Ferromagnetism in armchair graphene nanoribbon heterostructures

P. A. Almeida,1 L. S. Sousa,1 Tome M. Schmidt,1 and G. B. Martins1

1Instituto de Física, Universidade Federal de Uberlândia, Uberlândia, Minas Gerais 38400-902, Brazil.
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We study the properties of flat-bands that appear in a heterostructure composed of strands of different widths of graphene armchair nanoribbons. One of the flat-bands is reminiscent of the one that appears in pristine armchair nanoribbons and has its origin in a quantum mechanical destructive interference effect, dubbed ‘Wannier orbital states’ by Lin et al. in Phys. Rev. B 79, 035405 (2009). The additional flat-bands found in these heterostructures, some reasonably closer to the Fermi level, seem to be generated by a similar interference process. After doing a thorough tight-binding analysis of the band structures of the different kinds of heterostructures, focusing in the properties of the flat-bands, we use Density Functional Theory to study the possibility of magnetic ground states when placing, through doping, the Fermi energy close to the different flat-bands. Our DFT results confirmed the expectation that these heterostructures, after being appropriately hole-doped, develop a ferromagnetic ground state that seems to require, as in the case of pristine armchair nanoribbons, the presence of a dispersive band crossing the flat-band. In addition, we found a remarkable agreement between the tight-binding and DFT results for the charge density distribution of the so-called Wannier orbital states.

I. INTRODUCTION

Strong correlations in magic-angle twisted bilayer graphene (TBG), discovered in 2018 [1] (see Ref. [2] for a review), were associated to the presence of strongly correlated states in flat mini-bands of the hexagonal Moiré superlattice, as previously predicted by band structure calculations [3–5]. Recently, ARPES measurements [6] have provided direct evidence for the existence of flat-bands in magic-angle TBG. These developments have greatly increased the interest in the study of low-dimensional systems presenting bands with zero (or quasi-zero) dispersion.

Indeed, in the last one year alone, there has been new flat-band research in many different areas, like their experimental observation in atomically precise one-dimensional (1D) chains [7], as well as the study of flat-bands in strongly correlated systems [8–16], search for flat-bands in kagome-type lattices [17, 18], study of symmetry aspects of flat-band systems [19–21], holographic construction of flat-bands [22], flat-bands in pyrochlore lattices [23, 24], analysis of randomness in flat-band Hamiltonians [25], topological aspects of flat-band systems [26–31], construction of flat-band tight-binding models starting from compact localized states [32], and study of flat-bands in graphene and graphene-like lattices [33–37].

For a brief review of the research in flat-bands, describing initial theoretical proposals in the late 1980s [38, 39], their association to topological phases [40, 41], and their possible realization in superconducting wire networks, cold atoms in optical lattices, and photonic systems, see Ref. [42]. For a description of strongly correlated ground states associated to dispersionless bands, see Ref. [43].

Following the development of a bottom-up procedure for atomically precise synthesis of semiconducting graphene nanoribbons (GNRs) with different width, edge, and end termination [44], a seminal paper by Steven Louie’s group in 2017 [45] showed that these synthesized armchair GNRs (AGNRs) strands belonged to different topological phases, protected by spatial symmetries and with a $\mathbb{Z}_2$ topological invariant whose value was dictated by their width and terminating unit cell. Thus, the bulk-boundary correspondence principle [46–51] imposes that at the interface between two finite AGNRs, with different $\mathbb{Z}_2$ values, a topologically protected localized state should exist, with its energy located inside the AGNR gap. This expectation was confirmed by Density Functional Theory (DFT) calculations [45]. The following year, two experimental groups, one in Europe [52] and the other in the USA [53], published side-by-side Nature papers presenting DFT and tight-binding simulations of Scanning Tunneling Spectroscopy (STS) measurements in superlattices of short AGNR strands, alternating between finite and vanishing $\mathbb{Z}_2$ values, that indicated the presence, inside the (overall) AGNR gap, of a dimerized chain band structure. A Su-Schrieffer-Heeger (SSH) effective model (initially proposed to describe polyacetylene [54], and recently revived as a prototypical model for a one-dimensional topological insulator [55]), was shown to qualitatively describe the experimental results. Thus, in what was described as a "hierarchically engineered one-dimensional topological system" [52], the AGNR heterostructure, with topologically non-trivial properties (i.e., a topologically protected end state), is itself composed of alternating topologically-trivial and non-trivial building blocks. Besides the ability of considerably decreasing the AGNR’s spectral gap (with the recent observation of metallicity in an AGNR heterostructure [56]–notice that all AGNRs are actually semiconducting [57]), the properties of these heterostructures, as implied by the results presented in Refs. [52, 53], have generated much attention, as they represent one of the first stable materials (besides polyacetylene) that simulates the SSH model, which up to now had been simulated mainly in cold-atom [58], engineered atomic lattices [59, 60], photonic [61], acoustic [62], and mechanical [63, 64] experimental configurations. Very recent work, extending the results in Refs. [52, 53], may be found in Refs. [65, 66].

