

Magnetic properties of low-angle twisted bilayer graphene at three-quarters filling

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We present a theoretical investigation of the magnetic properties exhibited by twisted bilayer graphene systems with small twist angles, where the appearance of flat minibands strongly enhances electron-electron interactions effects. We consider a tight-binding Hamiltonian combined with a Hubbard mean-field interaction term and employ a real-space recursion technique to self-consistently calculate the system's local density of states. The $O(N)$ efficiency of the recursion method makes it possible to address superlattices of very large size by means of a full real-space analysis. Our procedure is validated by comparison with literature momentum-space calculations that find a magnetic phase in charge-neutral twisted bilayer graphene. We use our method to investigate the properties of small-angle twisted bilayer graphene at three-quarters filling of the conduction miniband. Our calculations indicate the emergence of a ferromagnetic phase that can be understood in terms of the Stoner mechanism, in line with recent experimental observations.

I. INTRODUCTION

Twisting two layers of graphene by a specific angle has led to the discovery of a wealth of unexpected phenomena [1, 2]. Particularly intriguing is the experimental discovery of insulating phases and superconducting states [3, 4] in small-angle twisted bilayer graphene (TBG) systems has opened new paths for the investigation of graphene systems [5–15], playing a key role in a line of investigation called *twistronics* [16], and has rapidly attracted a large and active community of researchers in condensed matter physics and materials science [2, 17].

The origin of these remarkable phenomena in low-angle TBG is attributed to the appearance of flat electronic bands near the charge neutrality point (CNT) [18, 19], as explained by the continuum model [20–23]. The corresponding electronic states are localized, leading to enhanced electron-electron interaction effects, which are believed to originate the rich properties observed in low-angle TBG. From a theoretical point of view, the challenge is to treat a system with a unit cell that has thousands of atoms with strongly interacting electrons.

The focus of our paper are the magnetic properties of low-angle TBG systems at finite doping. The motivation is a recent experiment [24] that observed the emergence of a ferromagnetic phase in a TBG with a twist angle $\theta \approx 1.16^\circ$ at 3/4 filling of the conduction miniband and the debate about the underlying mechanism responsible for this experimental finding [24, 25].

The onset of magnetism in graphene-based systems has been intensively studied, both experimentally and theoretically [26]. The latter span a wide variety of settings, such as zero-dimensional graphene nanofragments [27–29], one-dimensional graphene nanoribbons [30–35], defect-induced magnetism in bulk graphene [36, 37]. The description of magnetic moments induced by edge terminations and vacancies has been addressed by both *ab initio* calculations and the tight-binding approximation with an on-site Hubbard electron-electron interaction term. The magnetic properties of these systems are

nicely described by a mean-field theory, with the exception of vacancy-induced Kondo correlations [38–40].

In this paper, we employ a tight-binding Hamiltonian with a mean-field Hubbard on-site interaction term to compute the low-energy electronic and magnetic properties of low-angle TGBs. We use the Haydock-Heine-Kelly (HHK) recursive technique [41–44] to calculate the spin-resolved LDOS with spin $\sigma = \{\uparrow, \downarrow\}$ of TBG as a function of the bilayer twist angle. Being an order \mathcal{N} method, the HHK method makes possible to compute the Green's functions of TBG with very large primitive unit cells. The latter combined with a self-consistent mean-field calculation allows us to investigate the electron localization properties and the magnetization of TBGs at arbitrary filling factors of the flat minibands. Motivated by a recent experimental paper [24], our focus is the study of emergent ferromagnetism close to 3/4-filling at the conduction miniband of a TBG with $\theta \approx 1.16^\circ$ [24].

The paper is organized as follows. In Sec. II we review the geometric properties of commensurate TBG moiré structures, present the Hamiltonian model, and the numerical method developed for this study. In Sec. III we discuss the LDOS of low-angle TBG systems. First, we show our results for non-interacting electrons. Next, we calculate the magnetic properties of TBGs at the charge neutrality point (CNP). The excellent agreement between our results and those of Ref. [45] serves to validate our method. Finally, we investigate the magnetic properties of a TBG system with $\theta = 1.16^\circ$ at 3/4 filling and find the emergence of a ferromagnetic phase, in line with recent experiments [24]. We present our conclusion and an outline of future research in Sec. IV.

II. THEORY AND METHODS

A. TBG geometric and symmetry properties

The stacking geometry of TBGs is characterized, as standard [20, 46, 47], by the twist of one graphene layer

with respect to the other around a given site starting from the AA-stacked bilayer, forming a moiré pattern. We describe a TBG system by twisting the upper layer at an angle θ as described in Ref. [20, 21]. The primitive lattice vectors of the bottom layer are $\mathbf{a}_1^b = \sqrt{3}a_0\hat{e}_x$ and $\mathbf{a}_2^b = \sqrt{3}a_0/2(\hat{e}_x + \sqrt{3}\hat{e}_y)$ with the carbon-carbon bond length $a_0 = 1.42 \text{ \AA}$, and those of top layer as $\mathbf{a}_i^t = R(\theta)\mathbf{a}_i^b$, where $R(\theta)$ is the rotation matrix. In this study, we take the spacing between graphene layers as $d_0 = 3.35 \text{ \AA}$ [19] and do not consider lattice relaxation effects.

