Emergent magnetic texture in driven twisted bilayer graphene

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(Dated: September 23, 2019)

The transport properties of a twisted bilayer graphene barrier are investigated for various twist angles. Remarkably, for small twist angles around the magic angle $\theta_m \sim 1.05^\circ$, the local currents around the AA-stacked regions are strongly enhanced compared to the injected electron rate. Furthermore, the total and counterflow current patterns show high correlations in these regions, giving rise to well-defined magnetic moments that form a triangular Moiré superlattice. The orientation and magnitude of these magnetic moments changes as function of the gate voltage and implications regarding the electrical control of magnetic excitations and emergent spin-liquid behaviour are discussed.

PACS numbers:

Introduction. Despite its chemical simplicity, twisted bilayer graphene (TBG) hosts a number of surprising phenomena ranging from superconductivity, correlated insulator phase, emergence of a Hofstadter butterfly, anomalous Hall ferromagnetism, photonic crystal for nano-light, and intrinsic optical dichroism. There are also several new predictions such as chiral superconductivity, nematic phases, flat plasmonic bands, a longitudinal Hall effect, long-lived plasmons and marginal Fermi liquid. The tuneable twist angle thus changes the optical, plasmonic and electronic properties that may be used in novel twisttronic devices. There are also related carbon systems such as twisted double-bilayer graphene or ABC-trilayer graphene on a BN-substrate that show superconducting phases.

The plethora of new phenomena is closely linked to the emergence of a new intermediate length scale given by the Moiré-lattice constant of $\sim 10\text{nm}$. This superstructure is due to the different crystallographic orientations of the two graphene layers and its periodicity can be defined by quasi-circular AA-stacked regions where the two graphene layers lie on top of each other. These islands are arranged in a triangular lattice, surrounded by AB- and BA-stacked graphene which are the dual configurations of Bernal-stacked bilayer graphene. At small twist angles and low energies, the wave-functions become quasi-localised within the AA-stacked regions, also leading to quasi-localised neutral collective modes.

Remarkably, a simple tight-binding model for the two bands around the charge neutrality point cannot be constructed by Wannier functions which are localised within these AA-stacked regions, mainly because they would miss out on the fragile topology inherent to TBG. Instead, the Wannier functions are centred at the AB and BA-stacked region, giving rise to effective models for the bands around the neutrality point with ring-like Coulomb interaction and assisted hopping. Also in other 2D materials such as transition metal dichalcogenides, the crystallographic misalignment gives rise to an array of equally spaced ”quantum dots” where the lattice period depends on the twist angle. The appearance of an intermediate Moiré scale should also influence the coherent transport properties, especially for filling factors around the charge neutrality point as it gives rise to the aforementioned flat-band physics. In this regime, a paramagnetic orbital response to an in-
plane magnetic field is expected that becomes maximal around the magic angle $\theta_m = 1.05^\circ$.\cite{24,25} This response is caused by the so-called counterflow\cite{26,27} where the current densities of the two layers move in opposite directions.\cite{28}

In order to invoke this response electrically, an asymmetric driving with respect to the layers is required. Therefore, we shall here investigate the coherent transport properties of TBG by injecting electrons only in one of the layers. Our main result is the emergence of an AA-enhanced counterflow pattern, interpreted as a periodic magnetic texture due to the orbital motion of the electrons whose magnitude can be tuned by the external source-drain voltage and whose triangular geometry might lead to frustration and spin-liquid behaviour.

Model. In order to investigate electronic transport through a TBG region with a large magnetic (or counterflow) response, we consider the system sketched in Fig. 1. It consists of a monolayer armchair graphene nanoribbon of width $W_{nr}$ and length $L_{nr}$ with a graphene flake of size $W_f \times L_f$ on top. When the armchair edges of the nanoribbon and the patch are aligned we have an AB-stacked bilayer graphene region. Changing the orientation of the flake, a TBG barrier is created for the electrons flowing in the nanoribbon. We choose the following dimensions: $W_{nr} = 50$ nm, $L_{nr} = 50$ nm, $W_f = 40$ nm and $L_f = 40$ nm. This setup allows us to reduce the effect of corners, edges and evanescent states that would make the interpretation of the transport mechanisms more challenging. There are 97,468 atoms in the lower layer and 59,432 atoms in the top flake. The number of atoms connected to the source and drain contacts amounts to 237. For an infinite system, the local current density and the related local magnetic moment for a positive and negative twist angle $\theta \leftrightarrow -\theta$ are related as $(j_x, j_y, j_z) \leftrightarrow (j_x, -j_y, j_z)$ and $(m_x, m_y, m_z) \leftrightarrow (-m_x, m_y, -m_z)$, respectively. These symmetry relations are fulfilled for our finite system as well, see SM.\cite{29}

Although in our setup we can impose any twist angle, we will work with commensurate rotation angles $\cos(\theta) = 1 - \frac{2M(i+1)}{i+1}$ to facilitate notation and comparison to the continuum model.\cite{30} The conductance is calculated within the Landauer-Büttiker formalism $G = 2e^2/h \text{Tr}[\Gamma_L G_C \Gamma_R G_C^\dagger]$, where $G_C = [E - H_C - \Sigma_L - \Sigma_R]^{-1}$ is the Green’s function of the central region and $\Sigma_{L(R)}/\Gamma_{L(R)}$ are the self energy/coupling of the left (right) contact, respectively.