A much less studied aspect of these AGNR heterostructures is the presence of dispersionless bands in their band structure. In this work, using the tight-binding method and DFT, we systematically analyze how the presence or not of flat-bands, their proximity to the Fermi energy, their interplay...
The organization of the paper is as follows: In Sec. II we introduce the AGNR heterostructure parameters \( N \), \( n \), and \( m \), together with the tight-biding Hamiltonian that models it, while in Sec. III, to illustrate the appearance of flat-bands in these heterostructures, we present the tight-binding results for heterostructures with the second smallest unit cell, i.e., \( N = 3 \), \( n = 1 \), and \( m = 3 \), showing the presence of four valence flat-bands (with respective particle-hole symmetric conduction band partners). Then, in Sec. IV, we show that flat-bands survive for ‘backbones’ \( N = 5 \) and \( N = 7 \), and also present the profile of the ‘Wannier orbital’ states associated to each one of the four lowest energy flat-bands. For \( N = 9 \), the flat-bands present for smaller values of \( N \) acquire dispersion. In Sec. V, we keep \( N = 3 \) and vary the other two parameters, \( n \) and \( m \), and analyze their influence over the flat-bands and the corresponding ‘Wannier orbital’ states (which, from now on, will be called Wannier-like states). This will set the stage for an \textit{ab-initio} DFT analysis of the ferromagnetic ground state present for varying hole-doping in Sec. VI. Finally, in Sec VIII, we present a summary of the results obtained and our concluding remarks. For completeness sake, in Sec. A of the Appendix, we present the Hamiltonian in real and reciprocal space for an N-AGNR(1,3) heterostructure (see next Section, for notation). In addition, in Secs. B and C of the Appendix, we discuss the effects of adding a next-nearest-neighbor (NNN) hopping to the main-text tight-binding calculations and briefly present tight-binding and DFT band structures for an alternative (less symmetric) type of heterostructure that has also been synthesized in the laboratory \cite{52,53}.

II. MODEL FOR THE HETEROSTRUCTURES

A. The geometry of the N-AGNR(\( n,m \)) heterostructures

In Ref. \cite{53}, two types of AGNR heterostructures were introduced, the so-called ‘inline’ and ‘staggered’ heterostructures. In this paper, we will analyze the properties just of inline heterostructures (which we will name N-AGNR(\( n,m \)) heterostructures), since they present more flat-bands than the staggered heterostructures.

In Fig. 1, we schematically show how the unit cell of an N-AGNR(\( n,m \)) heterostructure is built. In the top panel, the parameter \( n \) indicates how many adjacent unit cells (delimited by vertical dashed lines) of the so-called backbone (a pristine N-AGNR, depicted in white), containing \( N = 5 \) dimers in each unit cell, as indicated in the right, will be extended into

with nearby dispersive bands, as well as if they give origin or not to a ferromagnetic ground state, depends on the parameters that define the AGNR heterostructure. Our results show that, indeed, the majority of the heterostructures studied through tight-binding present several flat-bands that can be associated to ‘Wannier orbital’ states, as formerly seen in pristine AGNRs \cite{67}. By appropriately hole-doping these heterostructures, i.e., bringing the Fermi energy close to a flat-band, a ferromagnetic ground-state is observed through DFT simulations. The ferromagnetic exchange coupling at the flat-band appears to be mediated by a dispersive band that crosses it \cite{67}.

Before presenting the organization of the paper, we want to emphasize that we do not investigate the topological properties of the heterostructures studied here. We just, eventually, point out some possible connections between ferromagnetism and the SSH effective model, which may motivate further research on that.
unit cells containing $N + 4$ dimers. As indicated in Fig. 1, this is done, for the first of the $n$ unit cells, by adding six carbons to the top and bottom of the unit cell. This adds three extra benzene rings, colored in cyan, to the top and bottom of the unit cell. To extend the next unit cell (adjacent to the right), just four extra carbon atoms are needed to add two more benzene rings, colored in green. This second step is repeated until all $n$ adjacent unit cells are extended. The top panel in Fig. 1 shows the result for $n = 3$. Finally, in the bottom panel, $m$ indicates how many unit cells away from the last extended unit cell we will repeat the process of extending $n$ unit cells. There is an important detail here: we count $m$ from the center of the last extended unit cell to the center of the first extended unit cell of the next $n$-group to the right (notice the positioning of the vertical dashed lines in the bottom panel, see Fig. S2 in Ref. [53]). Therefore, the unit cell of the N-AGNR(n,m) heterostructure thus obtained will contain $n + m - 1$ unit cells of the original backbone. It is clear that $m \geq 2$, since $m = 1$ produces an uniform AGNR with a width equal to $N + 4$.

