this model.

The effects of atomic relaxations on the electronic structure have also been explored. Energy minimizations were performed with the LAMMPS [2] package using the Kolmogorov-Crespi potential [3, 4] with a cutoff of 20 Å for non-bonded interlayer interactions and the REBO potential [5, 6] for bonded intralayer interactions. During minimizations, supercell dimensions (parallel to graphene sheets) were allowed to relax and iterations stop when the sum of absolute values of force components over the entire system is smaller than 0.5 eV/Å.

SIII. SIMPLIFIED TB MODEL

To understand the origin of the gap we did several tests by varying the number of intra- and interlayer hopping parameters in the TB model. We noticed that if only one intralayer hopping γ_0 is included, then at large angles there is almost no gap, due to the fact that this approximate Hamiltonian is more symmetric. However, for low angles the gap is open within this model. The origin of the gap opening is related to a shift in energy of the bands of the twisted inner layers.

In Fig.S2 (a-c) we show the results of the band structures for three angles with only nearest-neighbor intralayer hopping γ_0. The gap is very small for large angles, i.e., above 5° but it becomes noticeable for angles below 2°. An almost perfect electron-hole symmetry can be observed, and the width of the flat bands obtained for the 1.16° is narrower at the K-M direction. Notice that in the full TB model the bands are narrower in the M-Γ direction, as mentioned in Section III of the main text.

In Fig. S2d we explicitly show the origin of the gap in this model which is the second mechanism for gap opening explained in the main text. In dashed red lines we plot two coupled inner twisted layers and two uncoupled graphene outer layers. This system is like DTBG but with uncoupled outer layers. In this case we obtain two Dirac cones, one belonging to the twisted layers, with renormalized velocity, and the other being in fact two degenerate cones with the slope as in monolayer graphene. Notice the shift in energy of the twisted Dirac cone. We also superpose in blue lines a band structure calculation for the same system, but now for an small interlayer coupling of $0.1\gamma_1$ (γ_1 is the nearest interlayer hopping in AB-stacked bilayer graphene). The size of the gap in the parabolic bands is exactly the same as the shift of the twisted Dirac cone. The other two bands are the anti-

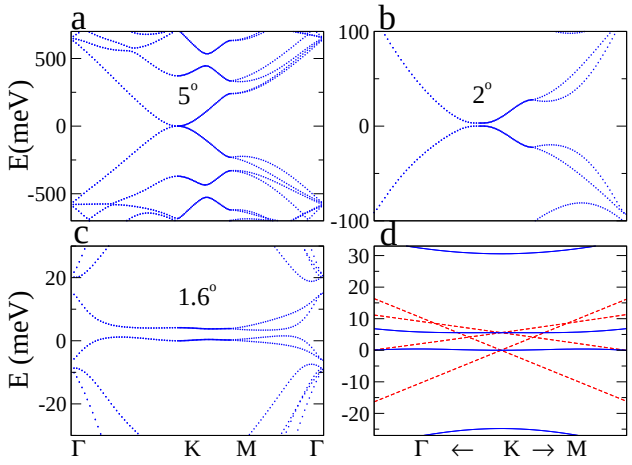

FIG. S2. (a-c) Band structures calculated with a TB model including only one intralayer term (γ_0), one direct interlayer hopping (γ_1) between AB-stacked graphene and an exponential decay hopping between atoms in the inner twisted layers. There is no gap with this model for large angles. In d we show two calculations of the bands of DTBG with a rotation angle of 2.0°. Dashed red lines: the outer Bernal layers are uncoupled to the inner ones; Continuous blue lines: outer Bernal-stacked layers coupled with a 1/10 fraction of γ_1. Notice that the size of the gap in the coupled case is exactly the shift of the Dirac cone of the twisted layers.

bonding bands of AB bilayer graphene, now located near the Fermi level due to the smaller coupling. Importantly, the size of the gap does not change with γ_1.

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