To describe the low energetic electronic properties, we use a tight-binding Hamiltonian (above denoted as $H_C$) with hopping amplitude $t_{ij}(d_{ij}) = V_{pp}(d_{ij}) \cos^2(\alpha) + V_{ppr}(d_{ij}) \sin^2(\alpha)$ between sites $i$ and $j$, where $d_{ij} = |\vec{R}_j - \vec{R}_i|$ is the bond length and $\alpha$ is the angle formed by $\vec{d}_{ij}$ and the $z$-axis. The value of the inter-atomic matrix elements is a function of the bond length,\cite{31,32} $V_{pp} = V_0^{pp} e^{-\delta/\delta_{pp}}$, $V_{ppr} = V_0^{ppr} e^{-\delta/\delta_{ppr}}$, where $V_0^{pp} = 6.0 \pm 0.48$ eV, $V_0^{ppr} = t_0 = -2.7$ eV, $a = 0.142$ nm, $d_0 = 0.335$ nm and $\delta = 0.184\sqrt{3}a$.

Conductance. There are three electronic regimes in TBG.\cite{33} For large rotation angles $\theta > 10^\circ$, the layers are basically decoupled at low energies. For intermediate angles with $2^\circ < \theta < 10^\circ$, the two layers become coupled leading to a renormalisation of the Fermi velocity and the emergence of van Hove singularities inside the first Moiré band.\cite{34} For small angles $\theta < 2^\circ$, the layers are strongly coupled and low-energetic wave-functions become localized.\cite{35}

We can observe these regimes also in the conductance of our system where we are especially interested in the transport properties around the charge neutrality point (CNP). Due to self-doping effects, the chemical potential of the CNP is no longer at zero energy but located at $E = 0.29t_0 \sim 0.8eV$, see SM.

For large angles $\theta > 10^\circ$, the TBG barrier is completely transparent and the conductance is not modified by the twisted patch on top, see the dashed green line in Fig. 2 (a). The reduction of the twisting angle gradually increases the strength of the TBG barrier and in the intermediate coupling regime $\theta = 4.4^\circ (i=7)$, resonant peaks superimposed on the first conductance plateau as well as the reduction of the width of the plateau are observed. In the strong coupling regime $\theta = 1.2^\circ (i=27)$, the conductance quantization is completely gone and rapid oscillations around the CNP appear. In general, frequency

![FIG. 2: (Color online) Panel (a): Conductance of the twisted bilayer graphene barrier in the intermediate regime ($\theta = 4.4^\circ (i=7)$) and strong ($\theta = 1.2^\circ (i=27)$) coupling regime. The conductance of the weak coupling regime ($\theta > 10^\circ$) is represented by the green dashed line. Panel (b): Density of States for the same twist angles as in panel (a).](image-url)
as well as intensity of these oscillations become stronger as the twist angle is reduced. The physical origin behind these oscillations is the high Density of States (DOS) around the CNP, see Fig. 2 (b), i.e., the TBG barrier scatters the incident electrons into a large number of available states with the same energy, causing the observed interferences.

**Local current patterns.** We will now investigate the local current distribution through the TBG sample.\(^{121,122}\) The total current at each atomic site \(I_i\) is the sum of the bond current between sites \(i\) and \(j\), i.e., \(I_i = \frac{1}{2a} \sum_j I_{ij}(\vec{r}_i - \vec{r}_j)\) with

\[
I_{ij} = \frac{2e}{\hbar} \int_{E_F - eV_{SD}/2}^{E_F + eV_{SD}/2} t_{ij} \left[ G^<_{ij} - G^>_{ij} \right] dE ,
\]

where \(G^>_{ij}\) is the lesser Green’s function.\(^{121}\) To make contact with experiment,\(^{122}\) we fix the bias voltage \(V_{SD} = 100\ \mu\text{eV}\) and study the current distribution as a function of the gate voltage measured from the charge neutrality point \((V_g = E_F - 0.8 \ \text{eV})\).

In Fig. 3 the magnitude of the current on both layers is plotted for \(V_g \approx 0.1\ \text{meV}\), see panels (a) and (b), and for \(V_g = 7\ \text{meV}\), see panels (c) and (d). The current is normalised by the average bond current injected into the drain contact \(I_{SD}^{\text{bond}}\). For \(V_g = 0.1\text{meV}\), the total source-drain current \(I \approx 12.5\ \text{nA}\) is divided by the number of atoms connected to the drain contact, i.e., \(I_{SD}^{\text{bond}} = 12.5\ \text{nA}/237\).