### B. Tight-binding Hamiltonian

The band structure of these N-AGNR(n,m) heterostructures will be simulated using a tight-binding Hamiltonian

$$H_{\text{tb}} = -t \sum_{(i,j)\sigma} c_{i\sigma}^\dagger c_{j\sigma},$$

(1)

where $c_{i\sigma}^\dagger$ creates (annihilates) an electron in site $i$ with spin $\sigma$ and $(i, j)$ runs over nearest-neighbor sites. This Hamiltonian describes nearest-neighbor hoppings with transfer integral $t$, which is a typical value found in the literature for this parameter is $t \sim 3.0$ eV [68]. In Appendix A, a specific expression will be given for Eq. (1) for a 3-AGNR(1,3) heterostructure, in real and reciprocal space.

In Sec. VI, long-range Coulomb interactions will be added within the DFT framework. A hybrid functional for the exchange-correlation term will be included in the DFT to better describe the Coulomb interactions as well as the Wannier-like states. The calculation methodology will be detailed in Sec. VI as well.

In the next section, we will present tight-binding results for the band structure of a 3-AGNR(1,3) heterostructure. Note that the tight-binding and DFT band structures will be given in units of eV.

### III. FLAT-BANDS FOR A 3-AGNR(1,3) HETEROSTRUCTURE

In Fig. 2, we show the tight-binding band structure for a 3-AGNR(1,3) heterostructure, for $t = 3.00$ eV (the nearest-neighbor hopping integral value we will use for all tight-binding calculations). For the energy-interval shown, we label the negative energy flat-bands as 1, 2, 3a, 3b, and 4, starting from the closest one to the Fermi energy (at half-filling). Their respective energies are $E_1 = -0.56$, $E_2 = -1.61$, $E_{3a} = E_{3b} = -3.00 = -t$, and $E_4 = -4.08$ eV, where the band at $-t$ is double-degenerate.

It is relatively well known [67] that N-AGNRs (pristine, with no extensions) with odd-N present two perfectly flat-bands at $\pm t$, and Fig. 2 shows that this also happens for the 3-AGNR(1,3) heterostructure (energy $E_{3a} = E_{3b} = -t$). As a matter of fact, this is true for all odd-N N-AGNR(1,3) heterostructures we have investigated, with the difference that for $N = 3, 5$ and 7 there are additional flat-bands at higher and lower energies, as shown in Fig. 2. For $N \geq 9$, these additional flat-bands acquire dispersion (see Sec. IV). One inter-

![Fig. 4. Wannier-like states for all four flat-bands in a 3-AGNR(1,3). From top to bottom, corresponding band energies are $E_1 = -0.56$, $E_2 = -1.61$, $E_{3a} = E_{3b} = -3.00$, and $E_4 = -4.08$ eV.](image)

![Fig. 5. Band structures for N-AGNR(1,3) heterostructures for $N = 5, 7, 9$ in panels (a), (b) and (c), respectively. Although it is not so apparent, for $N = 9$ the only flat-bands left is the pair $\pm t$.](image)
est point is that, in the N-AGNR(1,3) heterostructures, the ±t bands are double-degenerate for N = 3 and 5, however, this degeneracy is lifted for N ≥ 7 (see Sec. IV).

A. The Wannier-like states

In Ref. [67], a very interesting analysis is done of the magnetism of these ±t flat-bands that are present in the odd-N AGNR (without extensions, i.e., pristine AGNR). Indeed, the origin of the zero-dispersion is that the Bloch states associated to the ±t bands are formed by ‘isolated’ clusters of charge inside each unit cell (the so-called ‘Wannier orbital’ states, or Wannier-like states), which have zero overlap with the clusters in adjacent unit cells. This happens because of destructive quantum interference [67]. This phenomenon is shown in Fig. 3, which shows the integrated charge density (over all k-values) for \( E = -t \) in each site of an \( N = 3 \) pristine AGNR. Figure 3 simulates the local density of states (LDOS) an Scanning Tunneling Microscope tip would observe in case its parameters were set to capture just the \( E = -t \) states of a 3-AGNR. It is remarkable that such Wannier-like states exist in the odd-N pristine AGNRs, survive (basically unaffected) the \((n, m)\) extensions that give origin to the heterostructure. This can be seen in the LDOS (charge density) profile shown in Fig. 4(c) for state \( E_{3a} \), which shows exactly the same structure as the one in Fig. 3, with the difference that now the extended unit cell is wider, thus it accommodates four occupied dimers along the vertical direction, in contrast to the pristine 3-AGNR, where the Wannier-like state is composed of just two dimers (see Fig. 3). On the other hand, Fig. 4(d) shows the other Wannier-like state, \( E_{3b} \), that is degenerate at \( E = -t \). Interestingly, its charge profile near the edge of the extended unit cell is clearly reminiscent of the pristine 3-AGNR, while, at the center of the unit cell it is a mixture of the \( E_{3a} \) state and some charge density occupying the maximally-separated sites that are left empty by the \( E_{3a} \) state.

