

Itinerant magnetism in the triangular-lattice Hubbard model at half doping: Application to twisted transition metal dichalcogenides

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We use unrestricted Hartree-Fock, density matrix renormalization group, and variational projected entangled-pair state calculations to investigate the ground-state phase diagram of the triangular-lattice Hubbard model at “half doping” relative to single occupancy, i.e., at fillings of $(1 \pm \frac{1}{2})$ electrons per site. The electron-doped case has a nested Fermi surface in the noninteracting limit, and hence a weak-coupling instability toward density-wave orders whose wave vectors are determined by Fermi-surface nesting conditions. We find that at moderate-to-strong interaction strengths, other spatially modulated orders arise, with wave vectors distinct from the nesting vectors. In particular, we identify a series of closely competing, itinerant long-wavelength magnetically ordered states, yielding to uniform ferromagnetic order at the largest interaction strengths. For half-hole doping and a similar range of interaction strengths, our data indicate that magnetic orders are most likely absent.

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The triangular-lattice Hubbard model plays a paradigmatic role in studying the interplay between electronic interactions and geometric frustration. At half filling ($\nu = 1$ electron per site), double occupancy is suppressed by Hubbard repulsion; electron spins are then the dominant degrees of freedom, but their ordering is frustrated by the nonbipartite nature of the triangular lattice [1]—a scenario believed to favor the formation of quantum spin liquids. On doping away from half filling, strong-coupling expansion yields a picture of geometrically frustrated magnetic moments coupled to itinerant electrons. The existence and nature of magnetic order in this setting remain a challenging question.

Experimentally, layered materials with triangular-lattice structure [2–8] provide a natural platform for realizing the triangular-lattice Hubbard model (TLHM). More recently, effective extended TLHMs have also been constructed [9] to describe electrons in homo- and hetero-bilayer transition metal dichalcogenide (TMD) moiré materials [10–12]; experimental evidence of strong correlations has recently been reported [12–20]. The TLHM has also been realized in a more controlled setting in cold-atom experiments [21–23].

Most existing theoretical efforts to establish the ground-state phase diagram of the TLHM and related or extended models [24–37] have studied half filling or a small filling range straddling it. A smaller body of work [21,25,38–49] has studied the case of “half electron doping” ($\nu = 3/2$), mostly motivated by experiments described above. Theoretical efforts have also built on the resemblance of the noninteracting problem to that of graphene doped to a filling of $1 \pm 1/4$ electron per site [50–54], namely, a nested Fermi surface whose associated Van Hove singularity signals a weak-coupling instability to broken-symmetry order at the nesting wave vectors. In the half-electron-doped TLHM case, the weak-coupling order is predicted to be an unusual magnetic insulator with tetrahedral spin order [39,55]. The possibility of realizing this exotic broken-symmetry state, and its potential to stabilize chiral superconductivity, have stimulated much experimental and theoretical work.

However, two recent experimental studies of twisted TMDs [12,17]—theoretically modeled as the single-band TLHM model or simple extensions—find no evidence for insulating states at $1/2$ electron doping. This motivates our study of the TLHM ground-state phase diagram at larger interaction strengths, where the weak-coupling assumption is no longer valid. We will also consider the quantum phases at $1/2$ hole doping, demonstrating the clear particle-hole asymmetry of the TLHM.

In the absence of a controlled weak-coupling calculation, we have deployed a variety of numerical variational methods to study the TLHM: (a) unrestricted self-consistent Hartree-Fock [56] (HF), (b) the density matrix

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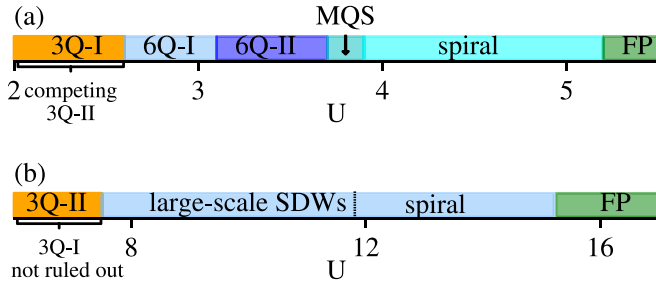


