

Charge-density waves and stripes in quarter metals of graphene heterostructures

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Motivated by recent experiments, here we identify valley-coherent charge-density wave (VC-CDW) order in the non-degenerate quarter-metal for the entire family of chirally-stacked n layer graphene, encompassing rhombohedral multi-layer, Bernal bilayer, and monolayer cousins. Besides the hallmark broken translational symmetry, yielding a modulated charge-density over an enlarged unit-cell with a characteristic $2\mathbf{K}$ periodicity, where $\pm\mathbf{K}$ are the valley momenta, this phase lacks the three-fold (C_3) rotational symmetry but only for even integer n . The VC-CDW then represents a stripe order, as observed in hexalayer graphene [arXiv:2504.05129], but preserves the C_3 symmetry for odd n as observed in trilayer graphene [Nat. Phys. **20**, 1413 (2024) and arXiv: 2411.11163]. From a universal Clifford algebraic argument, we establish that the VC-CDW and an anomalous Hall order can lift the residual valley degeneracy of an antiferromagnetically ordered spin-polarized half-metal, when these systems are subject to perpendicular displacement fields, with only the latter one displaying a hysteresis in off-diagonal resistivity, as observed in all the systems with $2 \leq n \leq 6$. We showcase a confluence of VC-CDW and anomalous Hall orders within the quarter-metal, generically displaying a regime of coexistence, separating the pure phases.

Introduction. A periodic modulation of charge, realized at the cost of the underlying lattice translational symmetry gives rise of a novel quantum state of matter, known as the charge-density wave (CDW). Such a phase can result from electron-phonon interaction in weakly correlated systems or from electronic repulsions in correlated materials. Furthermore, when such a density modulation breaks crystallographic discrete rotational symmetry, an analog of the smectic order is realized in electronic liquids, also known as the stripe phase [1–8]. In this work, we identify the footprints of such CDW and stripe orders in the so called quarter-metal phase, devoid of the spin and valley degeneracies, in chirally-stacked n layer graphene, a family that encompasses monolayer ($n = 1$), Bernal bilayer ($n = 2$), and rhombohedral multi-layer ($n \geq 3$) graphene heterostructures (Fig. 1).

Summary. We show that the CDW always breaks the translational symmetry, leading to a charge modulation that is commensurate in an enlarged unit-cell (Fig. 2) with a characteristic $2\mathbf{K}$ periodicity, where $\pm\mathbf{K}$ are the valley momenta, thus representing a valley-coherent (VC) order. Additionally it breaks the three-fold rotational (C_3) symmetry, but only for $n = 1, 2, 4, 6, \dots$ yielding a stripe phase therein as observed in hexalayer graphene [9]. By contrast, the C_3 symmetry remains preserved when $n = 3, 5, \dots$ as observed via the scanning tunneling microscopy in trilayer graphene [10, 11]. Our proposed universal mechanism for the cascade of degeneracy lifting due to the simultaneous presence of competing orders, ultimately leading to the identification of such a VC-CDW order in the quarter-metal, rests on the following robust and general Clifford algebraic argument. If the electronic bands possess a 2^N -fold degeneracy, then its 2^M -fold degeneracy can be lifted in the simultaneous presence of $M + 1$ number of orders for which the corresponding matrix operators mutually

commute, where $M \leq N$ and $N = 2$ due to the valley and spin degrees of freedom for any n . Thus, in a half-metal (quarter-metal), requisite 2-fold (4-fold) degeneracy lifting is accomplished in the simultaneous presence of two (three) mutually commuting orders. Specifically, we recognize that in the presence of perpendicular displacement (D) field-induced layer polarization, an anti-