The lattice structure of a TBG system is periodic if the periods of the two graphene layers match, giving a finite unit cell. Hence, the periodicity condition requires the lattice translation vector $m\mathbf{a}_1^b + n\mathbf{a}_2^b$ of the bottom (unrotated) layer and $n\mathbf{a}_1^t + m\mathbf{a}_2^t$ in the top (rotated) layer, with m and n integers, to coincide. Hence, the twist angle for a commensurate structure is related to (m, n) by [48–50]

$$\theta(m, n) = \arccos\left(\frac{1}{2}\frac{m^2 + n^2 + 4mn}{m^2 + n^2 + mn}\right), \quad (1)$$

with a lattice constant

$$L = a_0\sqrt{3(m^2 + n^2 + mn)} = \frac{|m - n|\sqrt{3}a_0}{2\sin\theta/2}. \quad (2)$$

The commensurate unit cell contains $N = 4(m^2 + n^2 + mn)$ atoms.

Due to the symmetry of the honeycomb lattice, when the translation vector of the top layer is fixed to $\delta^t = 0$, a twist at an angle $\theta = 120^\circ$ transforms the AA-stacked bilayer into itself. In turn, a twist by $\theta = 60^\circ$ transforms the bilayer from AA- to AB-stacking. TBGs with θ and $-\theta$ are mirror images that share equivalent band structures [48].

Each graphene layer is constituted by two sublattices, $A^{b(\text{or } t)}$ and $B^{b(\text{or } t)}$. The commensurate structures occur in two different forms distinguished by their sublattice parities [49, 50]. Figure 1(a) shows an example of an even commensurate structure under sublattice exchange. It is characterized by having 3 three-fold symmetric positions that correspond to the stacking of the A^bA^t and B^bB^t sites and by hexagonal centers that overlap, H^bH^t . In contrast, Fig. 1(b) shows an odd commensurate structure. Here, the top and bottom coinciding sites correspond to A^bA^t at the origin, and the two remaining three-fold symmetric positions are occupied by B^b (or B^t)-sublattice sites of one layer aligned with the hexagon centers H^t (or H^b) of its neighbor layer. Therefore, the angles $60^\circ - \theta$ and $-\theta$ followed by a relative translation of the upper layer by $\delta^t = (\mathbf{a}_1^t + \mathbf{a}_2^t)/3$ form commensurate partners with unit cells of equal areas but opposite sublattice parities.

A TBG with $\theta = 30^\circ$ is a special case, since its crystal structure is its own commensurate partner and corresponds to an elementary two-dimensional quasicrystalline lattice [51, 52].

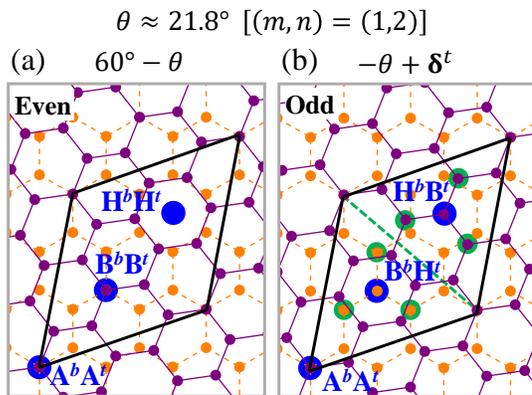


FIG. 1. Commensurate structure partners with $\theta \approx 21.8^\circ$ that corresponds $(1, 2)$. Purple (orange) circles indicate the carbon sites of the top (bottom) layer. The black lines indicate the primitive unit cell. The 3 blue disks correspond to the three-fold symmetry positions. (a) The twist angle $60^\circ - \theta$ corresponds to an even symmetry under sublattice exchange at three-fold symmetric positions. (b) The twist angle $-\theta$ followed by δ^t translation represents an odd symmetry under sublattice exchange. The green disks indicate the equivalent points. The green dashed line corresponds to the two-fold rotation axis.

Here, we work with odd commensurate structures. In Fig. 1(b), the carbon atom sites, indicated by the green disks, are equivalent due to the three-fold symmetric positions and two-fold rotation axis (green dashed line). These symmetry properties allow us to reduce the numerical computation by identifying the equivalent sites within the primitive unit cell.

B. Model Hamiltonian

We model the electronic properties of low-angle TBG systems using a tight-binding Hamiltonian with a Hubbard interaction term [26, 53], namely

$$H = H_{\text{TB}} + H_{\text{U}}, \quad (3)$$

where H_{TB} stands for the TBG tight-binding Hamiltonian and H_{U} for a Hubbard on-site electron-electron interaction term.

The tight-binding Hamiltonian that describes the low-energy electronic structure of TBG reads

$$H_{\text{TB}} = \sum_{i,j\sigma} \left(t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c.} \right), \quad (4)$$

where the operators $c_{i\sigma}$ and $c_{i\sigma}^\dagger$ annihilate and create an electron with spin $\sigma = \{\uparrow, \downarrow\}$ at site i , respectively. t_{ij} is the transfer integral between the Wannier electronic orbitals centered at the carbon sites i and j . The transfer integral t_{ij} depends on the interatomic distance and the relative orientation between p_z orbitals at each site and

is usually parameterized as [19, 48, 54]

$$t_{ij} = V_{pp\pi} \left[1 - \left(\frac{d_0}{d} \right)^2 \right] + V_{pp\sigma} \left(\frac{d_0}{d} \right)^2, \quad (5)$$

