

# Unrestricted Hartree-Fock analysis of the Hubbard model on single-layer graphene and Bernal bilayer graphene

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We perform unrestricted Hartree-Fock calculations on the two-dimensional Hubbard model on a honeycomb and bilayer honeycomb lattice at both zero and finite temperatures. Finite-size real-space calculations are supplemented with random-phase approximation calculations in the thermodynamic limit. Our motivation comes from high doping levels achieved in graphene and Bernal bilayer graphene by intercalation. We present phase diagrams in doping and temperature for a moderate Hubbard interaction. The magnetic states we find are classified systematically based on the dominant Fourier components of their spin patterns, their average magnetization and spin incommensurabilities. The dominant spin patterns are Néel order and various types of stripes. Around Van Hove filling, we resolve the competition between stripe and chiral spin density waves in the symmetry-broken regime. We also investigate the effect of an applied external displacement field on the spin patterns of Bernal bilayer graphene.

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## I. INTRODUCTION

The Hubbard model in its minimalist setup is a key model for understanding strongly correlated materials. Despite its simplicity, it can give rise to a rich variety of interaction-induced ordered phases and more exotic correlation phenomena, such as antiferromagnetism, superconductivity, or a pseudogap phase. While impressive progress on its phase diagram was achieved, the large possible parameter space continues to provide unexplored regions, including its application to different lattice geometries in two spatial dimensions [1,2].

In the most prominent representative—the square-lattice Hubbard model—a Néel antiferromagnet arises at half filling, and makes room for various magnetic or charge-ordered phases, such as spirals [3–15] or stripes [16–30]. Due to the many possible states, an unbiased approach toward this zoo of states is desirable and extensive numerical efforts start to converge toward a phase diagram [2,31]. Interestingly, many of these ordered phases already appear within mean-field theory, and a recent study revealed the Néel-to-spiral-to-stripe transitions using unrestricted Hartree-Fock calculations in real space [32,33]. While such mean-field calculations tend to overestimate ordering tendencies and violate the Mermin-Wagner theorem, they still provide valuable insights about dominant correlations and can serve as guidance for numerical studies or gauge theories of fluctuating magnetic order [34–36].

It is an open question how much of the square-lattice phenomenology can be transferred to other lattices. We take this as a motivation to study the phase diagram of the Hubbard model on a single- and bilayer honeycomb lattice. The model can be used to study interaction effects in graphene and Bernal bilayer graphene (BBG) [37,38]. Previously, interaction-induced ordered states in these systems were theoretically studied mainly at charge neutrality and Van Hove filling. A competition between different quantum spin, charge, or valley Hall states arises at charge neutrality, in which a (layered) Néel antiferromagnet was predicted to win for a Hubbard interaction above a critical value [39–52]. To see this in BBG, trigonal warping or self-energy effects need to be taken into account [53]. Around Van Hove filling, the interplay between magnetism and superconductivity was investigated [54–58]. An approximately nested Fermi surface at this filling enables three degenerate spin density wave states with symmetry-related nesting vectors  $M_i$ . Their collinear or orthogonal combination leads to a gapped chiral or a half-metallic uniaxial spin density wave [59–62].

Experimentally, single-layer graphene is a stable semimetal at charge neutrality. In contrast, suspended samples of BBG show signatures of interaction-induced ordered states including a layered antiferromagnet, quantum anomalous Hall, and nematic states [63–71]. Furthermore, it was demonstrated that high doping regimes can be achieved by intercalation in both single- and bilayer graphene [72–76]. Single-layer graphene was even doped beyond its Van Hove singularity [74]. This promises the possibility to also realize interaction-induced ground states in highly doped graphene systems.

In this work, we perform a comprehensive analysis of the phase diagram of graphene and BBG as modeled by a Hubbard model on the single- or bi-layer honeycomb lattice. We

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employ unrestricted Hartree-Fock calculations in real space, complemented by an instability analysis via a randomphase approximation (RPA) in the thermodynamic limit. We complete the phase diagram for the entire range of doping from empty to fully filled bands at zero and finite temperatures. Similar to the square-lattice Hubbard model, we find that a (layered) Néel antiferromagnet transitions into a cascade of stripe phases. However, for larger dopings the phase diagram on the single- and bilayer honeycomb lattices is qualitatively different due to their Van Hove singularities. We observe a competition between spin density waves with three Fourier modes. This includes the detailed temperature and density dependence of the chiral and uniaxial spin density waves that were previously studied at Van Hove doping. Furthermore, we analyze the effect of unequal layer occupation in BBG, which can arise from asymmetric intercalation or an external displacement field. Note that we focus on density regimes beyond the cascade behavior recently observed in BBG in a displacement field at the lowest densities [77–79]. We find that for intermediate interaction strengths, the overall field dependence can be well understood based on the phase diagram of two almost decoupled single layers.

The rest of the paper is structured as follows. In Sec. II, we introduce the Hubbard model on a single-layer and Bernal-stacked bilayer honeycomb lattice, and we describe our unrestricted Hartree-Fock and RPA calculations. Furthermore, we provide a classification of the ordered states we find based on their real-space spin and charge patterns, and Fourier modes. In Sec. III, we present the phase diagram, magnetization, and wave vectors of the ordered states for the single- and bilayer case. We vary doping, temperature, and for BBG a displacement field. We provide a discussion of our results and a brief outlook about possible further research directions in Sec. IV.

## II. MODEL AND METHOD

Motivated by graphene and Bernal bilayer graphene, we consider the Hubbard model on a single- and AB-stacked bilayer honeycomb lattice

$$H = H_0 + H_{\text{int}} = \sum_{j,j',\sigma} t_{jj'} c_{j\sigma}^\dagger c_{j'\sigma} + U \sum_j n_{j\uparrow} n_{j\downarrow}, \quad (1)$$

with local interaction  $U > 0$  and hopping amplitudes  $t_{jj'}$ . Here,  $c_{j\sigma}^\dagger$  ( $c_{j\sigma}$ ) is the fermion creation (annihilation) operator for an electron with spin  $\sigma$  on site  $j$ , and  $n_{j\sigma} = c_{j\sigma}^\dagger c_{j\sigma}$ . The index  $j$  runs over all atoms in both layers. The hopping matrix  $t_{jj'}$  depends only on the distance of the sites  $j$  and  $j'$ . We model the single-layer case with only the nearest-neighbor hopping amplitude  $t$  being nonzero. In the bilayer case, we include nearest-neighbor hopping within a layer, and vertical  $t_\perp = 0.121t$  and skew hopping terms  $t_3 = 0.120t$ , and  $t_4 = 0.044t$  between layers [38] (see Fig. 1). We note that the hopping  $t_4$  breaks particle-hole symmetry and the bipartite structure of the system. The value of the interaction  $U$  was estimated via a constrained random-phase approximation (cRPA) to be  $U \approx 3.3t$  in graphene and  $U \approx 2.9t$  in graphite [80], which is why we choose  $U = 3t$  for our calculations below.

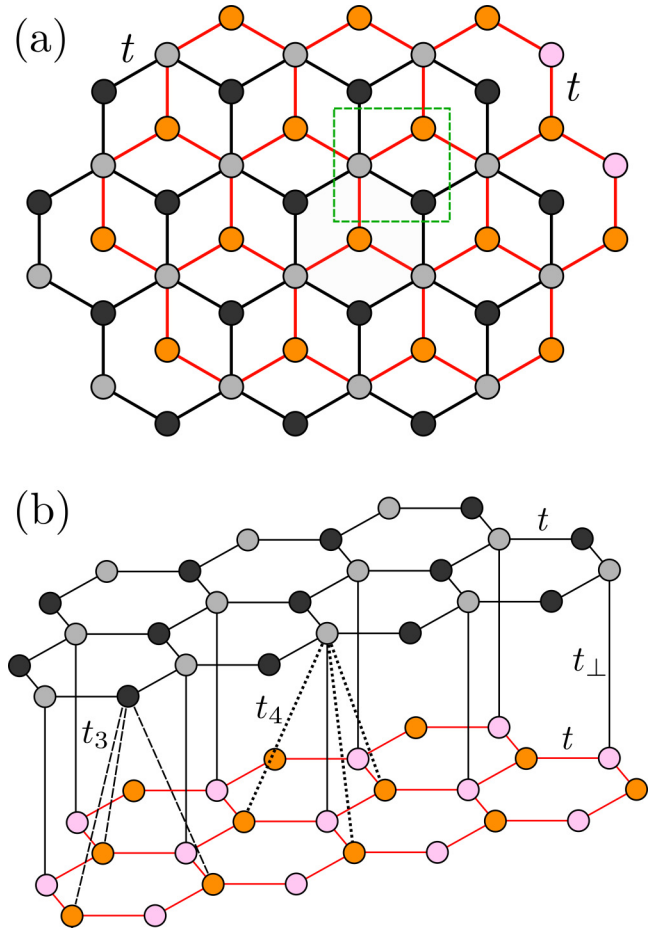


FIG. 1. Overview of the real-space structure of Bernal bilayer graphene showing (a) the top view and (b) a view from the side with an indication of the four different types of hoppings,  $t$ ,  $t_\perp$ ,  $t_3$ , and  $t_4$ . The green dashed box is one possible choice for a BBG unit cell, which contains four sites.