The current intensity presents similar patterns and values on both layers which is remarkable given the fact that the electrons are injected only into the bottom layer. Arguably even more remarkable is the strong enhancement of the current in the AA-stacked regions which can be up to twenty times stronger compared to \(I_{SD}^{\text{bond}}\). The formation of these hot spots can be linked to the enhanced density of states observed on the AA-stacked regions. It can also be observed in the continuum model, see SM.\(^{43}\)

**In-plane magnetic moment.** Let us now discuss the current response of the driven TBG sample in magnetic language. We can define the in-plane magnetic moment in two ways, i.e., globally and locally. The first definition can be written in terms of the bond current \(I_{ij}\) between site \(i\) and \(j\), i.e., \(\vec{m} = \sum_{i<j} I_{ij}(\hat{r}_i \times \hat{r}_j)/2\) where the sum is over each pair \(i < j\).\(^{22}\) The second definition involves the magnetic current or counterflow \(\vec{I}_m = (\hat{I}_1 - \hat{I}_2)/2\) which discriminates the currents in the bottom \((\hat{I}_1)\) and top \((\hat{I}_2)\) layers and reads \(\vec{m}_L = a_{\text{eff}}(\hat{I}_m \times \hat{z})\), where \(a_{\perp} = 0.335\ \text{nm}\) is the interlayer distance, \(\hat{z}\) the out-of-plane unit vector, and \(\hat{I}_m \rightarrow \hat{I}_m(i)\) the local counterflow current at site \(i\) and \(\hat{z}\). We also introduced and effective length \(a_{\text{eff}} = 0.7a_0\), see SM.\(^{121}\) After summing the local definition over all sites, both approaches yield very similar results for the in-plane magnetic moment per site in units of Bohr magneton \(\mu_B\), as shown in Fig. 3 (f).

The above expressions are only well-defined if the total current \(\vec{I}_T = \hat{I}_1 + \hat{I}_2\) vanishes, i.e., in a closed system.\(^{22}\) This is not the case in our driven system as a net current flows through it. Still, the magnetic moment as obtained from the first definition is almost independent of the choice of the origin, see SM.\(^{121}\) This remarkable result can be understood by looking at the current map of the magnetic and total current whose absolute values differ by two orders of magnitudes, see Fig. 4 (a) and (b).

This is particularly clear in the AA-stacked regions: zooming in on one of these regions, a strongly enhanced and highly oriented counterflow is appreciated, see Fig. 4 (c). Therefore, a well-defined local magnetic moment can
be attached to each AA-patch, because \( \vec{I}_m \gg \vec{I}_T \). The presence of well-localised counterflow patterns must be accompanied by a source and drain. This can be seen from the vectorial map of the counterflow that clearly shows the presence of a source and a sink of the magnetic current on the AA-stacked region. The non-zero divergence of the magnetic current leads to a accumulation of charge on the two layers with opposite sign provoking a current in z-direction and thus closing the loop current.

It is worth noticing that large orbital magnetic moments also appear in \( C_{60} \) molecular bridges \(^{25}\) and carbon nanotubes \(^{25}\) however, its appearance highly depends on the source and drain electrodes. This is not so in our case since our electrodes are not directly attached to the TBG barrier, in fact, they are far from the barrier by approx. 5 nm. Furthermore, the continuum model (without leads) yields the same picture for the in-plane magnetic moment, i.e., large magnetic and small total currents in the AA-stacked regions. This is true for the whole band (not only around the neutrality point) and it also confirms the paramagnetic nature of the excitations,\(^7\) i.e., the current on the bottom layer is opposite to the applied source-drain voltage, see SM\(^{13}\).

**Out-of-plane magnetic moment.** The general (global) definition also yields a finite magnetic moment in z-direction. This is consistent with a local interpretation since even though \( |\vec{I}_T| \) is much smaller than \( |\vec{I}_m| \), it is finite and shows a vortex structure on the scale of the AA-stacked region as seen in Fig.\(^4\) (d). Moreover, at the atomic level we observe microscopic loop currents around the hexagonal plaquettes of single-layer graphene, giving rise to additional out-of-plane moments, see \(^{13}\). Still, this effect seems not as robust as the counterflow.

**Magnetic lattice.** Similar to a Skyrmion lattice where the spin-texture is defined by circular domain walls which are arranged in a lattice configuration and recently seen in single layer graphene,\(^{29}\) we find a non-trivial texture of angular orbital momentum which is necessarily arranged in a triangular lattice. This genuinely two-dimensional system is highly tuneable since the induced magnetic moments are directly related to the source-drain voltage and can thus be changed from the quantum regime with small magnetic moments to a "more classical" regime with larger magnetic moments. Furthermore, close to the neutrality point there is no uniform direction of the magnetic moments in the AA-stacked regions due to the scattering of evanescent modes and incommensurable twist angles should add to the misalignment. This might favour a frustrated spin-liquid state\(^{25}\) induced by the dipolar interaction between two localised magnetic moments with its characteristic \( r^{-3} \) decay.