The interesting result shown in the other panels of Fig. 4, for the remaining three flat-bands [panels (a), (b), and (e)], is that they seem to also originate from Wannier-like states with different charge configurations [when compared to panels (c) and (d)] that also do not have overlap between adjacent unit cells. Thus, in principle, they may produce similar magnetic ground states as the one theoretically predicted for the ±t bands in pristine N-AGNRs [67], as long as these flat-bands are crossed by dispersive bands. Section VI presents a DFT analysis of this possibility.

IV. WANNIER-LIKE STATES FOR \( N ≥ 5 \)

In panels (a), (b), and (c) in Fig. 5, we see the band structure for N-AGNR(1,3) heterostructures, for \( N = 5, 7, \) and 9, respectively. Despite the fact that the complexity of the band structures increases with \( N \), we can ascertain some facts [69]:
(i) flat-band 1, seen in Fig. 2, remains perfectly flat for \( N = 5 \) and 7, although at a different energy position, while flat-band 2 has acquired a tiny dispersion; (ii) for all three values of \( N \) the \(-t\) flat-band is present. In reality, as far as we can tell, the \( \pm t \) flat-bands occur for any odd value of \( N \); (iii) for \( N = 5 \) and 7, flat-band 4 has already acquired some dispersion; (iv) likewise, for \( N \geq 9 \), except for \(-t\) flat-band, the other three flat-bands (1, 2 and 4) have acquired dispersion; (v) finally, the \(-t\) flat-band for \( N = 5 \) is still double-degenerate, while it is not anymore for \( N = 7 \). It is possible that farther from the Fermi level (\( E = 0 \), at half-filling) there are additional flat-bands (besides the \( \pm t \) ones) for \( N \geq 9 \), but we have not investigated this possibility.

In Figs. 6 and 7, we show the flat-band Wannier-like states corresponding to bands 1, 2 and 3 presented in Figs. 5(a) and 5(b), for a 5-AGNR(1,3) and a 7-AGNR(1,3) heterostructure, respectively. A careful comparison of Figs. 4, 6, and 7 shows that the Wannier-like states for the same band at different values of \( N \) are semi-quantitatively the same, indicating that the maximum \( N \) for which we can look for these interesting states is \( N = 7 \), which is an N-AGNR(n,m) heterostructure size that can be faithfully obtained in the laboratory [44, 52, 70], suggesting that the results obtained here can be tested experimentally.

As mentioned above, there is an interesting point regarding the \(-t\) flat-band Wannier-like states \( E_{3a} \) and \( E_{3b} \) as we vary \( N \) in an N-AGNR(1,3) heterostructure: they are still degenerate for \( N = 5 \), as can be seen in Fig. 6(c), where we show the combined charge density for both bands \( E_{3a} \) and \( E_{3b} \) however, for \( N = 7 \), it is not degenerate anymore. Notice that in Fig. 7 we show, in panel (c), the charge density just for the \( E_{3a} \) band, since band \( E_{3b} \) does not exist anymore. We speculate that, as may be inferred from the charge density distribution in Fig. 6(c), the Wannier-like state \( E_{3b} \) for \( N = 5 \) seems on the verge of losing its Wannier-like character. This occurs because there are only 4 sites (indicated by numbers 1 to 4, and showing perfect destructive quantum interference) preventing the existence of a continuous nearest-neighbor path that connects all unit cells with each other, which would result in a dispersive state.

V. DEPENDENCE ON PARAMETERS \( n \) AND \( m \).

A. Band structure dependence with \( n \)

In Fig. 8, we see tight-binding band-structure results for 3-AGNR(n,3), for \( n = 1 \) to 4, in panels (a) to (d), respectively. In panel (a), we repeat the results shown in Fig. 2 [for 3-AGNR(1,3)] to facilitate comparison. A trend with increasing \( n \) (size of the extended region of the heterostructure) can be clearly discerned. Indeed, we see that the \( \pm t \) flat-bands survive the increase in the unit cell, and a cluster of flat-bands (and some bands with very little dispersion) develops in the energy range \([-2.0 \leq E \leq -1.0\]) and \( 3-AGNR(n,3) \) heterostructures (not shown) and obtained qualitatively the same results as the ones shown in Fig. 8 for \( N = 3 \), which may be considered reasonable, since we can intuitively expect a lesser dependence of the electronic structure on \( N \) than on \( n \) and \( m \).