The unit cell of single-layer graphene contains two atoms (A,B) and that of bilayer graphene contains four ( $A_1, B_1, A_2, B_2$ ) [see the green dashed box in Fig. 1(a)]. In the sublattice basis and transforming to momentum space, the noninteracting Hamiltonian becomes  $H_0 = \sum_{\mathbf{k},\sigma} c_{\mathbf{k}\sigma}^\dagger H_{0,\mathbf{k}} c_{\mathbf{k}\sigma}$ , where  $\mathbf{c}_{\mathbf{k}\sigma} = (c_{A\mathbf{k}\sigma}, c_{B\mathbf{k}\sigma})$  or  $\mathbf{c}_{\mathbf{k}\sigma} = (c_{A_1\mathbf{k}\sigma}, c_{B_1\mathbf{k}\sigma}, c_{A_2\mathbf{k}\sigma}, c_{B_2\mathbf{k}\sigma})$ , and for single-layer graphene

$$H_{0,\mathbf{k}} = \begin{pmatrix} 0 & -t\delta_{\mathbf{k}} \\ -t\delta_{\mathbf{k}}^* & 0 \end{pmatrix}, \quad (2)$$

while for Bernal bilayer graphene

$$H_{0,\mathbf{k}} = \begin{pmatrix} 0 & -t\delta_{\mathbf{k}} & t_4\delta_{\mathbf{k}} & -t_3\delta_{\mathbf{k}}^* \\ -t\delta_{\mathbf{k}}^* & 0 & t_\perp & t_4\delta_{\mathbf{k}} \\ t_4\delta_{\mathbf{k}}^* & t_\perp & 0 & -t\delta_{\mathbf{k}} \\ -t_3\delta_{\mathbf{k}} & t_4\delta_{\mathbf{k}}^* & -t\delta_{\mathbf{k}}^* & 0 \end{pmatrix} \quad (3)$$

with  $\delta_{\mathbf{k}} = e^{ik_y/\sqrt{3}} + 2e^{-ik_y/(2\sqrt{3})} \cos(k_x/2)$ . We show the corresponding energy bands  $\epsilon_{\mathbf{k}}$  in Fig. 2. They feature linear band touchings at half filling at the  $\pm K$  points (see Fig. 3 for high-symmetry points in the Brillouin zone). BBG has three further linear band touching points very close to the

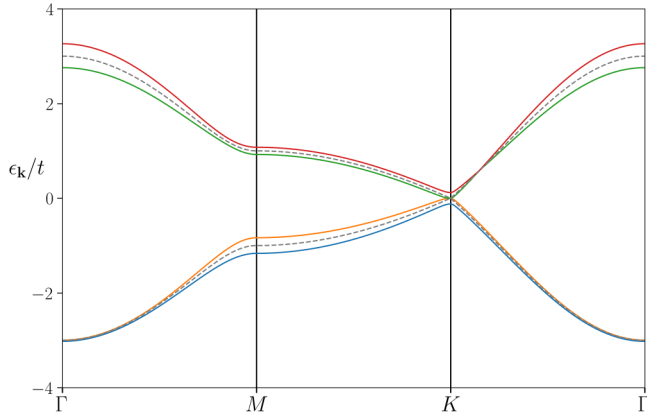


FIG. 2. The band structure of BBG (colored solid lines) and single-layer graphene (gray dashed lines). The bands are shown along the high-symmetry path  $\Gamma$ - $M$ - $K$ - $\Gamma$ .

$\pm K$  points. For large enough dopings of 25%, a Van Hove singularity can be reached in single-layer graphene. In bilayer graphene, there are two subsequent Van Hove energies of the two conduction/valence bands. The Fermi surface at the Van Hove energy is approximately nested with wave vector  $M$ .

In addition, the band structure of bilayer graphene can be tuned by an electric displacement field. For a nonzero displacement field, we add the term

$$H_D = \frac{D}{2} \sum_j \epsilon_j n_j \quad (4)$$

to the Hamiltonian where  $\epsilon_j = 1$  in the upper layer and  $\epsilon_j = -1$  in the lower layer. We note that doping of bilayer graphene via intercalation also leads to an asymmetry in the charge-carrier density of both layers and can be modeled in an analogous way [72,75].

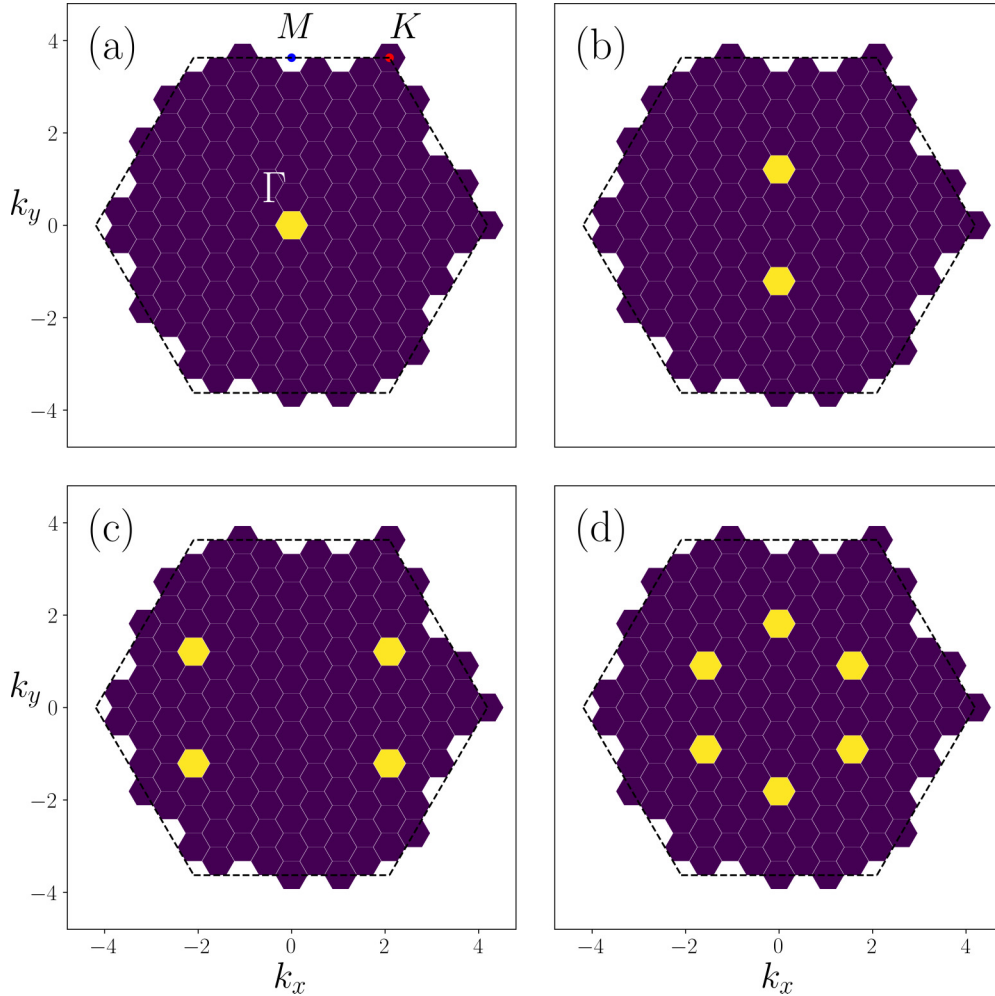


FIG. 3. The four most common Fourier patterns we find in our real-space Hartree-Fock calculations. (a) The spin order has only a single peak at  $\mathbf{Q} = 0$ , indicating a resulting Néel or ferromagnetic order depending on the sublattice structure. (b) There is a single contributing Fourier mode at  $\mathbf{q} = \pm \mathbf{Q}$ , corresponding to incommensurate single-mode stripe order. (c) The spin order consists of two symmetry-related Fourier modes, which we mainly find in the two orthogonal stripe-ordered phase. We here show the special case where they have the same amplitude. (d) Three symmetry-related Fourier modes occur with the same amplitude. For example, an order with three orthogonal or collinear stripes shows this Fourier pattern. As a special case, the three Fourier modes can occur at the three  $M$  points.

### A. Unrestricted Hartree-Fock in real space

For an unbiased analysis of spin and charge orders, we perform a mean-field decoupling of the interaction term of Eq. (1) in real space. The resulting mean-field expression is of the form [17,32,81,82]

$$H_{\text{int}}^{\text{MF}} = \sum_{j\sigma} \Delta_{j\bar{\sigma}} n_{j\sigma} + \sum_j (\Delta_{j-} c_{j\uparrow}^\dagger c_{j\downarrow} + \Delta_{j+} c_{j\downarrow}^\dagger c_{j\uparrow}) - \frac{1}{U} \sum_j (\Delta_{j\uparrow} \Delta_{j\downarrow} - \Delta_{j-} \Delta_{j+}), \quad (5)$$

where we defined  $\bar{\uparrow} \equiv \downarrow$  and  $\bar{\downarrow} \equiv \uparrow$ . We determine the parameters  $\Delta_{j\alpha}$  self-consistently by the gap equations

$$\begin{aligned} \Delta_{j\sigma} &= U \langle n_{j\sigma} \rangle, \\ \Delta_{j+} &= \Delta_{j-}^* = -U \langle c_{j\uparrow}^\dagger c_{j\downarrow} \rangle. \end{aligned} \quad (6)$$

The decoupling in Eq. (6) captures both the Hartree ( $\Delta_{j,\sigma}$ ) and the Fock ( $\Delta_{j,\pm}$ ) contributions and allows us to describe arbitrary spin and charge ordering patterns. The local charge and spin expectation values are given by

$$\begin{aligned} \langle n_j \rangle &= \frac{\Delta_{j\uparrow} + \Delta_{j\downarrow}}{U}, \\ \langle S_j^z \rangle &= \frac{\Delta_{j\uparrow} - \Delta_{j\downarrow}}{2U}, \\ \langle S_j^x \rangle &= -\frac{\Delta_{j+} + \Delta_{j-}}{2U}, \\ \langle S_j^y \rangle &= -\frac{\Delta_{j+} - \Delta_{j-}}{2iU}, \end{aligned} \quad (7)$$

with  $n_j = n_{j\uparrow} + n_{j\downarrow}$  and  $\vec{S}_j = \frac{1}{2} c_j^\dagger \vec{\sigma} c_j$ , where  $\vec{\sigma}$  is the vector of Pauli matrices.

We solve the mean-field equations on a finite lattice consisting of  $N$  unit cells and periodic boundary conditions. We choose a rhombus shape for the lattice with the same number of sites along both axes of the rhombus. The unit cells of single- and bilayer graphene contain two and four atoms, respectively. Thus, in total, the number of sites in our calculation is  $\mathcal{N} = 2N$  for single-layer and  $\mathcal{N} = 4N$  for bilayer graphene. We typically perform the real-space calculations on lattices with  $18 \times 18$  unit cells for single-layer and  $12 \times 12$  unit cells for bilayer graphene, i.e., we have 648 and 576 sites, respectively.