The dipolar interactions can directly be tuned by the twist angle which changes the lattice constant \( L_M \sim \theta^{-1} \) of the triangular Moiré-lattice, i.e., for \( 1^\circ \lesssim \theta \lesssim 2^\circ \) they would differ by a factor of \( 2^3 \sim 10 \) assuming constant localised magnetic moments adjustable by the source-drain voltage. It may alternatively lead to a magnon band with possibly topologically protected edge states.

We also envision the possibility of electrical control of magnetic excitations, a long-sought goal in the quest of improved heat management for current technologies based on charged carriers\(^{25}\). Our setup might also be interesting in view of nano magnetism, usually concerned with using chiral and topological excitations such as skyrmions to store information in small volumes. Here, it would be the size of a entire Moiré unit cell.

**Summary.** To conclude, we presented a transport study of a monolayer armchair ribbon with a twisted graphene flake on top. Around the neutrality point, the TBG barrier scatters electrons mainly into evanescent modes and for twist angles around the magic angle, the response is dominated by the large number of localized states on the AA-stacked regions. Interestingly, the high local DOS also gives rise to an enhanced localized counterflow when a source-drain voltage is applied to only one layer, resulting in a highly tuneable lattice of well-defined in-plane orbital magnetic moments.

In our finite sample, a high local DOS is only seen around the neutrality point, but we expect similar features to be observed for larger filling factors in macroscopic samples where the band-structure has fully developed. This is based on calculations made for the continu-
uum model where the enhanced counterflow exists over the entire first conduction and valence band, respectively.

Acknowledgments. We acknowledge interesting discussions with Nuno Peres. This work has been supported by Spain’s MINECO under Grant No. FIS2017-82260-P, FIS2015-64886-C5-5-P, and MDM-2014-0377 as well as by the CSIC Research Platform on Quantum Technologies PTI-001. DAB acknowledges financial support from FAPESP (process nos. 2015/11779-4 and 2018/07276-5), CAPES PrInt project no. 88887.310281/2018-00, CNPq process 306434/2018-0 and Mackpesquisa.

Supplemental Material

I. COHERENT TRANSPORT PROPERTIES OF TWISTED BILAYER GRAPHENE

FIG. 5: (Color online) Conductance of the twisted bilayer graphene barrier in the (a) intermediate twist angle regime for \( \theta = 9.4^\circ (i = 3) \), \( 4.4^\circ (i = 7) \) and \( 2.4^\circ (i = 13) \). (b) Small twist angle regime for \( \theta = 1.9^\circ (i = 17) \), \( 1.3^\circ (i = 24) \) and \( 1.2^\circ (i = 27) \). The green line on both panels (a-b) corresponds to the conductance of monolayer armchair graphene nanoribbon. (c) Density of States (DOS) for twist angles presented in panel (a). (d) DOS for twist angles in panel (b).

A. Conductance, momentum mismatch and charge neutrality point

This section contains a detailed discussion of the conductance and Density of States (DOS) observed in the twisted bilayer graphene (TBG) barrier. In Fig. 5(a), the conductance for the TBG barrier is shown for intermediate twist angles. The strength of the TBG barrier for \( \theta = 9.4^\circ (i = 3) \) is still very weak and the conductance lineshape is similar to the conductance for twist angles \( \theta > 10^\circ \) (green line in Fig. 5a). For \( \theta = 4.4^\circ (i = 7) \), there are resonant peaks superimposed on the first conductance plateau. There is also a reduction on the width of the same plateau. For \( \theta = 2.4^\circ (i = 13) \) the first conductance plateau is strongly reduced.

Irrespective of the rotation angle, there are two key attributes in the conductance of the TBG barrier in this twist angle regime: (i) the reduction of the twist angle introduces a continuous set of conducting states, since there are energy regions with conductance values higher than the ones obtained for the weak coupling regime. In those regions, interference is seen which is a consequence of having more than one conducting channel (ii) There is a conductance peak at \( E \sim 0.8 \) eV.
The physical origin of both attributes can be deduced from the DOS, shown in Fig. 5 (c). The first conductance feature can be understood by noticing that new conducting states rise around $E \sim 0.8$ eV when the twist angle is reduced. On the other hand, the transmission peak at $E \sim 0.8$ eV signals the position of the charge neutrality point (CNP) of the TBG barrier. This can be understood by noticing that the highly doped left contact injects electrons with a defined momentum $k_c$ which are scattered into a number of available states with momentum $k_{TBG}^x = V_g/h v_F$, where $V_g = E_F - E_{CNP}$ is the gate voltage and $E_{CNP}$ the energy at the CNP. At the CNP, the mismatch between the available momenta in the contacts ($k_c$) and the TBG barrier ($k_{TBG}^x = 0$) produces an evanescent state and partial reflection at the barrier. The constructive interference between these waves produces a transmission peak.