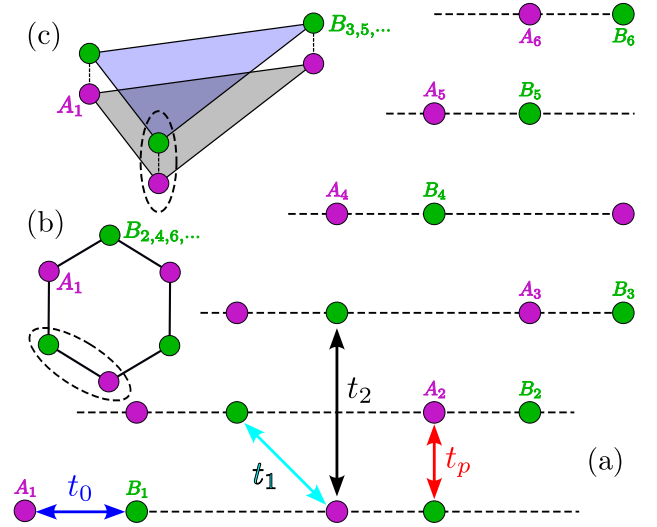


FIG. 1. (a) Lattice structure of chirally-stacked n layer graphene, where A_j (magenta) and B_j (green) correspond to the sites belonging to the A and B sublattice of the honeycomb lattice on the j th layer. In a system with n layers, the low-energy two-band model near each valley and for each spin projection is constituted by the Wannier states localized on A_1 and B_n sites. Hopping processes are denoted by double-headed arrows. The emergent lattice, constituted by low-energy sites, form (b) a honeycomb lattice when $n = 2, 4, 6, \dots$ and (c) a prism-like lattice for $n = 3, 5, \dots$, where the two-site unit cells are shown by dashed closed loops.

ferromagnetic order causes spin degeneracy lifting, yielding a valley-degenerate half-metal, while an additional formation of the VC-CDW or an anomalous Hall order (AHO) gives rise to the non-degenerate quarter-metal.

Experiments. Graphene-based electronic liquids are endowed with valley and spin degeneracies that also raises the possibility of realizing a plethora of broken symmetry phases that break some discrete and/or continuous symmetries of the normal state [12–16]. Such ordering tendencies receive an enhanced propensity with increasing number of layers (n) in chirally-stacked heterostructures, as the density of states therein scales as $|E|^{2/n-1}$. Indeed a wave of recent experiments has identified clear signatures of a cascade of spontaneous symmetry breaking in all these systems with $2 \leq n \leq 6$, especially when they are subject to perpendicular displacement electric (D) fields and gradually doped away from the charge-neutrality point (CNP) [9–11, 17–34]. By contrast, electronic liquid in monolayer graphene continues to feature nodal Dirac fermions without any symmetry breaking due to the linearly vanishing density of states therein [35–37]. See, however, a recent experiment, reporting a relativistic Mott transition in designer graphene [38].

Experimental findings allow us to construct a global phase diagram for systems with $2 \leq n \leq 6$. At relatively high doping, the system is described by a metallic phase, possessing four-fold valley and spin degeneracy. While at moderate doping the system loses the spin degeneracy, yielding a metallic state with only two-fold valley degeneracy, named half-metal, at low doping even the residual valley degeneracy gets lifted, yielding a non-degenerate quarter-metal; features clearly recognized from quantum oscillation frequencies. Such a systematic degeneracy lifting yielding fractional metals results from spontaneous breakdown of symmetries, causing nucleation of various ordered states in the system. Furthermore, superconductivity has been observed in the global phase diagram, primarily near metal and half-metal. These experimental observations caused a surge of theoretical works geared toward unfolding the nature and microscopic origins of various ordered phases and the pairing symmetries [39–68]. After a theoretical proposal [56], more recently superconductivity in the close proximity to the quarter-metal has also been observed [9, 24, 27], which due to the spin- and valley-polarized nature of the quasiparticles must correspond to an odd-parity chiral pair-density wave, triggering new theoretical interests [69–75].