With these specifications the mean-field Hamiltonian can be written as

$$H^{\text{MF}} = H_0 + H_{\text{int}}^{\text{MF}} = \sum_{j,j'} \sum_{\sigma,\sigma'} c_{j\sigma}^\dagger \mathcal{H}_{jj'}^{\sigma\sigma'} c_{j'\sigma'} + \text{const}, \quad (8)$$

where  $\mathcal{H}_{jj'}^{\sigma\sigma'} \in \mathbb{C}^{2\mathcal{N} \times 2\mathcal{N}}$  is a square matrix with  $4\mathcal{N}$  real parameters to be determined self-consistently ( $\Delta_{j\uparrow}$ ,  $\Delta_{j\downarrow}$ ,  $\text{Re}\Delta_{j+}$ , and  $\text{Im}\Delta_{j+}$  for each lattice site  $j$ ). The expectation values of Eq. (6) are then given by

$$\langle c_{j\sigma}^\dagger c_{j'\sigma'} \rangle = \sum_{\ell=1}^{2\mathcal{N}} (v_{j\sigma}^\ell)^* v_{j'\sigma'}^\ell f(\epsilon_\ell - \mu), \quad (9)$$

where  $f(x) = 1/(1 + e^{x/T})$  is the Fermi function,  $T$  the temperature,  $\epsilon_\ell$  are the eigenvalues of the matrix  $\mathcal{H}_{jj'}^{\sigma\sigma'}$  from

Eq. (8), and  $v_{j\sigma}^\ell$  is the normalized eigenvector corresponding to the  $\ell$ th eigenvalue. This formulation allows us to perform the mean-field approximation also at finite temperatures.

Furthermore, we keep the total average of the electron density  $n$  fixed in our calculation by enforcing the constraint

$$n = \frac{1}{\mathcal{N}} \sum_{j\sigma} \langle n_{j\sigma} \rangle = \frac{1}{\mathcal{N}} \sum_{\ell=1}^{2\mathcal{N}} f(\epsilon_\ell - \mu). \quad (10)$$

To achieve this, we adjust the chemical potential  $\mu$  in each iteration of the self-consistency loop.

We consider a state to be converged if either all parameters  $\Delta_{j\alpha}$  from Eq. (6) vary by less than  $10^{-8}t$  between successive iterations, or if the number of iterations reaches 3000. Lowering the threshold below  $10^{-8}t$  does not affect the results. The second criterion ensures that the computation eventually terminates; in practice, 3000 iterations are typically more than sufficient to obtain a well-defined state that can be classified as the same type of magnetic order as the fully converged solution.

Finally, when we obtain a convergent solution of the self-consistent mean-field equations, we calculate the free energy for the quadratic Hartree-Fock Hamiltonian as [17,83]

$$\begin{aligned} F/\mathcal{N} &= -\frac{T}{\mathcal{N}} \sum_{\ell} \ln(1 + e^{-\beta(\epsilon_\ell - \mu)}) \\ &\quad - \frac{1}{\mathcal{N}U} \sum_j (\Delta_{j\uparrow} \Delta_{j\downarrow} - \Delta_{j-} \Delta_{j+}) + \mu n. \end{aligned} \quad (11)$$

Here,  $\epsilon_\ell$  are the Hartree-Fock eigenvalues from above and the second term takes the condensation energy from the mean-field decoupling into account [cf. Eq. (5)]. We perform this Hartree-Fock procedure for varying temperature, density, and displacement field. To ensure that we do not converge to a local minimum, we repeat each calculation several times from random initial spin and charge patterns and after that from the (converged) states of adjacent points in the phase diagram and then compare their free energies via Eq. (11).

With a few exceptions, we consider calculations on a fixed lattice size. This means that the exact phase boundaries are subject to finite-size effects. The influence of these kinds of finite-size effects was analyzed in greater detail for the Hubbard model on a square lattice in [32,33]. For an even more precise picture of the phase diagram, one could repeat the calculations for various lattice sizes comparing their free energies or constrain the spin orders in order to work directly in the thermodynamic limit. For our purposes, we intend to give an overview of the regimes where different phases are dominant without calculating sharp phase boundaries, and leave their exact determination for future study. We still perform checks on  $18 \times 18$  and  $24 \times 24$  lattices for several points for BBG to ensure a correct convergence (see Appendix C).

### B. Random-phase approximation

We additionally performed calculations for the (paramagnetic) spin susceptibility in the thermodynamic limit using the RPA [84,85].

For the RPA summation we introduce the Green's function of the noninteracting system  $G_0$  and calculate the static spin

susceptibility

$$\begin{aligned}\chi(\mathbf{q}) &= \chi_0(\mathbf{q})[\mathbb{1} - U\chi_0(\mathbf{q})]^{-1}, \\ \chi_0^{\alpha\beta}(\mathbf{q}) &= -\int_k G_0^{\alpha\beta}(k+q)G_0^{\beta\alpha}(k), \\ G_0(k) &= (i\nu + \mu - H_{0,\mathbf{k}})^{-1},\end{aligned}\quad (12)$$

where we introduced the shorthand notation  $\int_k = T \sum_{i\nu} \int_{\mathbf{k} \in \text{BZ}} d\mathbf{k} \sqrt{3}/(8\pi^2)$  and  $k = (i\nu, \mathbf{k})$ ,  $q = (0, \mathbf{q})$  contain a fermionic Matsubara frequency  $\nu$  and a wave vector  $\mathbf{k}$  or  $\mathbf{q}$ .  $\alpha$  and  $\beta$  refer to the sublattices.

The RPA is performed in the paramagnetic phase to analyze the transition towards the symmetry-broken phase. In the paramagnetic phase, the Green's function  $G_0$  is independent of spin  $\sigma$  so that also  $\chi_0$  does not depend on which spin combination is considered. An instability in the susceptibility signals the onset of order and allows us to compare the mean-field critical temperature  $T^*$  of the finite-size and infinite systems. We assume that right below the critical temperature  $T^*$ , the spin order will be given by a single contributing mode with wave vector  $\mathbf{Q}$  (and symmetry-equivalent  $\mathbf{Q}$  vectors) [33], corresponding to a diverging  $\chi(\mathbf{Q})$ . To determine  $T^*$  and  $\mathbf{Q}$ , we focus on  $\mathbf{q}$  along the high-symmetry lines (i.e.,  $\Gamma$ - $M$ - $K$ - $\Gamma$ ), and calculate the maximal critical temperature  $T_{\mathbf{q}}^*$  where at least one eigenvalue of  $[\mathbb{1}_4 - U\chi_0(\mathbf{q})]$  becomes 0 so that the denominator of Eq. (12) vanishes. We then determine the mean-field critical temperature as

$$T^* = \max_{\mathbf{q}} T_{\mathbf{q}}^*, \quad (13)$$

and  $\mathbf{Q}$  as the corresponding  $\mathbf{q}$ , where  $T_{\mathbf{q}}^*$  becomes maximal. One can furthermore determine the sublattice structure of the order by considering the form of the eigenvector corresponding to the diverging eigenvalue. For example, if the divergence occurs at  $\mathbf{Q} = \mathbf{0}$ , one can distinguish between ferromagnetic and Néel order for graphene. If that eigenvector is proportional to  $(1, -1)$ , then the magnetic order corresponds to Néel antiferromagnetism and if it is proportional to  $(1, 1)$  it corresponds to ferromagnetism. Note that the chemical potential needs to be calculated for each set of filling and temperature. Both the Hartree-Fock approximation and the RPA overestimate the critical temperature of the system, as order-parameter fluctuations are neglected.

### C. Classification of real-space states

Before we discuss the phase diagrams, we give an overview of the types of magnetic states we find and how we categorize them. We perform the classification of states based on the pattern of the spin expectation values. This classification is inspired by similar classifications for spin patterns on the square lattice [32,86]. In agreement with [86,87], we generally find that the charge pattern of our states follows the spin pattern according to

$$\langle \rho_j \rangle \propto \langle \vec{S}_j \rangle^2 + \text{const.} \quad (14)$$

Note that in general this relation only holds for small spin expectation values, since it was derived from a low-order Landau theory [87].

To classify a magnetic state, we consider the Fourier transformation of the real-space spin pattern

$$\langle \vec{S}_j \rangle = \langle \vec{S}_{m,\alpha} \rangle = \sum_{\mathbf{q}} \vec{S}_{\mathbf{q},\alpha} e^{i\mathbf{q}\mathbf{r}_m}, \quad (15)$$

where we specify a site  $j$  by the unit cell ( $m$ ) at position  $\mathbf{r}_m$  and the sublattice ( $\alpha$ ). Since the real-space spin pattern is real valued, the Fourier components of the spin expectation values satisfy  $\vec{S}_{\mathbf{q},\alpha} = \vec{S}_{-\mathbf{q},\alpha}^*$ . We find that the classification is the same for each of the sublattices, so we drop the extra index  $\alpha$ . The sum runs over the  $N$  allowed momenta  $\mathbf{q}$  in the Brillouin zone given by the real-space lattice size. The Brillouin zone is discretized based on the  $12 \times 12$  lattice we used to calculate our phase diagrams, i.e., there are 144 allowed momenta for the spin pattern. In particular, this includes wave vectors for the spin orders at the  $M$  or  $K$  points. In Fig. 3, we show the Fourier weight of the four most common types of spin orders we find in our numerical calculations. Generally, we obtain nonzero Fourier components at one, two, or three different  $\mathbf{q}$  and the related  $-\mathbf{q}$ . They can be at high-symmetry points  $\Gamma$ ,  $K$ , and  $M$  or along high-symmetry lines  $\Gamma$ - $K$ ,  $\Gamma$ - $M$ , or  $K$ - $M$ . In addition, we determine the (complex) amplitudes of the spin components and the relative orientation of the spins on different sublattices, since we cannot completely classify a state with its Fourier weights only.

This leads us to the following categories for the most common spin orders we find. Note that the actual spin pattern we get in our Hartree-Fock calculations can be related to the states of the categories by a global  $\text{SU}(2)$  rotation or translations of the spin order. We present example patterns in Fig. 4, illustrated by showing the real-space configuration of one graphene layer projected onto the  $x$ - $y$  plane in spin space.

#### 1. Paramagnetism

In the paramagnetic state, the average spin magnetization on every lattice site is zero,  $\langle \vec{S}_j \rangle = 0$ , and the average electron density is uniform,  $\langle n_j \rangle = n$ .

#### 2. Néel or ferromagnetic order

The spin order in each sublattice is given by

$$\vec{S}_0 = S \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \quad \vec{S}_{\mathbf{q}} = \vec{0} \quad \text{for } \mathbf{q} \neq \mathbf{0}, \quad (16)$$

with spin amplitude  $S$ . This definition entails both ferromagnetism as well as Néel antiferromagnetism, depending on whether the spins on the different sublattices are arranged in a parallel or antiparallel way. Only if the spins are antiparallel do we classify the state as Néel order. In bilayer graphene, we additionally find that spins on sites that are directly on top of each other are antiparallel. All spins on each sublattice have the same amplitude and thus we do not find charge modulations. The Fourier weight is shown in Fig. 3(a). The real-space spin pattern is shown in Fig. 4(a).