FIG. 1. Ground-state phase diagram of the TLHM as a function of U at $1/2$ electron doping. The magnetic orders of the different phases are illustrated in Table I. The abbreviation nQ stands for spin-density waves with n momentum component. The I stands for noncoplanar, and II stands for collinear. MQS: multi-Q noncoplanar stripe. FP: full spin polarization. (a) Phase diagram obtained from unrestricted Hartree-Fock simulations. (b) Phase diagram inferred from tensor network calculations. The obtained large-scale SDWs include collinear orders akin to $6Q-II$.

renormalization group [57,58] (DMRG), and (c) infinite projected entangled-pair states [59–68] (PEPS). At $1/2$ electron doping we find that beyond a critical interaction strength the proposed insulating spin-tetrahedral state gives way to other magnetic orders with different wave vectors. This is in contrast to the half-filled square-lattice Hubbard model, where the spin-density-wave (SDW) momentum (π, π) is equal to the nesting vector for all interaction strengths. Starting from moderate interaction strengths, our numerics reveal a rich phase diagram hosting a series of different large-scale magnetically ordered metallic states, see Fig. 1. For weaker interactions, we also find a metallic collinear-spin state which competes with the insulating spin-tetrahedral state. At the largest interaction strengths, uniform ferromagnetism emerges. For $1/2$ hole doping, we find no evidence for magnetism up to reasonably strong interactions.

Hamiltonian. We consider the following Hamiltonian on the triangular lattice with nearest-neighbor ($\langle i, j \rangle$) hopping and density-density interactions:

$$H = -\sum_{\langle i, j \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + V \sum_{i \neq j} f(r_{ij}) n_i n_j, \quad (1)$$

where $c_{i\sigma}$ ($c_{i\sigma}^\dagger$) is the annihilation (creation) operator of electron at site i with spin index $\sigma = \uparrow, \downarrow$; $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ ($n_i = \sum_\sigma n_{i\sigma}$) is the density operator of spin- σ electrons (and the total density); and r_{ij} is the distance between site i and j in units of the lattice constant. We are primarily interested in the repulsive Hubbard model $U > 0$, $V = 0$, and will comment on the effects of long-range interaction $Vf(r)$. We focus on the quantum phases at $1/2$ electron doping with respect to half filling, i.e., $\langle n_i \rangle = 1 + \frac{1}{2} = \frac{3}{2}$, where the average (overline) is over all sites. We will also comment on the $1/2$ hole-doping case $\langle n_i \rangle = \frac{1}{2}$.

Spin-density-wave orders. The numerical results we detail below show that magnetic orders are ubiquitous in the $1/2$ -electron-doping phase diagram. These may be parameterized in terms of expectation values of the spin operators $S(j) = \frac{1}{2} \sum_{s,s'} c_{js}^\dagger \sigma_{s,s'} c_{js'}$, where $\sigma = [\sigma_x, \sigma_y, \sigma_z]$ are Pauli matrices. We decompose the corresponding spin textures as