with

$$V_{pp\pi} = V_{pp\pi}^0 e^{-(d-a_0)/\delta}, \quad V_{pp\sigma} = V_{pp\sigma}^0 e^{-(d-a_0)/\delta}, \quad (6)$$

where $d = |\mathbf{r}_i - \mathbf{r}_j|^{1/2}$. Here, $V_{pp\pi}^0 = -2.7$ eV is the transfer integral between the nearest-neighbor atoms in the monolayer graphene, $V_{pp\sigma}^0 = 0.48$ eV is the interlayer transfer integral between vertically located atoms, and $\delta = 0.319a_0$ is the decay length of the transfer integral [19, 54]. The transfer integral for $d > 5 \text{ \AA}$ is exponentially small and can be safely neglected. The intra- and inter-layer hopping matrix elements t_{ij} have been determined by fitting the dispersion relations of graphene monolayer and graphene AB-stacked bilayer obtained from *ab initio* calculations [19].

The Hubbard term reads

$$H_U = U \sum_i n_{i\uparrow} n_{i\downarrow}, \quad (7)$$

where $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ is the spin-resolved electron number operator at site i . The parameter $U > 0$ gives the magnitude of the on-site Coulomb repulsion [26, 53]. The magnitude of U in graphene systems is has been extensively discussed [55, 56] with estimates that vary from $0.5 \cdots 2.0 V_{pp\pi}^0$. In this study we consider, unless otherwise stated, $U = V_{pp\pi}^0$.

We solve the TBG Hamiltonian for a given filling factor in the mean-field approximation. As standard, we write $n_{i\sigma} \equiv \langle n_{i\sigma} \rangle + \delta n_{i\sigma}$ and neglect the quadratic terms in $\delta n_{i\sigma}$ that are responsible for electronic correlations. The mean field Hubbard Hamiltonian reads

$$H_U^{\text{MF}} = U \sum_i [n_{i\uparrow} \langle n_{i\downarrow} \rangle + n_{i\downarrow} \langle n_{i\uparrow} \rangle - \langle n_{i\uparrow} \rangle \langle n_{i\downarrow} \rangle]. \quad (8)$$

The last term is a constant for a given local magnetic configuration and can be absorbed in the chemical potential.

C. Numerical method

Here, we describe how we calculate the ground state charge density and magnetic properties of TBG systems modeled by the Hamiltonian $H = H_{\text{TB}} + H_U^{\text{MF}}$.

Due to the large number of atoms in a low-angle TBG primitive cell, even mean-field calculations can be computationally very costly. Several effective Hamiltonian models have been developed to reduce the numerical effort [45, 57–59]. Here, we perform a full real-space calculation using the HHK recursive method, which is very

efficient to compute the single-particle LDOS of large systems. The HHK method [41–44] puts forward a very efficient Lanczos-like $O(\mathcal{N})$ recursive procedure that transforms an arbitrary sparse Hamiltonian matrix in a tridiagonal one. Next, it evaluates the diagonal Green's function in real space by a continued fraction expansion, which is much more numerically amenable than a full diagonalization.

By a suitable choice of the chemical potential, our approach allows us to consider charge neutral as well as systems with a finite doping, namely, $n_{\text{dop}} = N_e/A_{\text{PUC}}$, where N_e is the number of electrons in excess to the CNP and $A_{\text{PUC}} = 3\sqrt{3}a_0^2/8 \sin^2(\theta/2)$ is the area of the TBG moiré cell with N atoms.

We implement the self-consistent mean-field calculation as standard:

(i) We start with an initial set of occupation numbers $\langle n_{i\sigma} \rangle$, with the constraint $\sum_{i\sigma} \langle n_{i\sigma} \rangle = N + N_e$, where the sum runs over all sites of the moiré unit cell, set by the considered doping. The occupations can be chosen randomly or respecting some given symmetry condition.

(ii) Using the HHK recursion technique [41–44], we compute the LDOS of the electronic system defined by the Hamiltonian, Eq. (3). The spin-resolved LDOS at a given site i can be written as

$$\nu_j(\epsilon) = -\frac{1}{\pi} \lim_{\eta \rightarrow 0^+} \text{Im} G_{jj}(\epsilon + i\eta). \quad (9)$$

In practice, the regularization parameter η is considered as finite and its magnitude can be attributed the self-energy correction due to disorder, that is ubiquitous in graphene systems. The HHK method enables us to compute the LDOS with $O(\mathcal{N})$ operations and the DOS with $O(\mathcal{N}^2)$. Hence, it is much more efficient that a direct diagonalization, which demands $O(\mathcal{N}^3)$ operations.

(iii) Next, we determine $\langle n_{i\sigma} \rangle$, the average electron occupation number with spin σ at the site i . In general, at a given temperature T , $\langle n_{i\sigma} \rangle$ reads

$$\langle n_{i\sigma} \rangle = \int_{-\infty}^{\infty} d\epsilon f_\mu(\epsilon) \nu_{i\sigma}(\epsilon), \quad (10)$$

with $f_\mu(\epsilon) = \{\exp[\beta(\epsilon - \mu)] + 1\}^{-1}$, where $\beta = 1/k_B T$, k_B is Boltzmann constant, and μ is chemical potential. At zero absolute temperature, μ is equal to the Fermi energy ϵ_F and the Fermi distribution becomes a step function. For simplicity, here we consider $T = 0$. As standard, the Fermi energy ϵ_F (or the chemical potential μ) is determined by the doping.