#### 3. Single-mode stripe order

We classify a state as single mode stripe order, if it can be described by only one mode  $\mathbf{Q}$  in momentum space,

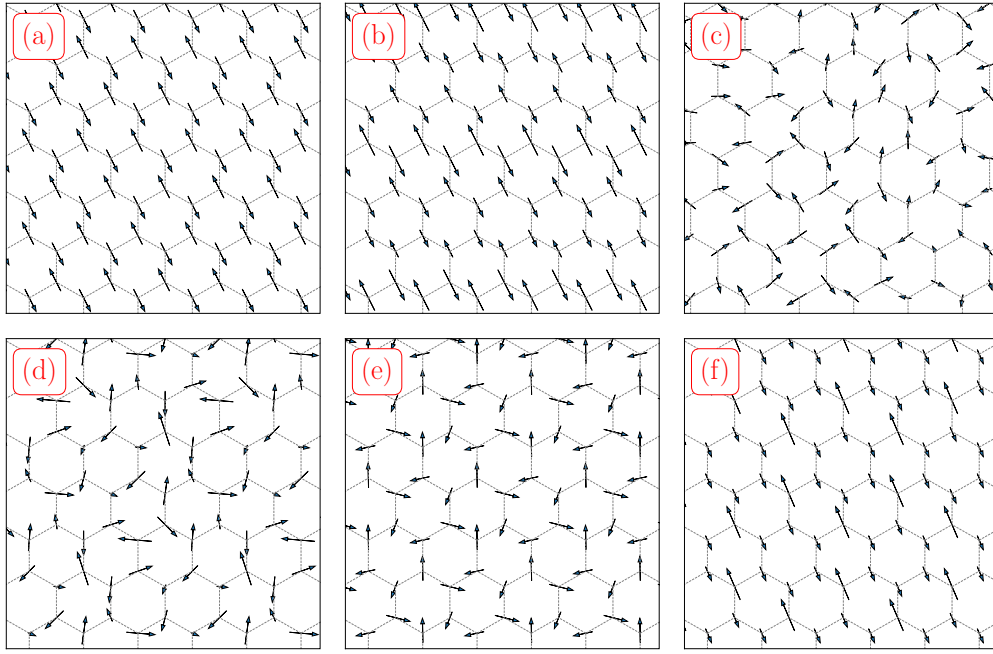


FIG. 4. Example representatives of the real-space patterns of the most common spin orders we find. We here show only one layer, but we found that in the case of BBG both layers display the same type of spin pattern for a given parameter set possibly with a relative phase. (a) Néel antiferromagnetism, (b) single mode stripe order, (c) two orthogonal stripes with the same amplitude, (d) an order with three modes with the same Fourier amplitude, which are neither orthogonal nor parallel stripes, (e) three orthogonal stripes in the special case of commensurate wave vector  $\mathbf{Q}_i = M_i$ , and (f) three collinear stripes, again in the commensurate case  $\mathbf{Q}_i = M_i$ . Orders in (d) and (e) are neither collinear nor coplanar and we only depict their projection on a plane. Spins in panel (e) have the same amplitude on each lattice site.

so that

$$\vec{S}_{\mathbf{Q}} = \vec{S}_{-\mathbf{Q}}^* = S \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \quad \vec{S}_{\mathbf{q}} = \vec{0} \quad \text{for } \mathbf{q} \neq \pm \mathbf{Q}. \quad (17)$$

All the spins are pointing collinearly along one axis with their amplitude modulated with wave vector  $\mathbf{Q}$ . The charge modulation on a sublattice will be to lowest order proportional to  $\langle n_m \rangle - n \propto \cos(2\mathbf{Q} \cdot \mathbf{r}_m)$ . The Fourier weight of single mode stripe order corresponds to Fig. 3(b). Its real-space representation is shown in Fig. 4(b). For comparison, note that, e.g., for a single-mode spiral order the Fourier amplitude of the spin pattern would have the form  $S(1, i, 0)$ .

#### 4. Two orthogonal stripes

If the spin order is given by

$$\begin{aligned} \vec{S}_{\mathbf{Q}_1} &= \vec{S}_{-\mathbf{Q}_1}^* = S_1 \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \\ \vec{S}_{\mathbf{Q}_2} &= \vec{S}_{-\mathbf{Q}_2}^* = S_2 \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}, \end{aligned} \quad (18)$$

we classify a state as two orthogonal stripes. The wave vectors  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  have to be symmetry related by a  $C_3$  rotation. We often find a special case of this state where  $S_1 = S_2$ . We show the Fourier amplitudes of this state in Fig. 3(c), where we present the common case with  $S_1 = S_2$ . In this case, the charge modulation follows  $\langle n_m \rangle - n \propto \cos(2\mathbf{Q}_1 \cdot \mathbf{r}_m) + \cos(2\mathbf{Q}_2 \cdot \mathbf{r}_m)$ . Its real-space representation is shown in Fig. 4(c).

so that

#### 5. Three modes with same amplitude

We classify a state as three modes with the same amplitude, if  $S_{\mathbf{q}}$  is nonzero only for three symmetry-related modes  $\pm \mathbf{Q}_1$ ,  $\pm \mathbf{Q}_2$ , and  $\pm \mathbf{Q}_3$ , where  $\mathbf{Q}_1$ ,  $\mathbf{Q}_2$ , and  $\mathbf{Q}_3$  are again related to each other by a  $120^\circ$  rotation and

$$|\vec{S}_{\mathbf{Q}_1}| = |\vec{S}_{\mathbf{Q}_2}| = |\vec{S}_{\mathbf{Q}_3}|. \quad (19)$$

We show an example in Fig. 3(d). We find several different types of spin orders in the form (19), which are not necessarily coplanar. We show the  $x$ - $y$  projection of one example in Fig. 4(d). To shorten notation, we will refer to this order in the following also as "three same modes." Note that the three modes do not necessarily have to be three stripes. For example, they can also be three parallel spirals if  $S_{\mathbf{Q}_1} = S_{\mathbf{Q}_2} = S_{\mathbf{Q}_3} = S(1, i, 0)$ . Most of the time, we do, however, find three stripes. Furthermore, we distinguish general states with three modes of the same amplitude from two special cases where the three spin modes are orthogonal or parallel stripes, which we specify in the following.

#### 6. Three orthogonal stripes

Three orthogonal stripes are a special case of three modes with the same amplitude, where the three spin amplitudes at

$\mathbf{Q}_1$ ,  $\mathbf{Q}_2$ , and  $\mathbf{Q}_3$  are orthogonal to each other, i.e.,

$$\begin{aligned}\vec{S}_{\mathbf{Q}_1} &= \vec{S}_{-\mathbf{Q}_1}^* = S \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \\ \vec{S}_{\mathbf{Q}_2} &= \vec{S}_{-\mathbf{Q}_2}^* = S \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}, \\ \vec{S}_{\mathbf{Q}_3} &= \vec{S}_{-\mathbf{Q}_3}^* = S \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}.\end{aligned}\quad (20)$$

The charge modulation is therefore proportional to  $\langle n_m \rangle - n \propto \cos(2\mathbf{Q}_1 \cdot \mathbf{r}_m) + \cos(2\mathbf{Q}_2 \cdot \mathbf{r}_m) + \cos(2\mathbf{Q}_3 \cdot \mathbf{r}_m)$ . Its Fourier weight looks like that of the three same mode orders, i.e., Fig. 3(d). Since this order is not coplanar, we show the projection of the spins to the  $x$ - $y$  plane in Fig. 4(e). In that panel, we show the special case where  $\mathbf{Q}_i = \mathbf{M}_i$ . This state was previously discussed as a symmetry-broken state of the doped triangular or honeycomb lattice [59–62]. It forms a four-sublattice state where the spins on each sublattice point into the four corners of a tetrahedron and the spins on each lattice site have the same magnitude.

### 7. Three collinear stripes

Three collinear stripes are a special case of three modes with the same amplitude, where the three spin amplitudes at  $\mathbf{Q}_1$ ,  $\mathbf{Q}_2$ , and  $\mathbf{Q}_3$  are collinear, i.e.,

$$\vec{S}_{\mathbf{Q}_1} = \vec{S}_{\mathbf{Q}_2} = \vec{S}_{\mathbf{Q}_3} = S \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \quad (21)$$

and its conjugate for  $\mathbf{Q}_i \leftrightarrow -\mathbf{Q}_i$ . The charge modulation is proportional to  $\langle n_m \rangle - n \propto [\cos(\mathbf{Q}_1 \cdot \mathbf{r}_m) + \cos(\mathbf{Q}_2 \cdot \mathbf{r}_m) + \cos(\mathbf{Q}_3 \cdot \mathbf{r}_m)]^2$ . In the special case  $\mathbf{Q} = \mathbf{M}$ , this state is equivalent to the collinearly ordered state proposed in [61]. Its Fourier weight is thus again represented by Fig. 3(d). We show the spin order pattern in Fig. 4(f).

### 8. Other orders

Any spin order we find that does not fit into one of the aforementioned categories, we classify as "other" spin orders.

Since we are performing our calculations on a finite system and completely unrestricted, there is the risk to converge to a local energy minimum, which is not the true lowest-energy state of the system. Generally, the other orders tend to disappear when we increase the system size. We therefore believe that the other orders are largely artifacts of convergences to local minima and finite-size effects, similar to [32].

We have also checked our Hartree-Fock solutions for possible states of phase separation, which would show themselves by exhibiting either sharp domain walls in real space or by an irregular behavior of the chemical potential [10,88–90]. We did not find strong evidence for either within the scope of our project. In Appendix A, we analyze the chemical potential of the phase diagram in more depth.

## III. RESULTS

### A. Single-layer graphene

For later reference, we perform Hartree-Fock and RPA calculations also for single-layer graphene. We vary the temperature between 0 and  $0.5t$  in steps of 0.01 and the density between 0.5 and 1.5 in steps of  $\frac{1}{108}$ . We choose an interaction strength of  $U = 3t$  because it is close to the value determined in cRPA of  $U = 3.3t$  [80]. Furthermore, this realistic value of  $U$  has the advantage that, on the one hand, it is big enough to find magnetism in a wide region of dopings and temperature, while on the other hand, it is still small enough for Hartree-Fock theory to provide meaningful results. We provide further RPA calculations for  $U = 2.75t$  and  $U = 2.5t$  in Appendix B.