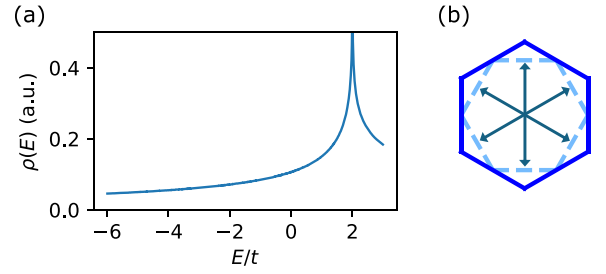


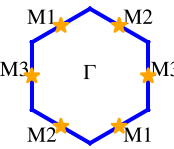
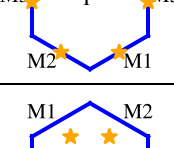
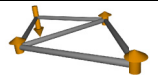
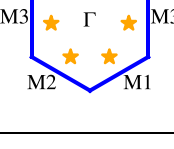
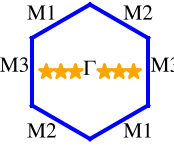
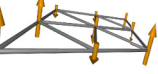
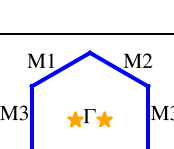
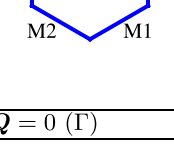
FIG. 2. (a) Density of states for the model at $U = 0$. A Van Hove singularity is located at single-particle energy $E = 2$. (b) Fermi surface (blue dashed line) in the Brillouin zone for $\langle n_i \rangle = \frac{3}{2}$. The Fermi energy is $E_F = 2$. There are three nesting vectors $\mathbf{Q}_{M_i} = -\mathbf{Q}_{M_i}$ modulo reciprocal lattice vectors.

$\langle S(j) \rangle = \sum_s \mathbf{m}(\mathbf{Q}_s) e^{i\mathbf{Q}_s \cdot \mathbf{r}_j}$. Here $\mathbf{r}_j$ is the coordinate of site j : $\mathbf{r}_j = r_{j,1} \mathbf{a}_1 + r_{j,2} \mathbf{a}_2$, with $\mathbf{a}_1$ and $\mathbf{a}_2$ the Bravais lattice vectors. Commensurate spin textures are periodic and have wave vectors of the form $\mathbf{Q}_s = s_1 \mathbf{G}_1 / N_{u,1} + s_2 \mathbf{G}_2 / N_{u,2}$, where $\mathbf{G}_1, \mathbf{G}_2$ are the reciprocal lattice vectors and $s_1, s_2, N_{u,1}$, and $N_{u,2}$ are integers. In the incommensurate case the $\mathbf{Q}_s$ are irrational linear combinations of the reciprocal lattice vectors, in which case there is no periodically repeating unit cell. A prototypical type of incommensurate orders is spin spirals with $\langle S_x(j) + iS_y(j) \rangle \propto e^{i\mathbf{Q}_s \cdot \mathbf{r}_j}$, $\langle S_z(j) \rangle = m_z$, where the spiral is canted if $m_z \neq 0$.

We find ground states via variational optimization of (a) Slater determinants (unrestricted HF), (b) matrix product states (DMRG), and (c) projected entangled-pair states (PEPS). These three numerical methods are complementary. We use HF for a first insight into candidate symmetry-breaking orders. By employing ODA self-consistent optimization [56], we can achieve convergence in total system size (144×144 up to 1024×1024 sites) to high precision and deal with relatively large unit cells (up to 48×48 sites). In addition to our unrestricted HF study, we have also used a “boosted frame ansatz” [69,70] to efficiently study incommensurate spiral states in HF. However, as it is limited to Slater determinants, HF is an uncontrolled approximation and is not guaranteed to provide reliable results, especially at strong coupling. Tensor network methods (DMRG and PEPS), on the other hand, are asymptotically exact methods, as their accuracy can be systematically improved by increasing the number of variational parameters characterized by the bond dimension D . DMRG is system-size limited but can reach very high accuracy. We hence use it to extrapolate to the two-dimensional (2D) thermodynamic limit from accurate small-system calculations. Variational PEPS calculations are performed directly in the 2D thermodynamic limit, with different choices of unit cell. By introducing automatic differentiation gradient descent [67,68] to fermionic PEPS [62], we are able to achieve good optimization efficiency.

$1/2$ electron doping: Hartree-Fock. At $1/2$ electron doping, the noninteracting band structure has Van Hove singularities at the Fermi energy, stemming from saddle points in the dispersion at the three M points (Fig. 2). Previous work [39] has built on this to propose the emergence of a weak-coupling “3Q” magnetic order characterized by the three independent choices of wave vector $\Delta \mathbf{Q}_{ij} \equiv \mathbf{Q}_{M_i} - \mathbf{Q}_{M_j} = \epsilon_{jik} \mathbf{Q}_{M_k}$ connecting

TABLE I. Hartree-Fock magnetic orders. Wave vectors $\mathbf{Q}_s$ of the magnetic orders are denoted as stars in the Brillouin zone.