Notice that the momentum mismatch and $E_{CNP}$ hardly depend on the twist angle and the peaks in the conductance and DOS observed for all twist angles at $E \sim 0.8$ eV indicate the position of $E_{CNP}$. The appearance of the high DOS peak at the same energy in the small angle regime confirms that our transport analysis is correct.

In the small angle regime ($0^\circ < \theta < 2^\circ$), the conductance quantization is completely gone. We can see in Fig. 5 (b) that the conductance shows rapid oscillations around $E \sim 0.8$ eV and the frequency as well as the intensity of these oscillations increase as the angle is reduced. The TBG barrier thus again scatters incident electrons into different channels with the same energy. However, in the small angle regime there is a higher DOS around $E \sim 0.8$ eV, shown in Fig. 5 (d). Consequently, the electrons transmit through a larger number of propagating states generating more complex interference patterns.

B. Local Density of States

In Fig. 6 (a) and (b), we have plotted the local Density of States (LDOS) for the for the state at $E = 0.8$ eV and twist angle $\theta = 4.4^\circ$. The state presents all the characteristics of an evanescent state: high LDOS at the edges that decays towards the center of the TBG barrier. However, the top and bottom layer are still weakly coupled since the LDOS is not evenly distributed over both layers. Consequently, in this regime the top patch behaves as an additional channel for the transport.

In the small angle regime, the interference discussed above is also appreciated looking at LDOS. In Fig. 6 (c) and (d), the LDOS is shown for $\theta = 1.2^\circ$ at $E = 0.8$ eV. The high LDOS is unevenly distributed over the AA-stacked regions as a result of the multiple electronic paths. The lower LDOS in the regions close to the edges of the top
graphene flake are finite size effects indicating a reduction of the Moiré confinement potential. In the small angle regime, electrons thus transmit through the sample via a number of degenerate states localized on AA-stacked regions.

C. Charge neutrality of bulk TBG

For an additional confirmation of the CNP location, we calculated the band structure for bulk TBG for different twist angles. The CNP of the bulk system is located around the same value we obtained from transport calculations of our finite system, see Fig. 7.

Based on the above and the conductance calculation, we can confirm that our finite system reproduces the main features reported for bulk TBG for \( \theta > 1^\circ \). Although DOS plots of our device show oscillations due to the confinement, we clearly observe: (i) new vHs with the reduction of the twist angle, (ii) Merging of vHs for small angles and (iii) localization of the wave function on AA-stacked regions.

D. Twist angles beyond the magic angle

For \( \theta < 1^\circ \), there is no high DOS at the charge neutrality point as shown in Fig. 8(a). Still, there is a high LDOS on the AA-stacked regions. For small twist angles, the Moiré periodicity \( D = a/\sin(\theta/2) > 16.2 \) nm almost exceeds the dimension of the top layer and the few AA-stacked regions are not enough to produce a DOS peak at the CNP. From the transport point of view, the TBG efficiently scatters electrons into the available states producing interference as seen from the rapid oscillations in the conductance, see Fig. 8(b).

Regarding the main results presented in the main text, we continue observing high current density and in-plane magnetic moments on the AA-stacked regions since these effects are the result of having high LDOS on those regions. To assert the above mentioned, we plot for a TBG barrier with \( \theta = 0.81^\circ (i = 40) \), \( V_g = 0.1 \) meV and \( V_{SD} = 100 \) \( \mu \)V the magnitude of the electric current divided by the source-drain current per bond in Fig. 8(c)-(d) and the in-plane magnetic moment in panels (e)-(f) calculated by the global formula \( \vec{m} = \sum_{<ij>} I_{ij}(\hat{r}_i \times \hat{r}_j)/2 \). The maps allow us to identify that in spite of the low number of AA-stacked regions the injected current still produces charge current and in-plane magnetic moments “hot spots”. Moreover, because of the greater Moiré periodicity the in-plane magnetic moments appear totally localized on the central AA-stacked regions. The current counterflow maps (Fig. 8(g)-(h)) also show high values and preferred orientation on the same regions.