B. Band structure dependence with \( m \)

In Fig. 9, we see the band structures for 3-AGNR(1,m) heterostructures for \( m = 2 \) to 5 in panels (a) to (d), respectively. Here, we also reproduced Fig. 2, in panel (b), to facilitate comparison. As seen with the variation of \( n \) (but to a lesser degree), we see in Fig. 9, for 3-AGNR(1,m), that increasing \( m \) from 2 to 5 results in an accumulation of flat-bands close to the Fermi energy. In addition, as observed for the \( n \)-variation, the results for the \( m \)-variation of the 5-AGNR(1,m) heterostruc-
FIG. 9. Band structures for 3-AGNR(1,m) heterostructures for m = 2 to 5, in panels (a) to (d), respectively.

1. The ±t flat-bands, present in the pristine AGNRs, are also present for all values of N, n, and m investigated here, and they are associated to the same Wannier-like states identified in the pristine AGNRs [67].

2. For N-AGNR(1,3) (N = 3 and 5), the ±t bands are double degenerate (in contrast to the pristine AGNRs) and the partner state is also a Wannier-like state, similar to the one mentioned in the item above. This degeneracy is lifted for N > 5.

3. Additional flat-bands appear around the ±t flat-bands for all heterostructures analyzed, and to each different flat-band it was possible to associate a Wannier-like state that seems like a variant of the ±t Wannier-like state.

4. Regarding the variation of these additional flat-bands with N, we see that they survive (i.e., have zero-dispersion) up to N = 7 for all heterostructures studied here.

5. With increasing n, we see that the overall number of flat-bands increases, with a cluster of them forming gradually closer to the Fermi-energy, with one of them seating almost at the Fermi energy already for the 3-AGNR(10,3) heterostructure. This description of the n dependence applies to all prime values 3 ≤ N ≤ 7.

6. Similar to the n-dependence, there is an increase in the number of flat-bands with m, with a similar accumulation close to the Fermi energy. As well, this description qualitatively applies to all prime values 3 ≤ N ≤ 7.

We should also mention that a brief study of the so-called ‘Staggered’ heterostructures, which are less symmetric than the ones analyzed here (see Refs. [52, 53]), has shown a tendency to form considerably less flat-bands, indicating that the heterostructures discussed here are the ones that should receive more attention in the quest for quasi-1D ferromagnetism.

VI. FERROMAGNETIC PHASE OBTAINED WITH DFT

To address the possible existence of any magnetic phase under hole-doping, we will use DFT, which is a more realistic calculation than tight-binding and that can treat correlations at the mean-field level. We will search for indications of a ferromagnetic ground-state on two heterostructures, viz., 3-AGNR(1,3) and 5-AGNR(1,3). According to Ref. [67], the presence of itinerant carriers is important to mediate ferromagnetism between the isolated magnetic moments in each unit cell of the Wannier-like states. The 3-AGNR(1,3) and 5-AGNR(1,3) heterostructures present dispersive bands intercepting the flat-bands, as can be seen in Figs. 2 and 5(a), respectively. We will postpone a careful DFT analysis of the ferromagnetic ground-state dependence on the parameters n and m to a future publication.

A. Details of the DFT calculations

We do a DFT calculation within the projector augmented wave scheme [71] for the pseudopotentials. The total energies and electronic structures are self-consistently computed within a plane-wave basis-set with a kinetic energy cut-off of 350 eV. We used the Vienna Ab initio Simulation Package (VASP) [72, 73]. For a better description of the exchange-correlation term of the DFT, we use a hybrid functional to improve the description of the many-electron interactions and
charge localization [74]. The HSE06 hybrid functional has been used [75], where the screened functional contains part of the exact Hartree-Fock exchange that has been shown to give accurate results for the exchange splitting, which is crucial to understand the magnetic properties in our system. Interestingly, our results show that the inclusion of the hybrid functional puts the $E_{3b}$ flat-band around 3 eV from the Fermi energy, matching the tight-binding results (see Fig. 10). By suppressing the hybrid functional, using just the generalized gradient approximation [76], the $E_{3b}$ band stays around 2.5 eV from the Fermi level. As we are using the periodic supercell approach within the first principles calculations, the exchange interactions between adjacent unit cells are also included.