Since single-layer graphene is particle-hole symmetric, we only consider hole doping. We present the phase diagram as a function of doping  $\delta = 1 - n$  and temperature, as well as the relevant spin ordering wave vectors  $\mathbf{Q}$  and the magnetization  $m$  [see Eq. (22)] in Fig. 5. In general, we find striking similarities between BBG and single-layer graphene (cf. Sec. III B).

The critical temperature as calculated in RPA coincides well with the critical temperature that we find in our real-space calculations, indicating that our system sizes are already big enough to give a good approximation of the thermodynamic limit. Overall we find a characteristic "two-dome" structure: a region of high critical temperatures around half-filling and another one around Van Hove filling. Around half-filling, we observe a Néel-ordered dome consistent with previous studies, despite the vanishing density of states because the considered interaction  $U = 3t$  is above the critical interaction strength needed in mean-field theory [39,44–47,50–52]. The critical temperature  $T^*$  is at a relative maximum because of particle-hole symmetry between conduction and valence bands  $\epsilon_k^+ = -\epsilon_k^-$  at charge neutrality. The regime of Néel order is followed by an intermediate regime of single mode stripe order. In particular, at  $T \approx 0.19$  and doping  $\delta \approx 0.0925$ , we find a triple point, where commensurate magnetism in the form of Néel order, incommensurate magnetism, and the paramagnetic state meet. Similar triple points have been discussed in previous works on different lattice geometries [90–92].

A secondary dome arises around Van Hove filling because the logarithmic singularity of the density of states together with nesting at wave vector  $\mathbf{M}$ , i.e.,  $\epsilon_{k+\mathbf{M}} = -\epsilon_{\mathbf{k}}$  boosts the critical temperature  $T^*$ . Within the secondary dome, we find an interplay of different types of three same modes, including three orthogonal and collinear stripes, at dopings between  $\delta = 0.15$  and  $\delta = 0.3$ . Consistent with previous studies we find three orthogonal stripes, i.e., a chiral spin density wave, at Van Hove doping [59–62]. Their competition with three collinear stripes is influenced by temperature [61], and more strongly by doping. For higher doping of 0.3 to 0.35, we find a single-mode stripe-ordered regime and for even higher dopings we see a small region of two orthogonal stripe order.

In the middle panel of Fig. 5, we present the  $\mathbf{Q}$  dependence of the spin patterns as a function of doping and temperature. We color code the  $\mathbf{Q}$  dependence according to their location in the Brillouin zone (cf. inset). For the real-space Hartree-Fock calculations we use the wave vector with the biggest weight of the corresponding Fourier mode of the spin pattern. The color of the outline is given by the  $\mathbf{Q}$  vector of the divergence

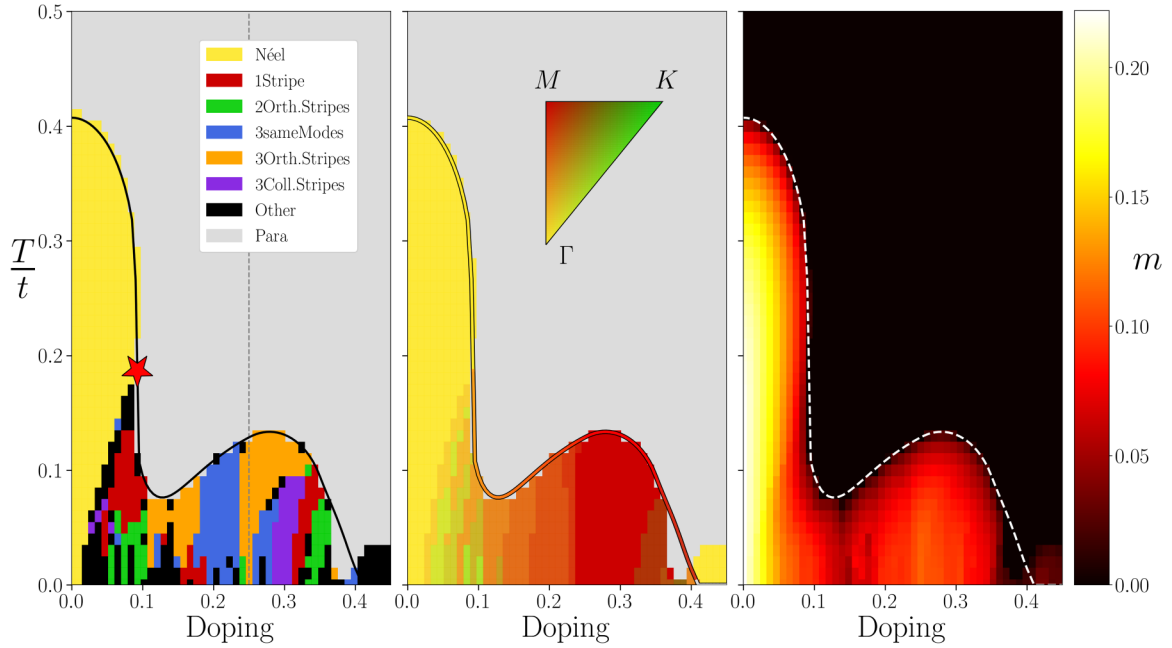


FIG. 5. The Hartree-Fock phase diagram, main wave vector  $\mathbf{Q}$  (color scale in the middle inset), and average magnetization  $m$  (right color scale) of single-layer graphene for  $U = 3t$  as a function of temperature and doping. Calculations were done on  $18 \times 18$  lattices. The gray dashed line at 25% hole doping in the left panel indicates the Van Hove filling of the noninteracting model. The  $T^*$  line and its corresponding  $\mathbf{Q}$  (color in the middle double line) were calculated in the thermodynamic limit using RPA. The red star in the left panel marks the triple point between paramagnetism, commensurate, and incommensurate magnetism.

in the RPA calculation. The  $\mathbf{Q}$  dependence of the magnetic orders is largely determined by the evolution of the Fermi surface with doping. For Fermi points and small pockets close to half-filling, we see Néel order with  $\mathbf{Q} = 0$ . For small dopings with Fermi pockets around  $K, K'$ , we find intermediate wave vectors with  $\mathbf{Q}$  moving along the  $\Gamma$ - $K$  line or the  $\Gamma$ - $M$  line. The wave-vector fluctuations in this regime indicate that these states are close in energy. In general,  $|\mathbf{Q}|$  increases for increasing doping or lower temperatures. In the secondary dome, the spin order reaches the  $M$  point approximately at Van Hove doping, which corresponds to the nesting wave vector. Temperature effects slightly shift the maximum away from Van Hove doping. At very high dopings,  $\mathbf{Q}$  shifts toward  $K$  along the  $M$ - $K$  line. This evolution is in agreement with our RPA calculations of single-layer graphene.

For each converged state, we calculated the average magnetization per site

$$m = \frac{1}{N} \sum_j \sqrt{\langle S_j^x \rangle^2 + \langle S_j^y \rangle^2 + \langle S_j^z \rangle^2}, \quad (22)$$

where the index  $j$  runs over all sites of the system. We show our results in the right panel of Fig. 5. The magnetization  $m$  is highest at half-filling and for small  $T$ . The transition from the paramagnetic phase to the magnetically ordered phase is a second-order transition within the resolution of our calculations. We also find a smooth magnetization profile for large ranges of doping. In particular, no magnetization jump occurs at the transition from Néel to incommensurate magnetic order, indicating a second-order transition. We see weak fluctuations in  $m$  between the main and secondary magnetic domes, as well as a lack of magnetization below  $T^*$  at large doping

values around  $\delta \approx 0.38$ . Since in the thermodynamic limit the magnetization should be stable up to the white RPA line, we believe these  $m$  fluctuations are due to finite-size effects of our lattice.

## B. Bernal bilayer graphene

In the following we state our results for BBG. In our calculations for BBG, we vary the temperature between  $T = 0$  and  $T = 0.5t$  in steps of  $0.01t$  and the density between 0.5 and 1.5 in steps of  $\frac{1}{144}$ . We again choose an interaction strength of  $U = 3t$  and provide further RPA calculations for  $U = 2.75t$  and  $U = 2.5t$  in Appendix B. We are interested in the possible interaction-induced ordered states that mean-field theory provides to get a comprehensive overview as a function of filling and temperature.

### 1. Phase diagram

In Fig. 6, we show the phase diagram in hole/electron doping and temperature as obtained by our unrestricted real-space Hartree-Fock calculations. Note that in contrast to single-layer graphene, the phase diagram is not perfectly particle-hole symmetric due to a finite  $t_4$  hopping parameter. The RPA critical temperature again coincides well with the critical temperature that we find in the real-space calculations, i.e., the system sizes are a good approximation of the thermodynamic limit. The only exception is a small paramagnetic regime at doping  $\delta \approx -0.1$  below the critical temperature. But we identify this as an artifact of the finite system size. When we repeat our calculations in this region on a  $24 \times 24$  lattice, we find a finite magnetic order parameter (see Appendix C). Around

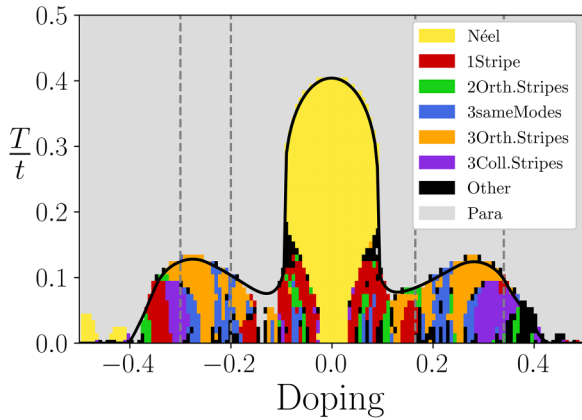


FIG. 6. Mean-field phase diagram for the Hubbard model on Bernal bilayer graphene with  $U = 3t$  as a function of doping and temperature. Calculations were done on  $12 \times 12$  lattices. The black  $T^*$  line was calculated in the thermodynamic limit using RPA. The gray dashed lines represent Van Hove fillings of the noninteracting model at doping values  $-0.299$ ,  $-0.200$ ,  $0.164$ , and  $0.339$ .

half-filling, we see a magnetic dome with relatively high critical temperatures of up to  $T^* \approx 0.40t$  in the phase diagram. As we increase the electron or hole doping, the critical temperature first shrinks to  $T^* \approx 0.08t$ . Then, a secondary dome appears where the critical temperature rises again between 13% and 28% doping, peaking at  $T^* \approx 0.13t$  and finally goes down to 0 at  $\delta \approx \pm 0.4$ . In highly doped BBG, there are two Van Hove energies from the two conduction (valence) bands. On the electron-doped side ( $\delta < 0$ ), the occurrence of the secondary dome coincides with a rise of the density of states and approximate nesting at Van Hove doping. On the hole-doped side, the maximum of  $T^*$  is strongly shifted away from the individual Van Hove points. We suspect the reason of the strong shift to come from interband effects in the particle-hole susceptibility, besides temperature effects and the overlap of the two Van Hove peaks.