II. GLOBAL DEFINITION OF THE MAGNETIC MOMENT
FIG. 8: (Color online) DOS (a) and conductance (b) for twist angles $\theta = 0.93^\circ (i = 35)$ and $\theta = 0.81^\circ (i = 40)$. The blue line in panel (a) corresponds to the DOS for $\theta = 1.2^\circ$. Total current normalized by the total source-drain current per bond for bottom (c) and top (d) layer. (e)-(f) In-plane magnetic moment calculated by $\vec{m} = \sum_{<ij>} I_{ij} (\vec{r}_i \times \vec{r}_j)/2$ in units of Bohr magneton for bottom and top layers. (g) Magnitude of the counterflow current ($|I_m|$) normalized by the total source-drain current per bond. (h) Vectorial map of $\vec{I}_m$ over one AA-stacked region. In panels (c)-(h), the parameters are: $\theta = 0.81^\circ (i = 40)$, $V_g = 0.1$ meV and $V_{SD} = 100$ $\mu$V.

FIG. 9: Absolute value and the $x,y,z$-components of the magnetic moment as function of $V_g$ for different shift vector $\vec{R}_0 = (0,0,0)$, $\vec{R}_0 = (a_0,0,0)$, $\vec{R}_0 = (0,a_0,0)$, and $\vec{R}_0 = (0,0,a_0)$ where $a_0 = 10$ nm ($V_{SD} = 100$ $\mu$V).

The total magnetisation induced by the current density $\vec{j}$ is defined as

$$\vec{m} = \frac{1}{2} \int d\vec{r} \times \vec{j}.$$  \hspace{1cm} (2)

For our lattice model, we write the total magnetization as \cite{53,54}

$$\vec{m} = \frac{1}{2} \sum_{<ij>} I_{ij} (\vec{r}_i \times \vec{r}_j),$$  \hspace{1cm} (3)

where $I_{ij}$ is the bond current between site $i$ and $j$ and the sum is over each pair $ij$. This expression is only well-defined for a closed system at equilibrium (for generalizations, see Ref. \cite{53}, since any change of origin $\vec{r}_i \rightarrow \vec{r}_i - \vec{R}_0$ would
lead to an additional contribution to the magnetic moment:

\[ \vec{m}_{\vec{R}_0} = \frac{1}{2} \vec{R}_0 \times \vec{I}_V, \]

(4)

with \( \vec{I}_V = \sum_{<ij>} \vec{I}_{ij} \) the total current.

To find out under what circumstances the magnetic moment is well-defined, we shift the center of our infinite device (TBG barrier + contacts) to positions \((0,0,0), (a_0,0,0), (0,a_0,0)\) and \((0,0,a_0)\) where \(a_0 = 10\) nm. The resulting magnitude, \(x, y\) and \(z\)-components of the magnetic moment are presented in Fig. 9. It is noticed that \(m_x\) and \(m_y\) are well defined for low gate voltages. The differences observed can be traced back to a small, but non-zero \(I_y\).

III. DEFINITION OF LOCAL SITE CURRENTS FROM BOND CURRENTS

Running backwards from the magnetic moment definition based on bond currents, we can provide a consistent definition of site currents that, by construction, leads to the same magnetic moment. We assume our formalism to provide an unambiguous expression for the magnetic moment:

\[ \vec{m} = \frac{1}{2} \sum_{<ij>} \vec{I}_{ij}(\vec{r}_i \times \vec{r}_j), \]

(5)

where \(\sum_{<ij>}\) means sum over pairs (bonds), counted once.

The expression of Eq. (5) can be manipulated as follows:

\[ \vec{m} = \frac{1}{2} \sum_i \sum_j \vec{I}_{ij}(\vec{r}_i \times (\vec{r}_j - \vec{r}_i))/2, \]

(6)

where we have used that \(I_{ij} = -I_{ji}, \vec{I}_{ij} \vec{r}_i \times \vec{r}_j = I_{ji} \vec{r}_j \times \vec{r}_i, \vec{r}_i \times \vec{r}_j = \vec{r}_i \times (\vec{r}_j - \vec{r}_i),\) and \(\sum_{<ij>} = \frac{1}{2} \sum_i \sum_j\), where now \(\sum_i\) (and \(\sum_j\)) runs over all sites. This leads to the following definition of site currents

\[ \vec{I}_i = \frac{1}{2a_{cc}} \sum_j \vec{I}_{ij}(\vec{r}_j - \vec{r}_i), \]

(7)

and the associated expression for the magnetic moment

\[ \vec{m} = \frac{a_{cc}}{2} \sum_i \vec{r}_i \times \vec{I}_i. \]

(8)

By construction, both expressions for the magnetic moment give the same answer, of course provided that local currents are associated to both sites of each bond. Notice that, if one just wanted the same magnetic moment, the carbon-carbon distance \(a_{cc} = 0.142\)nm could be any number, provided it is the same object in Eq. (7) and Eq. (8).

The choice of a length for \(a_{cc}\) is thus arbitrary and included for dimensional homogeneity of \(I_{ij}\) and \(I_i\). Eq. (8) can be interpreted as the discrete version of the textbook formula, \(\vec{m} = \frac{1}{2} \int dV \vec{r} \times \vec{j}\), at least for a regular array of sites.