(iv) We examine whether the output occupation numbers $\langle n_{i\sigma} \rangle$ coincide with the input ones, within a tolerance of 10^{-6} . If the answer is positive, we end the self-consistent loop. If not, we return to (ii). The computation of the updated spin densities is then repeated iteratively until all values of $\langle n_{i\sigma} \rangle$ are converged.

The self-consistent solution provides the spin polarization at the i th site,

$$p_{z,i} = \frac{\langle n_{i\uparrow} \rangle - \langle n_{i\downarrow} \rangle}{2}. \quad (11)$$

The $p_{z,i}$ is translated in a local magnetization $m_{z,i} = -g\mu_B p_{z,i}$, where μ_B is the Bohr magneton and the electron g -factor is $g = 2$. Hence, the total magnetization per moiré cell reads

$$M_{\text{PUC}} = \sum_{i=1}^N m_{z,i}. \quad (12)$$

We study two cases: (i) $N_e = 0$, corresponding to the CNP, and (ii) $N_e = 3$ corresponding to 3/4-filling of the conduction miniband [24]. For the CNP case, a previous theoretical study working in reciprocal space has predicted that low-angle TBG systems have an antiferromagnetic ground state [45]. In this case, we find computationally convenient to start the self-consistent loop with the configuration: $\langle n_{i\uparrow}^A \rangle = 1/2 + \Delta n^A$, $\langle n_{i\downarrow}^A \rangle = 1/2 - \Delta n^A$, $\langle n_{i\uparrow}^B \rangle = 1/2 + \Delta n^B$ and $\langle n_{i\downarrow}^B \rangle = 1/2 - \Delta n^B$, where $\Delta n^B = -\Delta n^A$. For the 3/4-filling case, we start with a random set of $\langle n_{i\sigma} \rangle$ with the constraint $\sum_{i\sigma} \langle n_{i\sigma} \rangle = N + N_e$, where $N_e = 3$.

III. RESULTS

We begin this section by presenting a study of the single-particle LDOS of TBG systems with a focus on the formation of low-energy minibands at small twist angles θ . Next, we use the obtained LDOS to calculate the magnetization of low-angle TBG systems using the self-consistent procedure outlined in Sec. II C. We address on two cases: (i) TBGs at the CNP and (ii) TBGs at 3/4-filling of the conduction band.

A. Non-interacting electronic densities

We compute the local electronic properties of TBGs using the HHK recursion method [41–44]. Being a real space approach, the HHK calculations take advantage of the symmetry properties of the odd commensurate TBG structures discussed above. We have numerically verified that the predicted equivalent sites indeed display the same LDOS. This is used to reduce the numerical effort by a factor of 6.

The regularization factor is set to be at most $\eta \approx 25$ meV, a value that can be attributed to a small disorder concentration. We return to this point later on.

To highlight the sites with the most prominent enhancement of the LDOS in TBGs, we consider the moiré region where the AA stacking is placed at the center of the TBG primitive unit cell. Figure 2(a) shows the LDOS for a moiré unit cell with $(m, n) = (22, 23)$ corresponding to a twist angle $\theta \approx 1.47^\circ$ and containing 6076 carbon atoms. The AB- (or BA-) stacking region is zoomed in the inset. Close to the CNP, well-localized states are found in the AA-stacking region, leading to an enhanced LDOS on atoms around the AA stacking, with

much smaller LDOS at AB- and BA-stacking regions, in agreement with previous studies [6, 19].

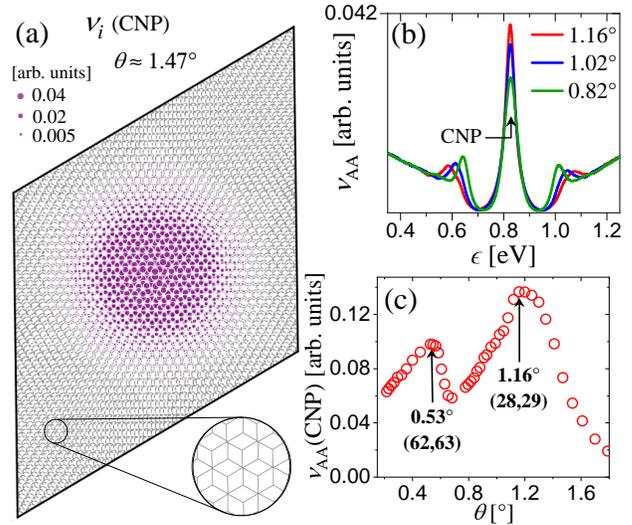


FIG. 2. Non-interacting electronic densities: (a) LDOS at the CNP, $\nu_i(\text{CNP})$ (in arbitrary units), for the $(m, n) = (22, 23)$ commensurate lattice, that corresponds to $\theta \approx 1.47^\circ$ and 6076 atoms within the moiré unit cell. The areas of the purple disks are proportional to $\nu_i(\text{CNP})$. (b) LDOS at the AA dimer sites, ν_{AA} , as a function of the energy (in electron volts) for odd commensurate lattices with $n - m = 1$. (c) ν_{AA} at the CNP as a function of θ (in degrees) corresponding to odd commensurate moiré structures with $n - m = 1$.