B. Band structure for 3-AGNR(1,3): comparison DFT/tight-binding

Panels (a) and (b) in Fig. 10 show a comparison of the DFT and tight-binding band structures for a 3-AGNR(1,3) heterostructure, at half-filling, respectively. As expected, the DFT bands are not particle-hole symmetric, but, other than that, there is a good qualitative agreement between DFT and tight-binding. The numbered bands are discussed in the text.

FIG. 10. DFT and tight-binding band structures for a 3-AGNR(1,3) heterostructure, at half-filling, in panels (a) and (b), respectively. As expected, the DFT bands are not particle-hole symmetric, but, other than that, there is a good qualitative agreement between DFT and tight-binding. The numbered bands are discussed in the text.

may create an overlap between the Wannier-like states in adjacent unit cells and result in dispersion (as discussed above in relation to the $E_{3b}$ tight-binding band for a 5-AGNR(1,3) heterostructure). On the other hand, the Wannier-like states [see panels (b) and (c) in Fig. 4] for the bands denoted 2 and 3a in Fig. 10 are much more concentrated at the center of the unit cell [especially for band 3a, see Fig. 4(c)] and thus they should be more robust against perturbations that could create an overlap between adjacent unit cells. Thus, as expected, DFT bands 2 and 3a are perfectly flat. Finally, the same reasoning leads us to expect that the DFT bands 3b and 4 should acquire dispersion, as they do indeed, the latter less so than the former.

A final point can be made, along the lines of the qualitative discussion above, if we compare our DFT results with the DFT results in Ref. [67]. There, it was obtained, for a pristine (no extensions) 5-AGNR, that the ±z DFT flat-bands, at zero doping, acquire a dispersion of ≈ 0.4 eV (see Fig. 4(a) in Ref. [67]). On the other hand, the DFT ±z bands for N-AGNR(1,3), for $N = 3$ [band 3a in Fig. 10(a)] and $N = 5$ (not shown), are perfectly flat. This seems to indicate that in an N-AGNR(n,m) heterostructure, which has a wider unit cell than a pristine AGNR, the charge density of the ±z Wannier-like states in each unit cell [like the ones shown in Figs. 4(c), 6(c), and 7(c)] is even more insulated from the charge density in adjacent unit cells, and thus can result in a more robust (more massive) DFT flat-band.

C. DFT bands at finite doping and ferromagnetic ground-state

To bring the Fermi energy close to the flat-bands, and thus investigate their properties, we start hole-doping the 3-AGNR(1,3) heterostructure. We measure the hole-doping $x_h$ from the half-filling point, thus $x_h = 1 - \langle n \rangle$ (therefore, $x_h = 0$ at half-filling), where $\langle n \rangle$ is the electron average site-occupancy.

FIG. 11. DFT band structures for a 3-AGNR(1,3) heterostructure at different hole-dopings: (a) $x_h = 0.10$, (b) $x_h = 0.0$ (half-filling), and (c) $x_h = 0.23$. In panels (a) and (c), majority-spin bands are in blue and minority-spin bands are in red.

In Fig. 11, we show the DFT bands for $x_h = 0.10, 0.0$ (half-filling), and 0.23, in panels (a) to (c), respectively. The Fermi
energy is at $E = 0.0$ in each panel. In panel (b), we repeat the results shown in Fig. 10(a) to better illustrate the hole-doping effects. In Figs. 11(a) and 11(c), at finite doping, we show the spin-decomposed band structure obtained through a hybrid DFT calculation, where the majority-spin bands are denoted in blue and the minority-spin bands are in red. The cyan arrows connecting the center panel to each one of the adjacent panels indicate the extent of the exchange splitting of each flat-band. The arrows connecting band 2 [in panel (b)] to the corresponding exchange-split bands in panel (a) indicate the extent of the exchange splitting energy acting over band 2 for $x_h = 0.1$, given by $E_{2,0.1} \approx 1.0\ eV$. Likewise, the arrows connecting panels (b) and (c) indicate the exchange splitting energy of band $3a$ for $x_h = 0.23$, corresponding to $E_{3a,0.23} \approx 2.0\ eV$.

In Fig. 12, we show the energy difference between the ferromagnetic and paramagnetic states, $\Delta E = E_{FM} - E_{PM}$, for both a 3-AGNR(1,3) (blue circles) and a 5-AGNR(1,3) (purple left-triangles), where $\Delta E < 0$ indicates a ferromagnetic ground-state. The most stable ferromagnetic configuration occurs when the hole-doping reaches the $3a$ flat-band, for both 3- and 5-AGNR(1,3). The inverse dependence of the ferromagnetic stability with $N$ can be attributed to the reduction of the overall band flatness as $N$ increases (see Figs. 2 and 5).