Model. We consider a minimal tight-binding description for free-fermions in chirally-stacked n layer graphene (Fig. 1). We take into account hopping amplitudes between the sites from two sublattices living on the same layer (t_0), yielding massless Dirac fermions and on the adjacent layers (t_1), yielding a momentum-dependent trigonal warping. We also account for two vertical hopping amplitudes between the sites from different sublattices, otherwise living on the adjacent layers (t_p), responsible

for the band splitting and on the second-adjacent layer (t_2), yielding a momentum-independent trigonal warping [76–80]. We neglect hopping between the sites from the same sublattice, yielding a momentum-dependent particle-hole asymmetry which we trade in terms of the chemical potential (μ), measured from the CNP. The effect of the D field enters the Hamiltonian in terms of an on-site potential on the m th layer of an n -layer system $V_{nm} = V[1/2 - (m-1)/(n-1)]$ with $m \leq n$, where $V = Dd(n-1)$ and d is the inter-layer distance.

The Bloch Hamiltonian in all the systems at a lattice momentum $\mathbf{q}$ can then be cast as a block matrix

$$H_{n \text{ layer}}(\mathbf{q}) = \begin{pmatrix} H_{LL} & H_{LH} \\ H_{HL} & H_{HH} \end{pmatrix}(\mathbf{q}), \quad (1)$$

where $H_{LL}(\mathbf{q})$ is a 2×2 matrix, operative over the low-energy sites (A_1 and B_n) and $H_{HH}(\mathbf{q})$ is a $(2n-2) \times (2n-2)$ matrix acting on the high-energy sites ($A_{2,\dots,n}$ and $B_{1,\dots,n-1}$) on which the split-off bands live predominantly. Here, A_m (B_m) corresponds to the sites belonging to the A (B) sublattice on the m th layer. See Fig. 1. The coupling between these two sets of sites is captured by $H_{LH}(\mathbf{q})$ and $H_{HL}(\mathbf{q}) \equiv H_{LH}^\dagger(\mathbf{q})$. To arrive at the effective low-energy description, we integrate out the high-energy sites, leading to the renormalized Hamiltonian $H_{n \text{ layer}}^{\text{renor}}(\mathbf{q}) = H_{LL}(\mathbf{q}) - H_{LH} H_{HH}^{-1} H_{HL}(\mathbf{q})$ [13, 81, 82]. The continuum model is derived by expanding each component of $H_{n \text{ layer}}^{\text{renor}}(\mathbf{q})$ to the leading order in a small momentum $\mathbf{k}$ around each valley with $\mathbf{q} = \pm \mathbf{K} + \mathbf{k}$. Inclusion of the spin degrees of freedom leads to a mere doubling due to a negligibly small spin-orbit coupling for light carbon atoms. We then arrive at the final form of the low-energy Hamiltonian $H_{n \text{ layer}}^{\text{renor,low}}(\mathbf{k}) = \sigma_0 \otimes [H_n(\mathbf{k}) - \mu]$, where $\{\sigma_\nu\}$ is a set of Pauli matrices operating on the spin index with $\nu = 0, \dots, 3$, $\otimes$ stands for the Kronecker product, and $H_n(\mathbf{k}) = \Gamma_{kj} d_{kj}^n(\mathbf{k}) + \Gamma_{03} d_{03}^n(\mathbf{k})$ with $k = 0, 3$ and $j = 1, 2$. A summation over the repeated indices is assumed. Lengthy expressions for $d_{\nu\rho}^n(\mathbf{k}) \equiv d_{\nu\rho}^n$ (for brevity) are not instructive, but displayed in the Supplemental Material (SM) [83]. Four-component Hermitian matrices are $\Gamma_{\nu\rho} = \tau_\nu \otimes \eta_\rho$, where the set of Pauli matrices $\{\tau_\nu\}$ ($\{\eta_\rho\}$) operates on the valley (sublattice or layer) index. Due to the valley inversion symmetry, generated by $\sigma_0 \otimes \Gamma_{10}$ under which $\mathbf{k} \rightarrow (-k_x, k_y)$ [36], for any momenta either $d_{01,32}^n(\mathbf{k})$ or $d_{31,02}^n(\mathbf{k})$ are finite.