The dome around half filling predominantly corresponds to Néel order. At  $T = 0$ , the Néel order seems to extend over a small doping range; however, this is likely due to the finite-size effects, as our system cannot resolve spin orders with very small incommensurabilities. For small dopings and low temperatures, a single mode stripe-ordered region emerges, extending from  $\delta \approx 0.02$  to  $\delta \approx 0.1$  and from  $T = 0$  up to the critical temperature. This region is increased compared to single-layer graphene. For larger dopings  $\delta \gtrsim 0.1$  in the region between the main magnetic dome and the secondary domes, the system undergoes several transitions between different stripe phases. As we describe below the wave vectors of magnetic orders remain stable when we increase the lattice size in our calculations. However, we attribute the absence of clear phase boundaries to finite-size effects.

Within the secondary domes, we see an interplay of the "three-orthogonal-stripe," "three-same-modes," and "three-collinear-stripe" phases with wave vector  $M$  similar to single-layer graphene. While this indicates that also in BBG the fermiology at Van Hove doping with approximate nesting is crucial for the order formation, the competition between the three phases is only resolved within a Hartree-Fock

calculation inside the symmetry-broken phases. In particular, three orthogonal stripes, i.e., chiral spin density waves, cover the largest area in the secondary domes. Three collinear stripes also cover extended regions for even larger hole or electron dopings  $\delta \approx \pm 0.3$ . Generally, when phases extend over such big areas in the phase diagram, it is an indication that they will remain stable in the thermodynamic limit [32]. The splitting of Van Hove points in BBG mainly due to  $t_\perp$  and  $t_3$  becomes more visible for smaller interactions as we show in Appendix B. On the electron-doped side, we find an additional single-mode stripe-ordered regime for dopings even smaller than  $-0.3$ , which is not present on the hole-doped side and thus induced by the particle-hole symmetry breaking  $t_4$ . For very high dopings of  $\delta \pm 0.4$ , we see indications of a two-orthogonal-stripe order. In this area, the two orthogonal stripes have the same amplitude, i.e.,  $S_1 = S_2$  in Eq. (18). Overall, the sequence of magnetic orders is very similar to that of single-layer graphene, in particular on the electron-doped side.

In general we see that the energy separating different types of spin order (e.g., single stripe order vs three collinear stripe order) is smaller than the condensation energy, i.e., the energy gain in comparison to the paramagnetic state. Typically the energy difference between different magnetic orders is of the order of magnitude of less than 10% of the condensation energy.

We also repeated the calculations on  $18 \times 18$  and  $24 \times 24$  lattices along the  $T = 0.04t$  and  $T = 0.05t$  lines to estimate how stable the phase diagram will remain in the thermodynamic limit (see Appendix C). In both of these lattice sizes, we find three collinear stripes, three orthogonal stripes, and the phase with three of the same modes in the secondary domes, in qualitative agreement with Fig. 6. The collinear stripe phases appear stable. The phases of three orthogonal stripes and three same modes have slightly different boundaries for the bigger systems, indicating that their energies are very close and they are competing. We did not find a single-mode stripe-ordered phase for dopings  $\delta \leq -0.3$  on the electron-doped side for the bigger lattices. Instead, the two orthogonal stripes take most of the phase space. Based on these checks, we believe that the overall phase structure will remain stable even for bigger lattices.

## 2. Q dependence

In Fig. 7, we present the  $Q$  dependence of the spin patterns corresponding to the phases in Fig. 6 as a function of doping and temperature. As before, we show the wave vector  $Q$  of the divergence of the RPA bubble and that of the real-space Hartree-Fock calculations given by the Fourier mode of the spin pattern with the biggest weight. We see that within the resolution allowed by our  $12 \times 12$  lattice, at the critical temperature the  $Q$  vectors of the RPA and the real-space Hartree-Fock calculations coincide, as expected.

Close to half filling, we see a wave vector of  $Q = 0$  for the Néel order. Generally, we then see an increase of  $|Q|$  as we increase electron or hole doping. Within our resolution, the transition from Néel order to the adjacent single-mode stripe phase is consistent with a second-order transition as  $Q$  increases by the smallest possible value allowed by the

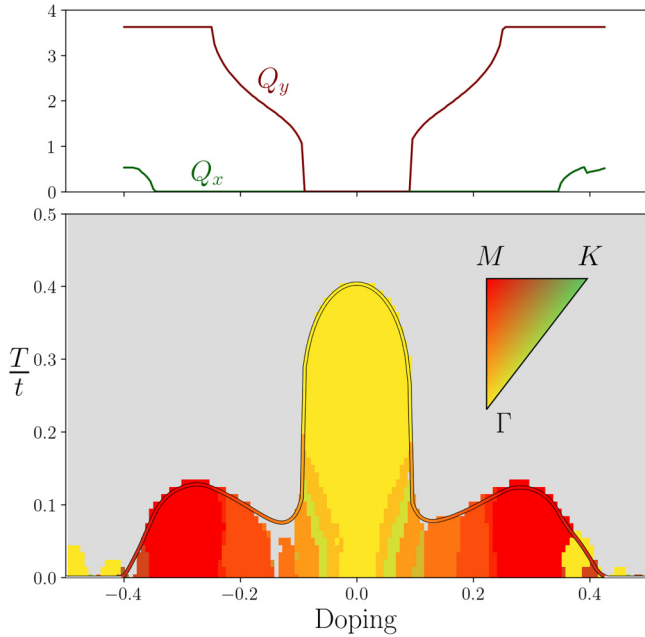


FIG. 7. Upper panel: The  $\mathbf{Q} = (Q_x, Q_y)$ , where we find a divergence of the RPA bubble in the thermodynamic limit. When the green line is zero, the divergence occurs at the  $\Gamma$ - $M$  line. For large electron or hole dopings, the divergence occurs at the  $M$ - $K$  line. Lower panel: The main  $\mathbf{Q}$  of the spin orders in real space. The color of the outline was calculated using RPA and corresponds to the upper panel.

finite-size system. As a function of temperature, we note an increase of  $|\mathbf{Q}|$  as we lower the temperature in this region around half filling. We then have a short doping region ( $\delta \approx 0.05$  to  $\delta \approx 0.1$ ) where wave vectors change back and forth between  $\Gamma$ - $K$  and  $\Gamma$ - $M$  lines indicating that the energies of both magnetic orders are very close together. This reflects the changes between different phases we described above in this region of the phase diagram. While we cannot completely rule out finite-size effects, the  $\mathbf{Q}$  on the  $\Gamma$ - $K$  and  $\Gamma$ - $M$  lines remain stable in our checks on bigger lattices ( $18 \times 18$  and  $24 \times 24$ ). At the critical temperature, the dominant  $\mathbf{Q}$  must lie on the  $\Gamma$ - $M$  line in this region, as suggested by the RPA results (upper panel of Fig. 7).

For dopings  $\delta \geq 0.1$ , the wave vector settles along the  $\Gamma$ - $M$  line corresponding to an instability toward incommensurate spin density waves. At a doping of roughly  $\delta \pm 0.25$  in the Van Hove region,  $\mathbf{Q}$  reaches the  $M$  point and the magnetic order becomes commensurate. At even larger dopings  $\delta \geq 0.35$ , the ordering wave vector within RPA becomes again incommensurate close to  $M$  along the  $M$ - $K$  line. While this agrees with the Hartree-Fock results for electron doping, we find  $\mathbf{Q} = \mathbf{0}$  ferromagnetism for hole doping instead. However, when we repeat the calculation on bigger lattices, we obtain  $\mathbf{Q} = M$  or close to the  $M$ - $K$  line, in agreement with the predictions of the RPA. The reason why we find ferromagnetism in this region of the phase diagram is that a  $12 \times 12$  lattice cannot resolve the  $M$ - $K$  line sufficiently well and therefore  $\mathbf{Q} = \mathbf{0}$  ferromagnetic order is energetically the next best state. In the thermodynamic limit, we therefore do not expect ferromagnetic order to prevail. This is also why we have classified it as "other" order in the phase diagram.

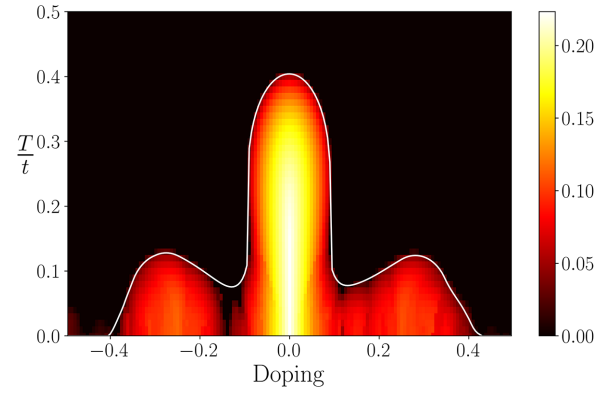


FIG. 8. Average magnetization  $m$  per site in Bernal bilayer graphene for  $U = 3t$  as a function of doping and temperature. The amplitudes are calculated using unrestricted Hartree-Fock and the white outline using RPA.

### 3. Magnetization

For each converged state, we calculated the average magnetization per site according to Eq. (22). We show our results in Fig. 8. Similar to the single-layer case, the transition between the paramagnetic and the magnetic regime appears like a continuous second-order transition within our density and temperature resolution. The magnetization takes its maximum at half filling. In general, we do not observe jumps in the magnetization as we transition between different magnetic phases. This includes the transition from the Néel state to the adjacent single-mode stripes. However, we do see fluctuations in  $m$  in the following doping region between the main magnetic dome and the secondary magnetic domes, particularly on the electron-doped side. Since in the thermodynamic limit the magnetization should be stable up to the white RPA line, we believe these  $m$  fluctuations are due to finite-size effects.