From Ref. [67], we obtain that the gain in energy due to ferromagnetic ordering of a pristine 5-AGNR is $\Delta E_p \approx -37.5\ meV$ (per unit cell). Since the number of occupied Carbon atoms in the ferromagnetic state in each unit cell is $N_{occ} = 6$ (see Fig. 5(b) in Ref. [67]), we obtain $\Delta E_p/N_{occ} = -6.25\ meV$. The corresponding results for the two heterostructures we analyzed through DFT, i.e., 3-AGNR(1,3) and 5-AGNR(1,3), were $\Delta E_3 = -150\ meV$, $N_{occ} = 8$, and $\Delta E_5 = -105\ meV$, $N_{occ} = 10$. This results in $\Delta E/N_{occ} = -18.8\ meV$ and $-10.5\ meV$, respectively. This shows that, if we compare the ferromagnetic energy gain for the pristine 5-AGNR and the 5-AGNR(1,3), the heterostructure had almost 70% more energy gain than that of the pristine AGNR. We believe that to be the case for two main reasons. First, the N-AGNR(n,m) heterostructures studied here through DFT present true flat-bands, contrary to what was seen in the pristine N-AGNRs studied in Ref. [67]. Second, the pristine N-AGNRs show a single low-dispersion band, while our N-AGNR(n,m) heterostructures show multiple perfectly flat-bands [two in the case of 3-AGNR(1,3), bands labeled 2 and 3a in Fig. 10(a)] and multiple almost flat-bands [two in the case of 3-AGNR(1,3), bands 3b and 4 in Fig. 10(a)], which should clearly result in a more robust ferromagnetic ground state.

**VII. WANNIER-LIKE STATES: COMPARISON BETWEEN DFT AND TIGHT-BINDING**

In this section, we want to highlight the fact that it is not only the DFT and tight-binding band structures that are qualitatively similar (as shown in Fig. 10), but also the Wannier-like states associated with the flat-bands obtained by either method that are qualitatively similar too.
pristine AGNR ±t bands are still present in the heterostructures, but with the interesting presence of a degenerate partner (for \( N = 3 \) and 5) in the tight-binding simulations. This degeneracy is slightly lifted in the DFT results for all values of \( N \). Importantly, our DFT results show that a few of the flat-bands observed in the tight-binding simulations remain perfectly flat in the DFT simulations as well. Thus, the ferromagnetism observed in our DFT results is considerably stronger than that observed in pristine AGNRs [67]. As a bonus, we found that the charge density associated with the flat-bands obtained via tight-binding agree surprisingly well with the corresponding results obtained through DFT.

Given the experimental availability of these heterostructures, our results suggest that it would be interesting to experimentally explore the possibility of ferromagnetism in these systems, which, given the variety of parameters that can be manipulated, opens up the possibility of looking for non-trivial topology in a ferromagnetic quasi-1D system.

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**Appendix A: Tight-binding Hamiltonian for an N-AGNR(1,3) heterostructure**

In this Appendix, we present explicit expressions for the Hamiltonian of an N-AGNR(1,3) heterostructure, in the real and reciprocal spaces. The modifications necessary to obtain the Hamiltonian for a general N-AGNR(n,m) heterostructure are straightforward. In Fig. 15, we show the \( l \)-th unit cell of an N-AGNR(1,3) heterostructure, where the A sublattice is represented by blue solid dots and the B sublattice by red solid dots. The sites are labeled \( p\alpha q \), where \( \alpha = A/B \), with \( 1 \leq p \leq N + 4 \) and \( 1 \leq q \leq 3 \), where \( p \) runs along the \( y \)-direction, as indicated in the right-hand side, and \( q \) runs along the \( x \)-direction (starting at the center of the unit cell and moving to its borders).

Using the labeling defined above, we can write the N-AGNR(1,3) Hamiltonian in real space as
where \( a_p(p) \) [\( b_p(p) \)] annihilates an electron on site \( pA_q \) \( (pB_q) \) on the \( l \)-th unit cell. Assuming periodic boundary conditions along the \( x \)-direction, we take a Fourier transform along that direction and obtain the reciprocal space Hamiltonian

\[
H = -t \sum_l \left[ \sum_{p \text{ odd}}^{N} b_{l,1}^\dagger(p) a_{l,1}(p) + \sum_{p \text{ even}}^{N-1} b_{l,1}^\dagger(p+1) a_{l,1}(p) + \sum_{p=2}^{N-1} a_{l,1}^\dagger(p+1) b_{l,1}(p) + \text{H.C.} \right]
\]

\[
- t \sum_l \left[ \sum_{p \text{ even}}^{N-2} b_{l,2}^\dagger(p) a_{l,2}(p) + \sum_{p \text{ even}}^{N-2} a_{l,2}^\dagger(p+1) b_{l,2}(p) + \text{H.C.} \right]
\]

\[
- t \sum_l \left[ \sum_{p \text{ even}}^{N-1} b_{l,3}^\dagger(p) a_{l,3}(p) + \sum_{p \text{ even}}^{N-1} b_{l,3}^\dagger(p+1) a_{l,3}(p) + \sum_{p=2}^{N-3} b_{l-1,3}^\dagger(p) a_{l,3}(p) + \text{H.C.} \right],
\]