Metal. While $d_{01,02,31,32}^n$ account for all the hopping processes, d_{03}^n stems from the D field, causing a layer polarization of electronic density and its k -dependence gives rise to a sagging of electronic bands, yielding annular (simply connected) Fermi rings at low (moderate) doping. As the d_{03}^n term anticommutes with the rest of the matrices in $H_n(\mathbf{k})$, the resulting layer polarization stands as an externally-induced mass order for gapless chiral fermions, producing an isotropic gap near the CNP. At this stage, the electronic bands are four-fold degenerate, with the energy spectrum $\pm[(d_{01/31}^n)^2 + (d_{32/02}^n)^2 +$

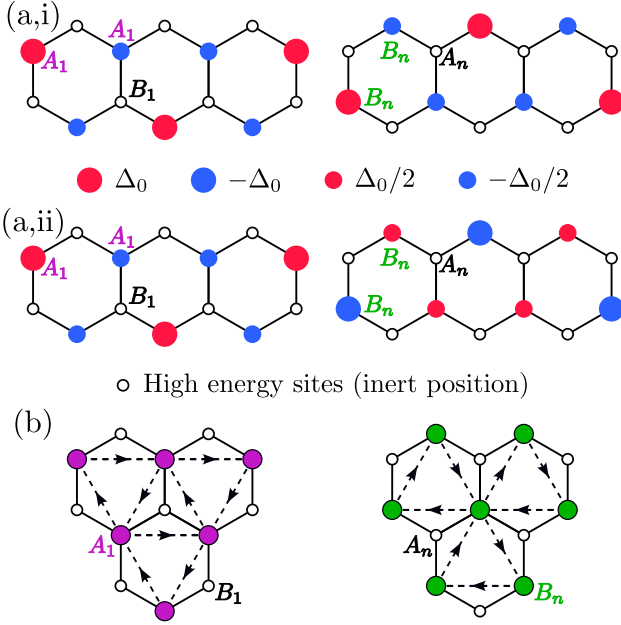


FIG. 2. (a) Lattice realization of the valley-coherent charge-density wave of real amplitude Δ_0 with (i) in-phase and (ii) out-of-phase modulations from the sites from the A_1 and B_n sublattices (Fig. 1). (b) Intra-sublattice/layer circulating currents in the direction of arrows, yielding the anomalous Hall order. The high-energy inert B_1 (A_n) sites in the bottom (top) layer are represented by open black circles. For $n > 2$, all the sites in intermediate layers are inert.

$(d_{03}^n)^2]^{1/2} - \mu$, where $\pm$ corresponds to conduction and valence bands, respectively. Hence, for any $\mu > \min. (d_{03}^n)$ the system describes a four-fold degenerate metal. Next we search for possible channels of symmetry breaking, leading to the formation of fractional metal.

Half-metal. Following the general principle of degeneracy lifting, outlined earlier, we proceed to search for an order for which the matrix operator *commutes* with $\sigma_0 \otimes \Gamma_{03}$ (accompanying d_{03}^n), such that its simultaneous presence with the D field induced layer-polarized mass order lifts the spin degeneracy from electronic bands. We note that among various competing orders, nucleation of the ones for which the matrix operator fully *anticommutes* with the Hamiltonian for gapless chiral fermions yields a maximal gain in the condensation energy as then all the states below the Fermi energy get pushed down uniformly. Such orders are named *mass*. With the inclusion of the on-site Hubbard repulsion U , the dominant component among all the finite-range interactions, these systems acquire a propensity toward the formation of an antiferromagnetic mass order, which is represented by $\sigma \otimes \Gamma_{03}$ and features a staggered pattern of electronic spin between two layers/sublattices [16, 47, 58]. As $[\sigma_0 \otimes \Gamma_{03}, \sigma \otimes \Gamma_{03}] = 0$, the condensation of the antiferromagnetic order (with an amplitude $|\Delta_0| = \Delta_0$) in the presence of D field induced

layer polarization lifts the spin degeneracy of electronic bands, leaving their valley degeneracy unaffected (as both orders appear with τ_0). This outcome can be appreciated from the eigenspectrum of the associated effective single-particle Hamiltonian $H_{n \text{ layer}}^{\text{renor, low}}(\mathbf{k}) + \Delta_0 \cdot \sigma \otimes \Gamma_{03}$, given by $\pm[(d_{01/31}^n)^2 + (d_{32/02}^n)^2 + \{d_{03}^n \pm \Delta_0\}^2]^{1/2} - \mu$. The system features a spin-polarized, valley-degenerate half-metal when $d_{03}^n(0) - \Delta_0 < \mu < d_{03}^n(0) + \Delta_0$.