### 4. Displacement field

In addition, we also considered the effects of an externally applied displacement field [see Eq. (4)]. We note that we cannot resolve the cascade of transitions observed at smallest density regions as a function of gate voltage. Instead, our motivation comes from intercalated BBG, which can lead to different charge densities in both layers. This can effectively be modeled by a term like Eq. (4) in the Hamiltonian, which translates to different chemical potentials in both layers [72,75]. We perform calculations at  $T = 0.01t$  to work at very low temperatures while having more numerical stability than for  $T = 0$ . We choose a hole-doping range from  $-0.1$  to  $0.5$ . We vary the displacement field from  $D = 0$  to  $D = 2t$  in steps of  $0.02t$  and set  $U = 3t$ .

In Fig. 9 we show a phase diagram of BBG as a function of displacement field and doping. We see that now the two layers show qualitatively different behaviors. At half filling, both layers exhibit Néel order up to displacement fields of  $D \approx 1.3t$ . We see that the lower layer has a wide region of Néel order in the hole-doped regime. In this region, the upper layer instead shows parallel vertical lines of constant phase. These vertical lines of constant phases extend to the  $D = 0$  line diagonally. In the lower layer, we also see these lines

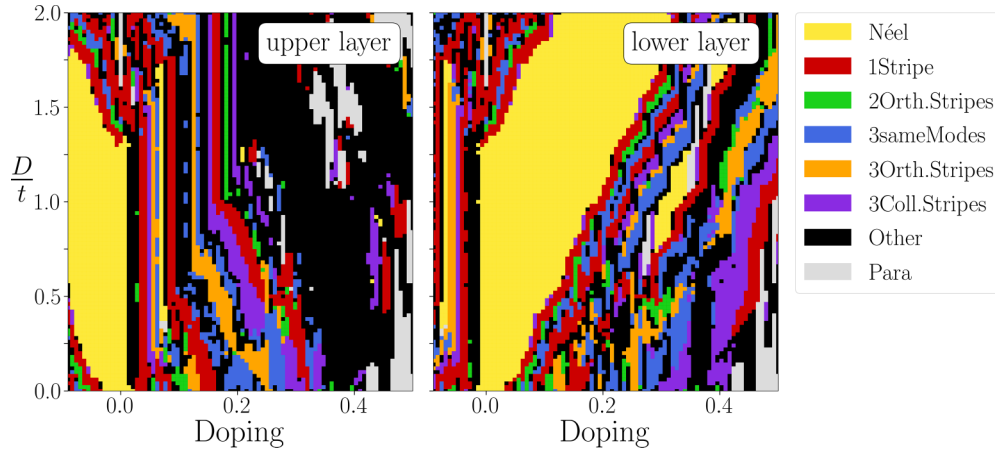


FIG. 9. Phase diagram of BBG for an applied external displacement field at  $T = 0.01t$  and  $U = 3t$  as a function of doping and  $D$ . As before, the doping refers to the total hole doping of the bilayer system. The left (right) panel shows the phase diagram for the sites on the upper (lower) layer. We can qualitatively describe the phase diagram by a cartoon picture of noncoupled graphene sheets, as shown in Fig. 10.

of constant phases in the electron-doped regime. In the hole-doped regime outside the Néel area, we again see diagonal lines of constant phase, this time with the diagonals going toward the top right. We note that the image is fairly pixelated, since we used less convergence time and still face the same finite-size effects as we do for the phase diagrams in doping and temperature. Nevertheless, we believe that the phase diagram gives a qualitatively correct picture of the dominant field effects, whose overall structure can be understood based on the phase diagram of single-layer graphene in Sec. III A. Additional hoppings  $t_{\perp}, t_3, t_4$  add perturbative effects for  $U = 3t$  as we discussed for the case without displacement field.

To this end, we think of the system as two almost decoupled sheets of graphene, which is a good approximation as long as  $t \gg t_{\perp}$ . The displacement field acts on the two layers like a chemical potential with flipped sign. A single layer of graphene in the Néel-ordered state at half filling and  $U = 3t$  has a magnetic gap  $\Delta_{\text{AFM}}$  in the density of states of  $\Delta_{\text{AFM}} \approx 1.3t$  [see Fig. 10(a)]. At half filling, we can see that both layers remain in the Néel state until  $D \gtrsim \Delta_{\text{AFM}}$ . The reason is that, as long as  $D < \Delta_{\text{AFM}}$ , both layers remain half filled [Fig. 10(b)] and thus, exhibit perfect Néel order. When  $D > \Delta_{\text{AFM}}$  exceeds the size of the magnetic gap, the different layers start to obtain different fillings. In this case the lower (upper) layer is filled slightly more (less) than half filling [see Fig. 10(c)]. Therefore, the order changes from a Néel pattern to a single-mode stripe phase following the phase diagram of BBG, Fig. 6.

As we start hole doping the system the upper layer is depleted first, while the lower layer remains half filled [for sufficiently big  $D$  and  $T = 0$ ; see Fig. 10(b)]. Effectively, the upper layer is then doped twice as much as the total system (e.g., when we consider 10% hole doping the upper layer has 20% and the lower layer 0% hole doping). As a consequence, the lower layer remains Néel ordered, while the upper layer shows incommensurate magnetic order. The opposite effect happens as we electron dope the system. All additional electrons populate the lower layer first, while the upper layer remains at half filling. This mechanism is responsible for the

extended Néel regions as well as the vertical lines of constant phases in the phase diagram of Fig. 9.

As we increase the doping even more, both layers become doped; however, with the lower (upper) layer remaining less doped across all hole (electron) dopings. This results in the diagonal lines of constant phases that can be seen in Fig. 9. They follow lines of constant doping of the individual layers. We can also identify a qualitatively similar sequence of phases. There is a fluctuating region of stripe phases with increasing doping, which transitions to larger regions of orders with

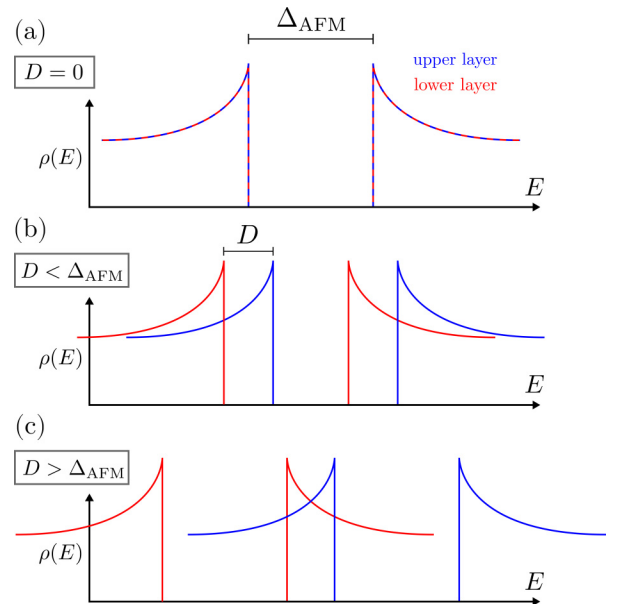


FIG. 10. Cartoon picture of the density of states (DOS)  $\rho$  of two nonconnected layers of graphene. (a) DOS of the system without displacement field. The DOS for the upper and the lower layers are degenerate. Both DOS exhibit a gap  $\Delta_{\text{AFM}}$  due to the Néel order. (b) DOS after applying a small displacement field  $D < \Delta_{\text{AFM}}$ . The DOS of the upper and lower panels are shifted with respect to each other, but the magnetic gap still remains. (c) DOS for  $D > \Delta_{\text{AFM}}$ . There is now no more magnetic gap remaining.

three same modes, three collinear stripes, and three orthogonal stripes.

#### IV. CONCLUSION

We performed unrestricted Hartree-Fock calculations on graphene and BBG lattice structures at moderate interaction strength  $U = 3t$ . The calculations were performed mainly on finite lattices with  $18 \times 18$  unit cells for single-layer graphene and  $12 \times 12$  unit cells for BBG. Bigger lattice sizes were used for several selected points to double-check our results and estimate finite-size effects. Additionally, we performed RPA calculations directly in the thermodynamic limit for spin orders close to the critical temperature  $T^*$ . We classified the resulting states based on their magnetic pattern, calculated their average magnetization, and showed the dominant Fourier weight of the spins.

We found that Néel order is the dominant spin order close to half filling for both single-layer and Bernal bilayer graphene consistent with previous studies [39–52]. Additionally, we studied the full phase diagram as the system gets doped away from half filling. We found that different stripe orders emerge with wave vectors along the  $\Gamma$ - $K$  and  $\Gamma$ - $M$  lines. In a second magnetic dome at  $|\delta| \approx 0.3$ , there is a competition of three orthogonal stripes, three collinear stripes, and other phases with three Fourier modes with the same amplitude. These correspond to the previously discussed chiral (three orthogonal stripes) and collinear (three collinear stripes) spin density waves at Van Hove filling [59–62]. We showed that they extend away from Van Hove filling and mapped out the regimes in doping and temperature where they appear.

The qualitative shape of the phase diagram of single-layer graphene and BBG is very similar, as well as the magnetic incommensurability  $Q$  and average magnetizations. We found a smooth magnetization profile and a smooth variation of the incommensurability. A commensurate  $Q = M$  emerges in an extended region in the secondary domes.

We also studied the effect of an external displacement field  $D$  on BBG. At the same time, the displacement field can also be understood as modeling unequal potentials in both layers, which is an effect that can occur in intercalation in BBG [72,75]. Motivated by this, we calculate a phase diagram for high doping levels as a function of  $D$ , which can qualitatively be explained by modeling BBG as two almost disconnected sheets of single-layer graphene.

In future works, it will be interesting to extend the Hartree-Fock analysis of this paper to clarify the exact nature of the phase boundaries, especially in the secondary dome. This can be done by limiting the allowed spin orders to the dominant spin orders that we found here and work directly in momentum space. This can also enable an investigation of the very small doping regime relevant for the recently observed cascade of phase transitions [77–79]. Finally, spin fluctuations can be taken into account by transforming the (long-range ordered) magnetic state into a fluctuating magnet with short-range (and potentially topological) order. This can be achieved by extending SU(2) gauge theories, incorporating Néel- and spiral-ordered chargons [34–36], to the new lattice geometry and also considering magnetic stripe order. Experimentally, the different magnetic phases we find can be detected by

scanning tunneling measurements, where a direct comparison with the Fourier modes and real-space charge patterns, which follow the magnetization of the local spin patterns, is possible. In this context, it will be interesting to further analyze the effect of extended Van Hove singularities observed in intercalated graphene samples [58,73,74,76,93]. Our results demonstrate that intercalation of graphene and BBG can lead to interesting interaction-induced phases.