Figure 2(b) shows the LDOS at the AA dimer site as a function of the energy ϵ around the CNP Fermi energy for $\theta \approx 1.16^\circ$, 1.02° and 0.82° , that belong to the family of odd commensurate lattices with $n - m = 1$. The appearance of the central peak can be interpreted in terms of the continuum model [21], which associates the enhancement of the LDOS at the vicinity of the magic angle to the formation of a flat band at the CNP. Figure 2(c) displays the evolution of the AA dimer site LDOS at the CNP as a function of the small twist angles θ for the $n - m = 1$ family of odd commensurate lattice structures.

Figures 2(b) and (c) show that when the twist angle decreases, the LDOS of the AA-stacking region increases significantly at the CNP. The maximum of the peak appears at the twist angle of $\theta \approx 1.16^\circ$ corresponding to $(m, n) = (28, 29)$ with 9748 carbon atoms within the moiré cell. For the $n - m = 1$ structures considered here, this twist angle is the closest to the magic angle $\theta \approx 1.1^\circ$ predicted by the continuum model [21, 23].

As the twist angle decreases ($\theta < 1.16^\circ$), the height of the central DOS peak becomes smaller, but the satellite peaks around the main one, both in the valence and conduction bands, see Fig. 2(b), become increasingly intense, and move closer to the CNP. Eventually, these peaks begin to overlap, showing another peak maximum at the twist angle close to 0.53° , see Fig. 2(c), corresponding to $(m, n) = (62, 63)$ with 46876 carbon atoms within the

moiré cell. This behavior is in good agreement with theoretical predictions based on the continuum model [21].

We note that in Fig. 2(c) we compute $\nu_i(\epsilon)$ using $\eta = 5$ meV. This choice guarantees that η is smaller than the flat-band broadening in a magic angle twisted bilayer graphene (MATBG) that is about 10 meV [4, 21] and helps to accurately determine the peak maxima at the CNP as a function of the twist angle θ . In contrast, in Figs. 2(a)-(b) we use $\eta = 25$ meV. For this parameter choice, the flat-band broadening is greater than 10 meV, but we stress that this does not change the possibility of filling just the flat band with 4 electrons per primitive unit cell [3–5, 7], since the satellite peaks are separated from the central one by gaps of approximately 0.17 eV. Considering that the physical origin of a finite η can be attributed to disorder, the latter can be the reason for the lack of experimental observations of magic angles $\theta_n = 1.1^\circ/n$ with $n > 1$.

Let us now depart from the CNP and discuss finite doping. Figure 3(a) shows the DOS of the single-particle miniband of a TBG with $\theta \approx 1.47^\circ$. Anticipating the appearance of a gap at the CNP when the interaction is switched on, we define a valence and a conduction miniband separated at the CNP. The red area represents the half-band filling of a TBG system where the Fermi energy is at the CNP. This filling corresponds to 4 electrons per moiré cell in the valence miniband. The green area represents the electron doping at the 3/4 filling of the conduction miniband. This doping corresponds to 3 electrons per moiré cell in excess of the CNP at a Fermi energy $\epsilon_F = 872$ meV.

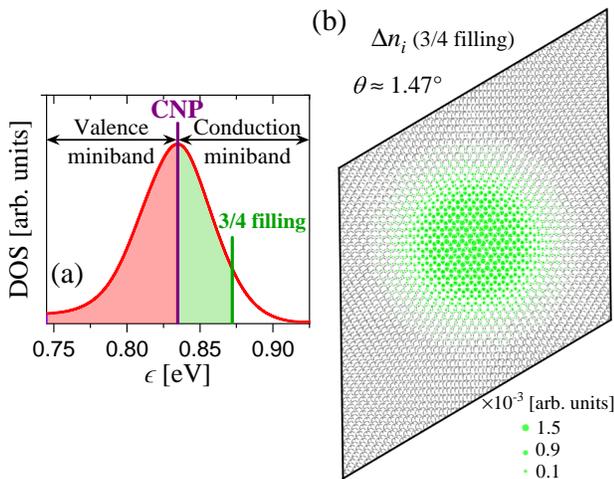


FIG. 3. Electron doping of the conduction miniband: (a) DOS (in arbitrary units) of the flat miniband as a function of the energy (in electron volts). The CNP corresponds to the DOS peak. The green area represents 3/4-filling of the conduction miniband. (b) Carrier distribution within the moiré cell (black line) in real space for $\theta \approx 1.47^\circ$. The areas of the green disks are proportional to Δn_i , the occupation in excess to the CNP filling.

Figure 3(b) shows the carrier distribution Δn_i in excess to the CNP distribution within the moiré unit cell for $\theta \approx 1.47^\circ$. The areas of the green disks are proportional to the carrier occupation number at the atomic site i , Δn_i . Owing to the localized LDOS, see Fig. 2, the carrier distribution is also concentrated at the AA-stacking region.

B. Magnetism at the CNP

Using the mean-field Hubbard Hamiltonian, we implement the self-consistent scheme described in Sec. II C to study the emergence of antiferromagnetic states in low twist angle TBGs at the CNP.

Figure 4(a) displays the local spin polarization, $p_{z,i}$, within the moiré unit cell for a $(m, n) = (22, 23)$ lattice corresponding to a rotation angle $\theta \approx 1.47^\circ$ with $U/V_{pp\pi}^0 = 1$. The regions with the largest local magnetic moments correspond to those of enhanced LDOS, see Fig. 2, strongly suggesting that the emergence of magnetism can be attributed to the Stoner mechanism [53]. In the AA stacking region, the imbalance between $\langle n_{i\uparrow} \rangle$ and $\langle n_{i\downarrow} \rangle$ leads to the emergence of an antiferromagnetic ground state at the CNP, as can be seen in the zoom. We note in passing that the converged solution is the same if the we start the self-consistent loop with a distribution of random or antiferromagnetic occupations $\langle n_{i\sigma} \rangle$.