Quarter-metal. We follow the road map of the Clifford algebraic argument for the systematic degeneracy lifting and next search for orders that would lift the residual valley-degeneracy of the half-metal, leading to the formation of a quarter-metal at lower chemical doping, devoid of any degeneracy. Naturally, we must search for the orders whose matrix operator commutes with the ones for layer polarization ($\sigma_0 \otimes \Gamma_{03}$) and antiferromagnet ($\sigma \otimes \Gamma_{03}$). If we restrict such a search within the territory of mass orders, then there exists a *unique* candidate $\sigma_0 \otimes \Gamma_{33}$ which corresponds to the AHO, resulting from intra-sublattice/layer circulating current, originally proposed by Haldane [84], see Fig. 2(b). The intra-layer next-nearest-neighbor Coulomb repulsion (V_2) [16, 47, 58], the next dominant component among all finite range interactions in chirally-stacked layered graphene systems, among spinless fermions (due to the formation of precursor spin-polarized half-metal) is a natural microscopic source for such an order that causes valley polarization (appearing with τ_3). As such an order yields a finite orbital magnetic moment, its nucleation can be identified from a finite anomalous Hall conductivity (σ_{xy}) and a hysteresis in the off-diagonal resistivity (R_{xy}), which have been observed in all systems with $2 \leq n \leq 6$ so far. The valley-degeneracy lifting due to the AHO (with an amplitude Δ_1) can be appreciated from the eigenspectrum of the effective single-particle Hamiltonian $H_n(\mathbf{k}) - \Delta_0 \Gamma_{03} - \Delta_1 \Gamma_{33} - \mu$ for spinless fermions, describing the spin-polarized half-metal, in the simultaneous presence of D field induced layer-polarization, antiferromagnet, and AHO, given by $\pm[(d_{01/31}^n)^2 + (d_{32/02}^n)^2 + \{d_{03}^n - \Delta_0 \pm \Delta_1\}^2]^{1/2} - \mu$. Therefore, when $|d_{03}^n(0) - \Delta_0| - \Delta_1 < \mu < |d_{03}^n(0) - \Delta_0| + \Delta_1$ (assuming $\Delta_0 > \Delta_1$ which is justified as $U > V_2$), the system features a fully polarized quarter-metal.

We, however, notice that besides masses there exists another class of orders that only partially anticommute with the free-fermion Hamiltonian. Although such orders typically tend to be energetically inferior to mass orders, strong electronic interactions in the suitable channels can, in principle, cause their dynamic nucleation. Motivated by recent experimental observations in the quarter-metal of rhombohedral trilayer and hexalayer graphene, reporting a second phase devoid of any σ_{xy} and hysteresis in R_{xy} , we re-search for its additional candidates, without deviating from our guiding Clifford algebraic principle; the corresponding matrix operators must *commute* with