Appendix B: Tight-binding with next-nearest-neighbor hopping

In Fig. 16 we present tight-binding and DFT results to assess the stability of the tight-binding flat-bands to the addition of a NNN hopping \( t_{\text{NNN}} \) to the calculations. In Fig. 16(a) we reproduce the tight-binding bands shown previously in Fig. 10(b) for 3-AGNR(1,3), which included just nearest-neighbor (NN) hoppings. In Fig. 16(b) we add NNN hoppings \( t_{\text{NNN}} = 0.1 \text{ eV} \) [68] to the calculations. As expected, the results are not particle-hole symmetric anymore. However, all the flat-bands (in the interval of energy shown) remain flat. Thus, since the DFT results [in panel (c), reproduced from Fig. 10(a)] show that flat-band 1 (the closest to the Fermi energy) has acquired dispersion, we conclude that longer hoppings than NNN are necessary in the tight-binding calculations to produce dispersion in flat-band 1. This can be understood by looking at the Wannier-like state for this band,
FIG. 15. Site labeling of a unit cell for an N-AGNR(1,3) heterostructure. The sites are labeled $p\alpha q$, where $\alpha = A/B$, $1 \leq p \leq N + 4$ and $1 \leq q \leq 3$, see text for details.

shown in Fig. 4(a). There, we clearly see that, to connect two unit cells, it is necessary at least a 3rd NN hopping. This may explain too, why flat-band 3b has acquired a small dispersion, while flat-band 4 has acquired just a slight dispersion.

FIG. 16. (a) Tight-binding band structure for 3-AGNR(1,3) with NN hoppings only [reproduced from Fig. 10(b)]. (b) Same as in panel (a), but adding a NNN hopping $t_{NNN}$ to the calculations, with $t_{NNN} = 0.1$ eV. [68] (c) DFT results for 3-AGNR(1,3) [reproduced from Fig. 10(a)].

Appendix C: Results for ‘Staggered’ heterostructures

In Refs. [52, 53] a second type of heterostructure has been introduced, less symmetric than the one we analyzed in this work. The reason we did not focus our attention in these so-called ‘Staggered’ heterostructures is that they show less flat-bands than the so-called ‘Inline’ heterostructures (which were the focus of this work). To exemplify that, in Fig. 17(a) we compare the tight-binding band structure results for a 5-AGNR-S(1,3) heterostructure [panel (a)] with that for a 5-AGNR-(1,3) one [panel (b)]. Notice the inclusion of an ‘S’ (in bold, for Staggered) to the label for the heterostructure. On top of Fig. 17(a) we show a single unit cell for the 5-AGNR-S(1,3) heterostructure. By comparing it to the single unit cell on top of panel (b) [for 5-AGNR-(1,3)], which was described in Sec. IIA, it is easy to understand the meaning of the (1,3) nomenclature, since the idea is the same as the one introduce in Sec. IIA.

By comparing the two panels, one notices that only the $\pm t$ flat-bands have survived in the Staggered heterostructure. We have checked that what appears to be two flat-bands (touched by a dispersive band, located, respectively, between energies $-1$ eV and $-2$ eV, and below energy $-3$ eV) are in reality two slightly dispersive bands, and not perfectly flat, like the $\pm t$ flat-bands. Our conclusion also rests in the fact that we could not discern a clear Wannier-like state associated to them. Thus, in the 5-AGNR-S(1,3) heterostructure there are just $1/4$ of the flat-bands present in the (Inline) 5-AGNR(1,3) heterostructure, shown in panel (b).

Finally, for completeness sake, in Fig. 18 we show a comparison of the DFT band structure for 3-AGNR-S(1,3), with the tight-binding band structure, in panels (a) and (b), respectively. Aside from the expected broken particle-hole symmetry in the DFT bands, it is easy to see the very good agreement between the two results. A careful analysis of the DFT results shows that the only flat-band that is perfectly non-dispersive is the $\pm t$ flat-band (located just below $-3$ eV), reinforcing our claim that the Inline heterostructures have more robust flat-bands.
FIG. 18. (a) DFT band structure for 3-AGNR-S(1,3). (b) Tight-binding band structure for 3-AGNR-S(1,3).

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