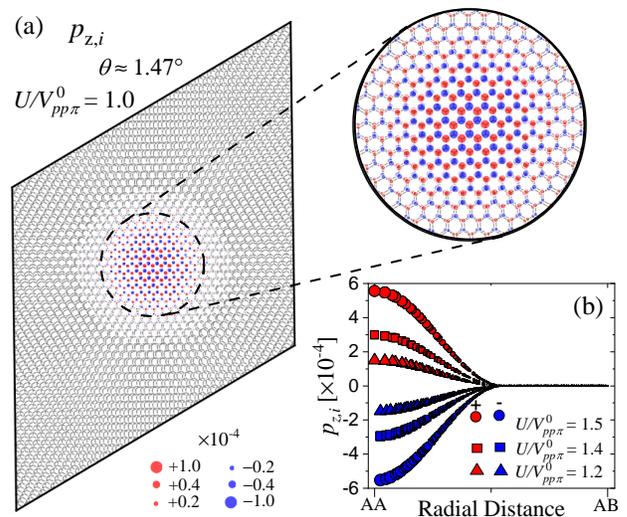


FIG. 4. Antiferromagnetic phase at the CNP: (a) Local spin polarization $p_{z,i}$ within the moiré cell (black line) for a TBG with $\theta \approx 1.47^\circ$ and $U/V_{pp\pi}^0 = 1$. The disk areas are proportional to $|p_{z,i}|$, where the red and blue disks correspond to positive and negative spin polarizations, respectively. (b) Local spin polarization $p_{z,i}$ as a function of the radial distance with respect to the AA dimer site (or AB non-dimer site) for $U/V_{pp\pi}^0 = 1.2, 1.4$ and 1.5 with $\theta \approx 1.47^\circ$.

Figure 4(b) shows the local spin polarization for a TBG system with $\theta \approx 1.47^\circ$ for different values of the on-site

Coulomb interaction, $U/V_{pp\pi}^0 = 1.2, 1.4$ and 1.5 , as a function of the radial distance to the AA dimer site. Here, the distance between the AA dimer site and the AB nondimer one is $D = 55.3 \text{ \AA}$. In line with Fig. 4(a), one observes that the spin polarization shows a maximum at the AA-stacking region, and gradually decreases and eventually vanishes for the sites that are closer to the AB stacking. The magnitude of local spin polarization becomes increasingly larger with increasing on-site Coulomb interaction U , and the overall dependence with the radial distance to the AA dimer site is similar for all U values considered. Note that since the ground state is antiferromagnetic, the total spin polarization per moiré cell is zero for all values of U . Figure 4 shows excellent agreement with the reciprocal space calculation reported in Ref. [45].

Figure 5 displays the LDOS of the flat miniband calculated for AA dimer site, namely, $\text{LDOS}(\text{AA}, \epsilon) = \nu_{\text{AA},\uparrow}(\epsilon) + \nu_{\text{AA},\downarrow}(\epsilon)$ for a twist angle of $\theta \approx 1.47^\circ$ with $U/V_{pp\pi}^0 = 1$. Due to the electron-electron interaction, the LDOS of the flat miniband in the AA dimer site is split into two peaks almost symmetric around the Fermi energy, here set to $\epsilon_F = 0$, and a small gap is opened. The separation between the two LDOS peaks is approximately 0.031 eV . Note that since the charge density is not homogeneous, one expects $\text{LDOS}(\text{AA}, \epsilon)$ to differ from the $\text{DOS}(\epsilon)$. However, since the two low-energy LDOS peaks are absent in the atomic sites at the AB-stacking region, the $\text{LDOS}(\text{AA}, \epsilon)$ captures the main features of $\text{DOS}(\epsilon)$ at low energies.

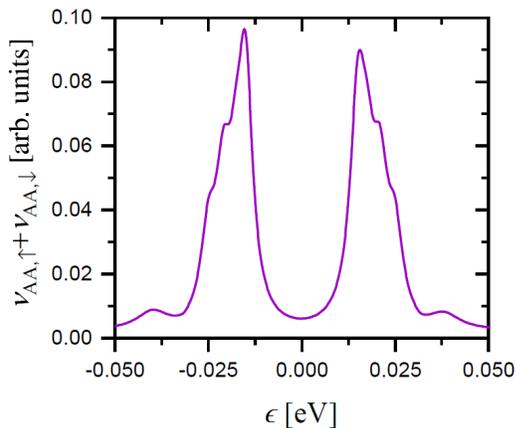


FIG. 5. LDOS at the AA dimer site, $\nu_{\text{AA},\uparrow} + \nu_{\text{AA},\downarrow}$ (in arbitrary units) as a function of energy ϵ (in eV) for $\theta \approx 1.47^\circ$ with $U/V_{pp\pi}^0 = 1$. For convenience, the Fermi energy at the CNP is set to zero.