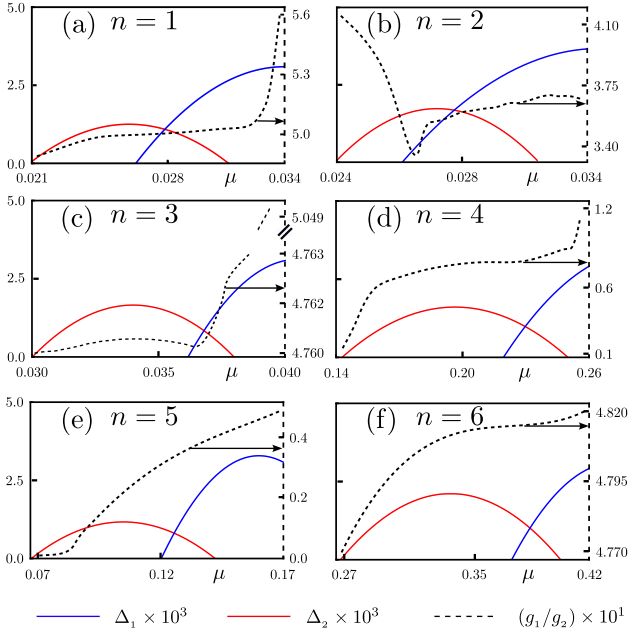


FIG. 3. Solutions of the mean-field gap equation [obtained from Eq. (2)] at a temperature $T = 10^{-5}$ displaying a competition between the anomalous Hall order with amplitude Δ_1 (blue) and valley-coherent charge-density wave with amplitude Δ_2 (red) with numbers on left vertical axes as a function of the ratio of the corresponding coupling constants g_1 and g_2 (black dashed curve with numbers on right vertical dashed axes), respectively, over a range of chemical potential μ in the quarter-metal of chirally-stacked n layer graphene (see legends) [83]. For smallest (largest) value of μ the system yields annular (simply connected) Fermi ring when $2 \leq n \leq 6$. For $n = 1$ the Fermi ring is always simply connected. Appearance of anomalous Hall (valley-coherent charge-density wave) order for large (small) chemical potential is chosen to qualitatively mimic the experimental observation in rhombohedral trilayer ($n = 3$) and hexalayer ($n = 6$) graphene. Here, Δ_1 , Δ_2 , and T are measured in units of t_0 (Fig. 1).

$\sigma \otimes \Gamma_{03}$ and $\sigma_0 \otimes \Gamma_{03}$. We immediately identify *four* candidate matrix operators Γ_{j0} and Γ_{j3} with $j = 1, 2$. Such an order breaks the translational symmetry, generated by Γ_{30} as $\{\Gamma_{j0/j3}, \Gamma_{30}\} = 0$ for $j = 1, 2$ and mixes two valleys (appearing with off-diagonal Pauli matrices $\tau_{1,2}$ in the valley space). Hence, the resulting state is formed via a coherent superposition of the fermionic states localized near two valleys. Now depending of η_0 or η_3 , we find either in-phase or out-of-phase fermionic density modulations between the layers/sublattices with a characteristic periodicity of $2\mathbf{K}$, as show in Fig. 2(a), which causes an enlargement of the unit cell, yielding a CDW [83]. Hence, this phase is named VC-CDW.

Furthermore, the VC-CDW displays an intriguing property depending on the number of chirally-stacked honeycomb layers (n). Notice that for $n = 2, 4, \dots$ ($n = 3, 5, \dots$) the low-energy sites form an emergent honeycomb (prism-like) lattice for which the generator of the

C_3 symmetry is Γ_{33} (Γ_{00}). Hence, the VC-CDW order additionally breaks the three-fold rotational symmetry when $n = 1, 2, 4, \dots$ as $\{\Gamma_{j0/j3}, \Gamma_{33}\} = 0$ for $j = 1, 2$ thereby representing a stripe order therein, as reported for rhombohedral hexalayer graphene. By contrast, the same phase preserves the C_3 rotational symmetry (as its generator is *trivial*) when $n = 3, 5, \dots$ as observed in rhombohedral trilayer graphene.

Finally, we address a competition between these two ordered phases in the quarter-metal phase within a mean-field approximation. Then, the free-energy density reads

$$F = \sum_{j=1,2} \frac{\Delta_j^2}{2g_j} - k_B T \sum_{\tau, \rho=\pm} \int_{\mathbf{k}} \ln \left[\cosh \left(\frac{|E_{\tau}^{\rho} - \mu|}{2k_B T} \right) \right], \quad (2)$$