IV. LOCAL DEFINITION OF THE MAGNETIC MOMENT AND CHIRAL RESPONSE

To check if the system size allows for a general analysis, we perform the calculations for a positive and negative twist angle. The infinite twisted bilayer system can be transformed from a positive to a negative twist angle by performing a parity-transformation \(\vec{r} \rightarrow -\vec{r}\) and subsequent mirror-transformation (\(\pi\) rotation around the \(y\)-axis). The position vector, current density, and magnetic moment transform accordingly, i.e., \((x,y,z) \rightarrow (x,-y,z), (j_x,j_y,j_z) \rightarrow (j_x,-j_y,j_z),\) and \((m_x,m_y,m_z) \rightarrow (-m_x,m_y,-m_z).\)

In Fig. 10 we can see that our finite system fulfill these requirements. Looking first at the map of in-plane magnetic moment \((V_g = 0.135\) meV and \(\theta = \pm1.2\)°) in panels (a)-(d), a large magnetic moment is seen at the AA-stacked regions.
FIG. 10: Map of the in-plane magnetic moment for $\theta = 1.2^\circ$, see panels (a) and (b), and $\theta = -1.2^\circ$, see panels (c) and (d), at gate voltage $V_g = 0.1$ meV. Components of the magnetic moment as function of $V_g$ for $\theta = 1.2^\circ$, see panel (e), and $\theta = -1.2^\circ$, see panel (f). In all panels, the magnetic moments are in units of $\mu_B$ and $V_{SD} = 100$ $\mu$V.

These regions transform as $(x,y,z) \rightarrow (x,-y,z)$ and can be linked to the high LDOS and current densities, present on the same spots. To underline the transformation of the magnetic moment, we plot the components $m_x(y)z$ in units of $\mu_B$ for two mirror-symmetric AA-stacked regions. The current pattern presents a complex structure, the number of neighbours considered in the tight-binding Hamiltonian and the interference effects introduce additional texture in the local current distribution. Despite of these circumstances, there exists a dominant current flow in all AA-stacked regions that produces an orbital magnetic moment. For this, we define the magnetic and the total current:

$$\vec{I}_m = (\vec{I}_1 - \vec{I}_2)/2$$

$$\vec{I}_T = \vec{I}_1 + \vec{I}_2$$

The magnetic current is well aligned and follows the transformation rules, see Fig. 11 (b) for $\theta = 1.2^\circ$ and in Fig. 11 (e) for $\theta = -1.2^\circ$. The local in-plane magnetic moment can now be defined as

$$\vec{m}_\parallel = a_{\text{eff}} a_\perp \vec{I}_m \times \hat{z}$$

where $a_\perp = 0.335$ nm is the interlayer distance and $\hat{z}$ the out-of-plane unit vector. We also introduced an effective bond length $a_{\text{eff}}$. Its value can be estimated projecting the bond length onto the vectors of the hexagonal lattice $a_{\text{eff}} = 2(a + a + a)/6 \approx 0.67a$. The nearly prefect match between the local and the global in-plane magnetic moment presented in the main text is achieved with $a_{\text{eff}} = 0.7a$.

Also the total current shows regular patterns. In Fig. 11 (c) and (f), the total current density is plotted where the circulating pattern is evident. As mentioned in the main text, the averaged total current is about 100 times smaller than the magnetic one, since the circulating pattern leads to a strong cancellation.

Localized magnetic moments would be correlated via dipolar interactions of the form

$$H = - \sum_{i \neq j} \frac{\mu_0}{4\pi |\vec{r}_{ij}|^3} [3(\mathbf{m}_i \cdot \vec{r}_{ij})(\mathbf{m}_j \cdot \vec{r}_{ij}) - \mathbf{m}_i \cdot \mathbf{m}_j]$$

This interaction (that is not included in our calculation) might favour the emergence of a spin-liquid.57
FIG. 11: (Color online) For $\theta = 1.2^\circ$ and $E_F = 0.8$ eV: (a) electric current density per unit energy, red arrow represents the electric current at atomic sites in the bottom layer ($\vec{I}_1$) and the blue arrow for current in the top layer ($\vec{I}_2$). (b) counterflow(magnetic) current per unit energy and (c) total current per unit energy. For $\theta = -1.2^\circ$ and $E_F = 0.8$ eV: (d) electric current density per unit energy in the bottom layer (red) and the top layer (blue). (e) counterflow(magnetic) current per unit energy and (f) total current per unit energy.

V. DEFINING THE IN-PLANE MAGNETIC MOMENT

The calculation of the magnetic and total current at the atomic sites as presented in Fig. 11 is only well-defined in the AA-stacked regions where the atoms have approximately the same $x$ and $y$ coordinates. At these sites, $\vec{I}_m(x,y) = [\vec{I}_1(x,y) - \vec{I}_2(x,y)]/2$ and $\vec{I}_T(x,y) = \vec{I}_1(x,y) + \vec{I}_2(x,y)$ are well defined. To extend the calculation to other regions of the device, it is necessary to average the current on both layers. Our process is divided in two steps.