Figure 6(a) shows the maximum spin polarization that corresponds to the AA dimer site, $p_{z,\text{AA}}$, as a function of the on-site Coulomb repulsion $U/V_{pp\pi}^0$ for different twist angles $\theta \approx 1.47^\circ, 1.30^\circ$ and 1.08° . We observe that the local spin polarization at the AA dimer site increases monotonically with increasing U . Furthermore, $p_{z,\text{AA}}$ versus $U/V_{pp\pi}^0$ shows a similar (almost linear) slope for

$\theta \approx 1.08^\circ$ and $\theta \approx 1.30^\circ$, while for $\theta \approx 1.47^\circ$ both $p_{z,\text{AA}}$ and its slope with respect to $U/V_{pp\pi}^0$ are much smaller. This suggests that $\theta \approx 1.47^\circ$ is close to a critical angle where the magnetic phase ceases to exist. A study of the critical values of $U_c/V_{pp\pi}^0$ for the zero temperature normal antiferromagnetic transition of TBG systems at the CNP can be found in Ref. [45], which estimates the critical values of $U_c/V_{pp\pi}^0$.

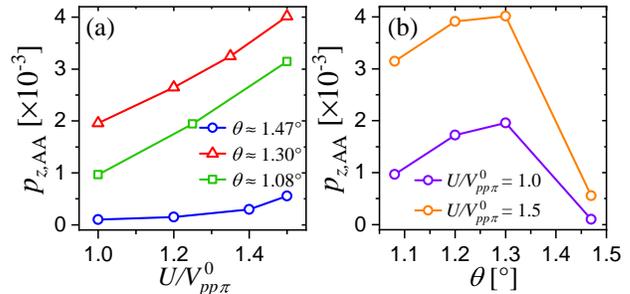


FIG. 6. (a) Local spin polarization at the AA dimer site versus $U/V_{pp\pi}^0$ for $\theta \approx 1.47^\circ, 1.30^\circ$ and 1.08° . (b) Local spin polarization at the AA dimer site as a function of the twist angle θ (in degrees) for $U/V_{pp\pi}^0 = 1.0$ and 1.5 .

Figure 6(b) shows the maximum spin polarization, $p_{z,\text{AA}}$, as a function of the magic angle θ for $U/V_{pp\pi}^0 = 1.0$ and 1.5 . We note that $p_{z,\text{AA}}$ shows a maximum close to $\theta = 1.30^\circ$ for different values of $U/V_{pp\pi}^0$, consistent with Fig. 6(a). This value of θ is slightly shifted with respect to the magic angle, $\theta_{\text{MA}} \approx 1.1^\circ$, which can be attributed to small differences in the corresponding single-particle model Hamiltonian parameterizations.

C. Magnetism at 3/4-filling of the conduction miniband

Let us now examine the ferromagnetic phase in a TBG system with a twist angle $\theta \approx 1.16^\circ$ at 3/4-filling of the conduction miniband whose origin has been the subject of discussion in the literature.

We consider an odd commensurate lattice with $(m, n) = (28, 29)$ corresponding to 9748 carbon atoms. Our results are obtained by following the self-consistent procedure described in Sec. II C for a finite doping. Here, we set $U/V_{pp\pi}^0 = 1$.

Figure 7(a) displays the local spin polarization, $p_{z,i}$, within a circular area with a radius of 21 \AA centered at the AA dimer site. These results show that the ground state is ferromagnetic as characterized by the imbalance between $\langle n_{i\uparrow} \rangle$ and $\langle n_{i\downarrow} \rangle$ in the AA stacking region. Since the spin polarization is largest at the regions of enhanced LDOS, we attribute the emergence of ferromagnetism to the Stoner instability mechanism.

Figure 7(b) shows the local spin polarization, $p_{z,i}$, as a function of the distance D from the AA dimer site. Similarly to the CNP case, the spin polarization is largest

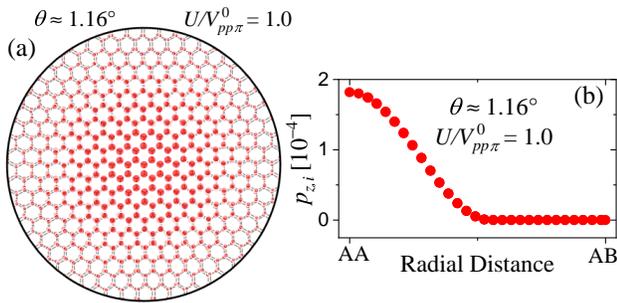


FIG. 7. Ferromagnetic phase at 3/4-filling of the conduction miniband: (a) Local spin polarization of a TBG with $\theta \approx 1.16^\circ$ and $U/V_{pp\pi}^0 = 1$ within the AA-stacking region. The areas of the red disks are proportional to the spin polarization $p_{z,i}$. (b) Local spin polarization $p_{z,i}$ versus the distance D from the i th site to the AA dimer site up to the AB nondimer site.

at the AA dimer site and becomes smaller, as D increases and eventually vanishes as the i th site approaches the AB nondimer position. On the other hand, in distinction to the previous case, since the ground state is ferromagnetic, the total magnetization is not zero. We find that the total spin polarization per moiré cell is ~ 0.19 and $M_{\text{PUC}} = -0.22 \text{ eV} \cdot \text{T}^{-1}$.