where k_B is the Boltzmann constant, which we set to be unity, $\int_{\mathbf{k}} \equiv \int d^2\mathbf{k}/(2\pi)^2$, Δ_1 (Δ_2) is the amplitude of the AHO (VC-CDW) with the corresponding coupling constant g_1 (g_2), and E_{τ}^{ρ} are the eigenvalues of the effective single-particle Hamiltonian $H_n(\mathbf{k}) + \Delta_1 \Gamma_{33} + \Gamma_{10} \Delta_2$ in the presence of these two orders in the spin-polarized half-metal phase. The coupling constants g_1 and g_2 are treated as phenomenological inputs whose strengths depend on the chemical potential μ in the spirit of the renormalization group, where these quantities are running variables. Minimizing F with respect to Δ_1 and Δ_2 , we arrive at coupled gap equations, which are solved numerically to mimic the experimentally observed phase diagrams in rhombohedral trilayer and hexalayer graphene, suggesting appearance of the VC-CDW (AHO) at lower (higher) doping in quarter-metal [83]. As the matrix operators for these phases mutually *anticommute*, a region of coexistence between them is generically observed that separates two pure phases, see Fig. 3, which we construct by taking parameter values from various experiments.

Discussions. To conclude, from a Clifford algebraic universal protocol for the systematic degeneracy lifting of electronic bands, we have identified two candidate orders for non-degenerate quarter-metals in chirally-stacked n layer graphene, namely VC-CDW and AHO. The VC-CDW (AHO) mixes two valleys (causes valley polarization) and thus breaks (preserves) the translational symmetry. The hallmark anomalous σ_{xy} and hysteresis of R_{xy} in AHO have been observed in the quarter-metal of all the systems with $2 \leq n \leq 6$ [9–11, 17–34]. However, thus far the VC-CDW ordering has been observed in the quarter-metal of rhombohedral trilayer and hexalayer graphene, where the C_3 symmetry is respectively preserved [9] and broken [10, 11], leading to a stripe phase only in the latter system, in agreement with our theoretical findings. As disorder (also breaking the translational symmetry) couple with VC-CDW as a *random field*, their inevitable presence in real materials can be detrimental for the nucleation of such an order [85]. Nonetheless, we are optimistic that with increasing sample quality, the VC-CDW order can be observed in other chirally-stacked graphene

heterostructures. Despite some previous works on the VC-CDW [40, 86], its connection to the quarter-metal and n -dependent stripe phase remained unexplored. In the future, it will be worth identifying an exact microscopic model, favoring the VC-CDW.

Even though no ordering has been observed in pristine monolayer graphene so far, a recent experiment has revealed a relativistic Mott transition into a mass ordered state in designer graphene [38], where a similar cascade of degeneracy lifting can be observed with the following protocol. The analog of layer polarization can be engineered by creating a small density imbalance between two sublattices. On such an engineered band-insulating system, one should observe a cascade of degeneracy lifting when it is doped away from the CNP, similar to the situation in chirally-stacked graphene multi-layer. However, due to the E -linear density of states, the on-site and next-nearest-neighbor repulsions need to be stronger, which can possibly be achieved in designer systems. Optical honeycomb lattices constitute yet another promising platforms where one can observe the cascade of degeneracy lifting and formation of fractional ‘thermal’ metals of neutral atoms, where Hubbard repulsion-driven antiferromagnetism has already been emulated [87]. Thus with an externally induced sublattice staggered potential, the realization of thermal half-metal should be within the reach of existing experimental facilities. Even though tuning next-nearest-neighbor or other finite-range repulsion to a desired strength to lift the residual valley degeneracy can be challenging therein, the requisite AHO and VC-CDW can be engineered externally, as has been achieved with the former one [88]. Therefore, with engineered AHO or VC-CDW, a thermal quarter-metal on Hubbard-dominated optical honeycomb lattices can be observed with a small staggered sublattice potential.

Our proposed Clifford algebraic protocol for systematic band degeneracy lifting should be generically applicable in any multi-band system, among which twisted bilayer graphene is the most prominent one. However, due to sufficiently narrow band width, such a system becomes an insulator upon degeneracy lifting at various integer fillings, as observed in experiments [89–92]. A detailed analysis on such a system is left for a future investigation.

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