Names	Momentum $\mathbf{Q}$	Magnetic unit cell
3Q-I		
3Q-II		
6Q-I		Incommensurate noncoplanar order
6Q-II		
MQS		Commensurate noncoplanar order
Spiral		Incommensurate coplanar order, here show an example with period 8.
FP	$\mathbf{Q} = 0$ (Γ)	

the M points $\mathbf{Q}_{M_i}$, $i = 1, 2, 3$, since $\mathbf{Q}_{M_i} = -\mathbf{Q}_{M_i}$ modulo reciprocal lattice vectors. The 3Q order leads to a 2×2 enlarged unit cell in real space. This state, that we term “3Q-I” for reasons that will be clear shortly, is a Chern insulator with tetrahedral spin order (Table I) and was found in restricted HF calculations [39]. Subsequently, it was found that within HF ferromagnetic states have a lower energy than the 3Q-I state [42,44,55] for $U > 3.5$, suggesting the breakdown of the weak-coupling picture.

Here, we find that at intermediate interaction strengths several other magnetically ordered metallic states appear between a 3Q order and the ferromagnet. This results in a remarkably rich phase diagram, with the striking trend that the ordering vectors appear to systematically evolve from M to Γ with increasing U . The evidence for large-scale spin textures is first obtained from our unrestricted HF calculations. In our HF phase diagram [Fig. 1(a)], we find the 3Q Chern insulator is further restricted to the range $U < 2.5$. In this region we also find a competing 3Q collinear state (Table I) that we dub “3Q-II,” with $\lesssim 10^{-4}$ higher energy per site. This competing collinear state is distinct from that proposed in Refs. [39,40], as it has net spin polarization and also a charge modulation induced by the spin order, though both are an order of magnitude smaller than the spin modulation. Similar orders have also been found in renormalization group (RG) studies [53], and the collinear state or its unpolarized sibling

has been argued to be favored over the 3Q insulator at finite temperatures in both mean-field and RG studies [40,53].

As U is increased further, the 3Q tetrahedral order is first replaced by the “6Q-I” state: a noncoplanar spin texture with ordering wave vectors shifted away from the M points toward Γ . At still larger U , the 6Q-I order gives way to the 6Q-II order, which is collinear and has a small net magnetization and small induced charge density modulation. Our data obtained on different momentum grids indicate that the 6Q-II order is most likely commensurate and has a fixed 3×3 unit cell, and can be considered as a larger unit cell version of the competing 2×2 collinear state (the 3Q-II state) found at small U .

Upon further increasing U , a multi-Q noncoplanar stripe (MQS) takes over from the 6Q-II state. The MQS state has an 8×1 unit cell and is found to have the lowest energy for a short interval of U . It has three pairs of ordering wave vectors $\pm \mathbf{Q}_i$. For even larger U , an incommensurate single-Q uncanted spiral phase is found to be favored until $U \sim 5.2$. Within the spiral region, the ordering wave vector varies continuously toward Γ , approaching it very closely for $U \sim 5.2$, after which the fully polarized (FP) ferromagnetic state becomes more favored. While the ordering wave vector of the spiral is still located on the $\Gamma - M$ lines, the energy density difference by rotating to other momentum directions can be as small as 10^{-4} . The Slater determinant of the FP state is an exact eigenstate of H . Since HF is variational, the region of full spin polarization is therefore bounded below by $U \sim 5.2$.