#### ACKNOWLEDGMENTS

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#### DATA AVAILABILITY

The data that support the findings of this article are openly available [94].

#### APPENDIX A: CHEMICAL POTENTIAL OF BBG

We here give an overview of the chemical potential of BBG. The chemical potential for single-layer graphene behaves qualitatively similar.

In Fig. 11, we show the chemical potential we find from our real-space Hartree-Fock calculations as a function of doping and temperature. We subtracted a factor of  $\frac{U}{2}$  so that we find a zero crossing point at half filling. The chemical potential increases for decreasing doping (i.e., increasing  $n$ ). We see a steep drop of the chemical potential for low temperatures close to half filling due to the antiferromagnetic gap. This drop gets washed out for higher temperatures. In Fig. 12, we show the chemical potential as a function of doping for different constant temperatures. Within the resolution of our finite-size calculations, the behavior of  $\mu$  as a function of doping is monotonous, therefore not indicating a phase separated region.

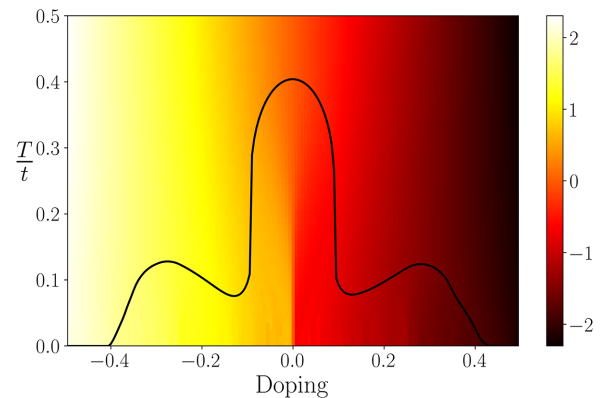


FIG. 11. The shifted chemical potential  $\frac{\mu - U/2}{t}$  for BBG as a function of temperature and doping, as calculated for  $U = 3t$  on the  $12 \times 12$  lattice. The black line is the critical temperature as calculated in RPA.

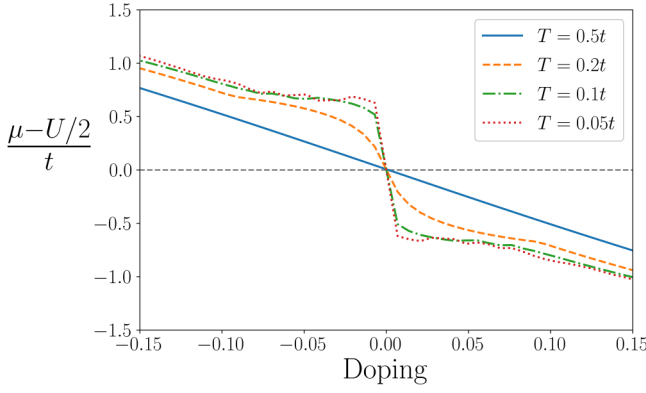


FIG. 12. The shifted chemical potential  $\frac{\mu - U/2}{t}$  for BBG as a function of doping for different temperatures, as calculated for  $U = 3t$  on the  $12 \times 12$  lattice. The solid blue line is calculated at  $T = 0.5t$ , the orange dashed line at  $T = 0.2t$ , the green dash-dotted line at  $T = 0.1t$ , and the red dotted line at  $T = 0.05t$ . The gray dashed line at  $\mu - U/2 = 0$  serves as a guide to the eye.

#### APPENDIX B: RPA RESULTS FOR DIFFERENT $U$

To understand how the three peak structure emerges and to investigate the effect of the interlayer hoppings  $t_\perp$ ,  $t_3$ , and  $t_4$ , we have calculated the critical temperature lines via RPA for different interaction strengths.

In Fig. 13, we show the critical temperatures for BBG hopping parameters and interaction strengths  $U = 2.5t$ ,  $U = 2.75t$ , and  $U = 3t$ . In Fig. 14 we show the critical temperatures for the same  $U$ , but this time with the interlayer hoppings set to zero, corresponding to single-layer graphene.

We see that for lower  $U$  the magnetic domes are strictly disconnected. As  $U$  increases, all three domes increase in

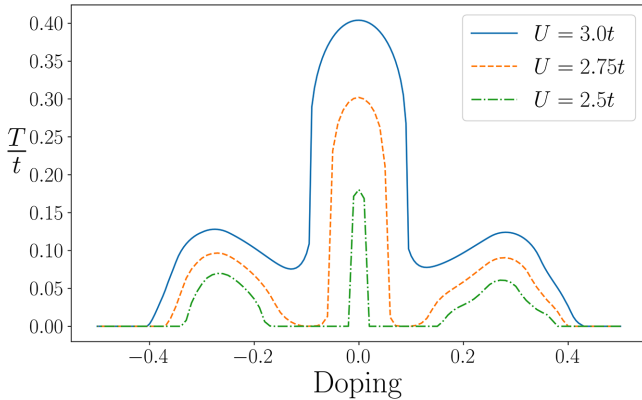


FIG. 13. Critical temperatures for BBG hopping parameters and different interaction strengths. The solid blue line is the critical temperature for  $U = 3t$ , which was already presented in the main text, the orange dashed line is the critical temperature for  $U = 2.75t$ , and the green dash-dotted line is the critical temperature for  $U = 2.5t$ .

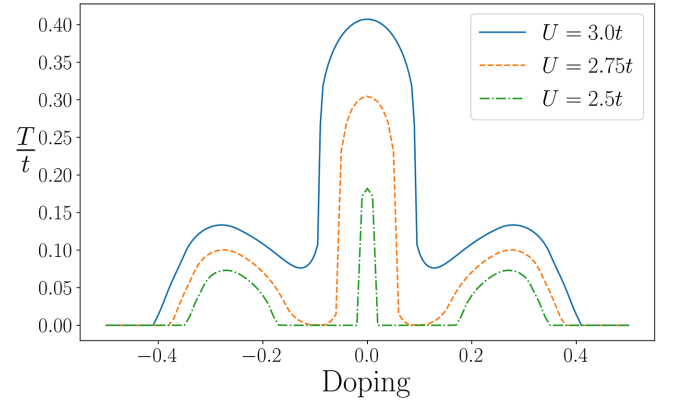


FIG. 14. Critical temperatures for single-layer graphene hopping parameters and different interaction strengths. The solid blue line is the critical temperature for  $U = 3t$ , which was already presented in the main text, the orange dashed line is the critical temperature for  $U = 2.5t$ , and the green dash-dotted line is the critical temperature for  $U = 2t$ .

width and critical temperature and eventually they merge into the three peak magnetic structure for  $U = 3t$ , which we have described in the main text.

One can also notice the effect that the hopping  $t_4$  has on the particle-hole symmetry breaking. In particular for the green dash-dotted curve, one can see that the secondary domes have different shapes on the electron- and hole-doped sides, with the dome on the hole-doped side being slightly flatter and wider.

#### APPENDIX C: HARTREE-FOCK RESULTS FOR BBG ON BIGGER LATTICES

We repeated the real-space HF calculations for BBG on different lattice sizes. Since big lattices take a very long time to compute, we have only calculated the magnetic phases for  $T = 0.04t$  and  $0.05t$ . In Fig. 15, we present the phases along this strip for system sizes  $12 \times 12$ ,  $18 \times 18$ , and  $24 \times 24$ . One can see that the Néel-ordered phase around half filling remains stable; however, it slightly shrinks, since the bigger lattices can host smaller  $\mathbf{Q}$  values, so the commensurate-incommensurate transition can occur already for smaller dopings. Furthermore, the paramagnetic region around  $\delta \approx -0.13$  disappears for bigger lattice sizes, in agreement with the RPA critical temperature result, which corresponds to the thermodynamic limit. The delicate interplay between the different magnetic orders remains also for bigger lattice sizes. In the strongly electron-doped side, the two orthogonal stripe order seems to take more space in the phase diagram for bigger lattices.

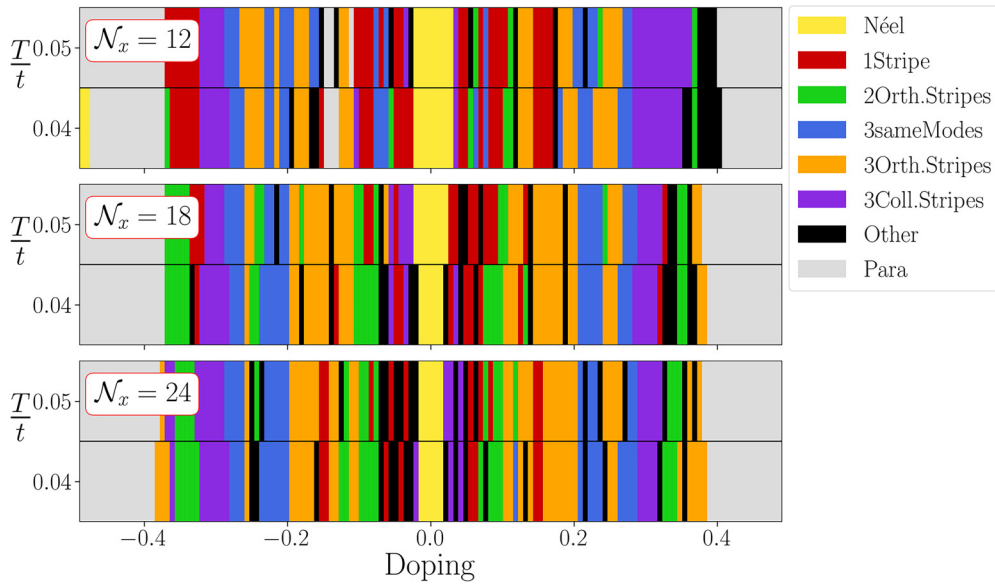


FIG. 15. The phase diagrams for BBG along the  $T = 0.04t$  and  $T = 0.05t$  line for lattice sizes  $12 \times 12$ ,  $18 \times 18$ , and  $24 \times 24$ .

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- [94] The code is publicly available at <https://github.com/RoScholle/BilayerGraphene>.