Figure 8(a) presents the LDOS calculated at the AA dimer site, $\text{LDOS}(\text{AA}, \epsilon)$, within an energy window that contains the flat minibands near the Fermi energy (set to $\epsilon_F = 0$ for clarity). Similar to the previous case, we argue that the $\text{LDOS}(\text{AA}, \epsilon)$ reproduces the behavior of $\text{DOS}(\epsilon)$ in the vicinity of ϵ_F . The separation of the two LDOS peaks around the Fermi energy is approximately 0.029 eV . The existence of two LDOS peaks in the valence miniband suggests the existence of two minibands that overlap in the CNP, as indicated in Ref. [3].

Figure 8(b) depicts the spin up and spin down LDOS at the AA dimer site, denoted as $\nu_{\text{AA},\uparrow}(\epsilon)$ and $\nu_{\text{AA},\downarrow}(\epsilon)$, respectively. A clear spin-imbalance is evident between the spin-up and spin-down occupation numbers in the filled miniband (fmb). Quantitatively, the occupation numbers are $\langle n_{\text{AA},\uparrow}^{\text{fmb}} \rangle = 9.7 \times 10^{-4}$ and $\langle n_{\text{AA},\downarrow}^{\text{fmb}} \rangle = 6.1 \times 10^{-4}$. This imbalance translates to a spin polarization $p_{z,\text{AA}} = 1.8 \times 10^{-4}$ at the AA dimer site, corresponding to a local magnetic moment $m_{z,\text{AA}} = -0.21 \text{ meV} \cdot \text{T}^{-1}$.

IV. CONCLUSIONS

In this work, we investigate the emergence of magnetism in low-angle TBG systems using a numerical real-space approach. We have considered a tight-binding Hamiltonian with a mean-field Hubbard term and obtained the ground state of TBG systems by a self-consistent iteration procedure. Notably, owing to the HHK recursive technique [41–44], our approach allows calculations to be efficiently performed for very large moiré cells.

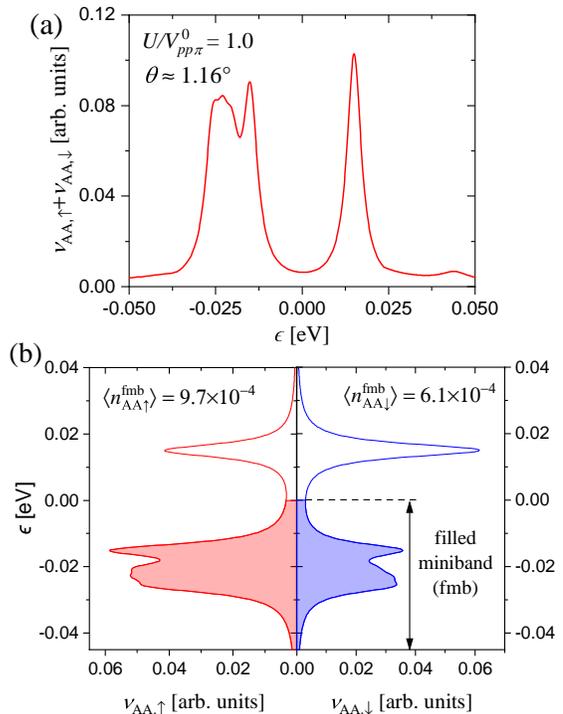


FIG. 8. (a) LDOS in the AA dimer site, $\nu_{\text{AA},\uparrow} + \nu_{\text{AA},\downarrow}$ (in arbitrary units) as a function of the energy (in eV). (b) LDOS for spin up $\nu_{\text{AA},\downarrow}$ (red line) and spin down $\nu_{\text{AA},\uparrow}$ (blue line) at the AA dimer site (in arbitrary units) as a function of the energy (in eV). Both (a) and (b) correspond to $\theta \approx 1.16^\circ$ with $U/V_{pp\pi}^0 = 1$. For convenience, the Fermi energy at 3/4-filling of the conduction miniband is set to zero.

To validate our method, we compare our results for low-angle TBGs at the CNP with those previously reported in the literature [45]. Specifically, we have found the emergence of an antiferromagnetic phase for low-angle TBGs with a maximum local spin polarization $p_{z,i}$ near the magic angle. For all computed twist angles θ , the agreement of $p_{z,i}$ with Ref. [45] is excellent, demonstrating the accuracy of our computational procedure.

The main finding of this study is the emergence of a ferromagnetic phase in low-angle TBGs at 3/4-filling of the conduction miniband. Motivated by recent experiments [24, 60] we have investigated the DOS, the local and total magnetization of TBGs with $\theta \approx 1.16^\circ$. Our calculations for the case of 3/4-filling of the conduction miniband (3 electrons in excess to the CNP per moiré cell) show that the system ground state is a ferromagnetic insulator with a gap of $\Delta \approx 0.029 \text{ eV}$, in agreement with the experimental findings [24]. We attribute this behavior to the large LDOS at the AA-stacking region, as predicted by the continuum model [21, 23], that causes a strong enhancement of the electron-electron interaction, a key element for the Stoner mechanism. Our results provide an alternative scenario to the substrate-induced gap proposed in Ref. [25].

We expect our methodology to be useful for the study

of interaction effects of other moiré 2D systems with large primitive unit cells. For instance, with modest adaptations, our approach can be used in the analysis of anisotropic ferromagnetism dominated by the orbital magnetic moment in 3/4-filling at the conduction miniband of MATBG systems [60]. By combining our calculations with a molecular dynamics procedure, we plan to analyze lattice relaxation effects as a function of twist angle. In addition, the presence of an external magnetic field, that typically requires the use of large supercells in standard approaches, in our method is trivial to account.

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