- The current on the top layer is averaged at the center of each hexagonal plaquette. To avoid double counting of the atomic sites, we average over the centers of every third hexagonal plaquette which form a triangular lattice with lattice parameter $3a$, where $a$ is the carbon-carbon distance. To cover up the top layer, we have three different possible triangular lattices. These are identified in Fig. 12 (a) by the symbols □, ◦ and △. We used these lattices to define the current for each in the hexagonal plaquettes of the top layer as:

$$\vec{I}_2^{\square(\diamond)(\triangle)} = \sum_{s=1}^{6} \vec{I}_2(s)$$  \hspace{1cm} (12)

- The current in the bottom layer, $\vec{I}_1$, is averaged using the same triangular lattice defined for the top layer, but
FIG. 12: (Color online) (a) Real atomic lattice and the dual triangular lattices □, ⋄ and △ used to average the magnetic and total current. Vector map of $\vec{I}_m$ normalised by the total source-drain current per bond for triangular lattice (b) □, (c) ⋄ and (d) △. In panel (b) to (d) $\theta = 1.2^\circ$, $V_g = 0.1$ meV and $V_{SD} = 100$ µV.

this time we select the atomic sites within a radius $R = 1.5a$:

$$\vec{I}_1^{□(⋄)(△)} = \sum_{<1.5a} \vec{I}_1.$$

(13)

Using the above procedure the coordinates of the top and bottom current are the same and we can proceed to calculate $\vec{I}_m$ and $\vec{I}_T$.

To confirm that the results obtained do not (strongly) depend on triangular lattice used, we present the resulting magnetic current using the local definition in Fig. [12] (b) - (d). It is clearly appreciated that the enhanced counterflow current in AA-stacked regions remains a robust feature irrespective of details of the calculational method.
FIG. 13: (color online): Current map (band average) within the Moiré unit cell for twist angle $\theta = 1.05^\circ (i = 31)$, as obtained from the continuum model of Ref. 1. The current is the response to the adiabatic introduction of a uniform vector potential along the negative $x$ axis acting only on the layer 2. The current is strongly enhanced around the AA-stacked region (center of lower triangle) and minimal around the AB-stacked (corners) and BA-stacked (center of upper triangle) regions. Notice that the current of layer 2 is opposite to the field direction giving rise to a paramagnetic response.

VI. SPATIAL DISTRIBUTION OF CURRENTS IN TWISTED BILAYER GRAPHENE IN THE CONTINUUM MODEL

Here we discuss the spatial distribution of currents induced by the adiabatic introduction of a uniform vector potential along the negative $x$ axis acting only on the layer 2, within the continuum model of Lopes dos Santos et al. 1 We expect this asymmetric driving to best mimic the scattering calculation of the main text, even though the geometry differs: here the calculation corresponds to an infinite system. The twist angle is $\theta = 1.05^\circ (i = 31)$, and the calculation is standard linear response for the continuum model 27, adapted to obtain the response current at position $r$, given by

$$ j(r) = \frac{1}{2}(|r\rangle\langle r|\sigma + \sigma|r\rangle\langle r|), $$

where $\sigma$ are pseudospin (current) operators. The calculation is restricted to Fermi levels within the lowest electron and hole bands around the neutrality point. Main results are:

1) The current is largest in the AA-stacked region, and opposite in both layers with near cancellation, as expected from previous work 27,28. This is a generic property of the considered bands, as shown in Fig. 13 where the band average of currents is presented.

2) The near cancellation makes the counterflow (magnetic) current, $J_1 - J_2$, to be largely enhanced in the AA-stacked regions, as shown in Fig. 14.

3) The enhancement factor is high, particularly close to the neutrality point (in fact, infinite at the Dirac point, where the total current should vanish but the counterflow does not, but this calculation is not enough to show it). This is shown in Fig. 15.

All these features agree with the main message of this work: enhanced counterflow in AA-stacked regions close to the magic angle. It is interesting to remark that, although the total current flows in the positive $x$-direction, which coincides with the (transient) electric field as expected, the current in the layer where the field is applied (layer 2) runs opposite to the field, see right panel of Fig. 13. This fact is at the heart of the large paramagnetic response previously reported 27,28.
FIG. 14: (color online): Left panel: Counterflow current (band average). Right panel: Counterflow for a Fermi energy corresponding to the vanHove peak closest to the Dirac point.

FIG. 15: (color online): Left Panel: Color map of the enhancement factor (band average). Right Panel: Color map of the enhancement factor for a Fermi energy corresponding to the vanHove peak closest to the Dirac point.

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See Supplementary Material for more details and additional numerical results, which includes the additional Refs. [43-45].

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