Except for the 3Q-I tetrahedral state, all other states are found to be metallic. By the Lieb-Schultz-Mattis (LSM) theorem [71–73], and assuming there is no topological order, states with a fractional charge per unit cell must be metallic. For circular spiral orders, a generalized LSM theorem [69] further states that a fractional charge per site leads to metallic behavior. Of the states found in HF, only the 3Q-I (spin-tetrahedral), 3Q-II (collinear), and the MQS states can in principle be insulating according to the (generalized) LSM theorem. Nevertheless, we find the latter two states to be metallic.

1/2 electron doping: DMRG. We now go beyond mean-field theory and numerically test the predictions of HF with tensor network methods. The inferred schematic phase diagram obtained from both DMRG and PEPS is shown in Fig. 3(a). Compared to HF, the phases and their boundaries are shifted to stronger interactions, in line with the expectation for comparing mean-field results with those of more controlled methods [74,75].

We use DMRG to obtain ground states on small systems (a 36-site triangular flake, a 48-site hexagonal flake, and a finite cylinder). The results indicate strong-coupling ferromagnetism and demonstrate the challenges of studying the possibility of SDW orders at weaker interactions on small system sizes. Our simulations explicitly conserve charge $U(1)$, spin $SU(2)$ [76,77]; transverse momentum [78] conservation is additionally used as validation for cylinder data. Broken-symmetry phases are identified by measuring the spin polarization S_z and correlation functions. The calculated S_z per site as a function of U [Fig. 3(a)] is quite different for different geometries, but the critical U ’s to reach full polarization are in reasonable agreement ($U = 17 \pm 4$). This difference is not very surprising because most large-scale spin

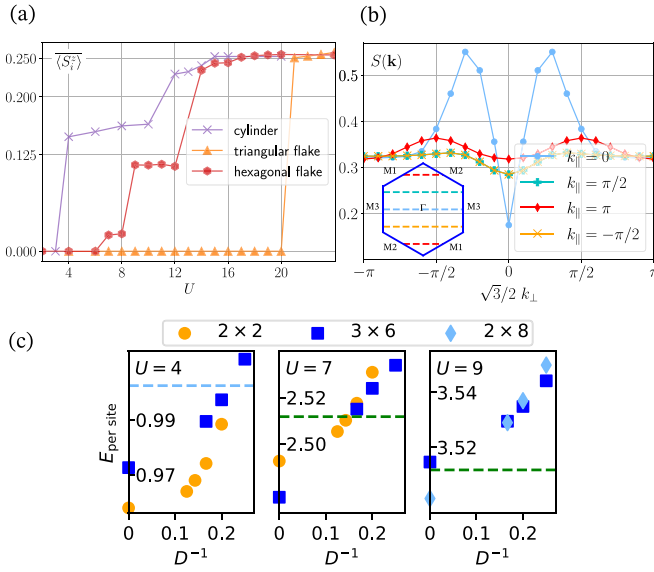


FIG. 3. Tensor network results for 1/2 electron doping. (a), (b) DMRG data. (a) Polarization $\langle S_z(i) \rangle$ (polarization per site) as a function of U for different geometries: four-leg zigzag cylinder with length 20, 36-site triangular flake, and 48-site hexagonal flake. The $\langle S_z(i) \rangle$ is evaluated for the highest-weight ground state averaged without edge sites. (b) Static structural factor $S(\mathbf{k}) = \sum_m e^{i\mathbf{k} \cdot (\mathbf{r}_m - \mathbf{r}_n)} [\langle \mathbf{S}(m) \cdot \mathbf{S}(n) \rangle - \langle \mathbf{S}(m) \rangle \cdot \langle \mathbf{S}(n) \rangle]$ averaged for the central sites m of the cylinder geometry, $U = 8$. The $\mathbf{k}$ can be projected to two orthogonal components $k_\perp$ and $k_\parallel$. The four possible values of $k_\parallel$ correspond to four colored cuts in the Brillouin zone, shown in the insets. (The red dashed lines are along the same cut through M1 and M2.) (c) PEPS variational energy data for different unit cells. The dashed lines denote HF energies for comparison, with full spin polarization at $U = 7, 9$. Different PEPS unit cells $m \times n$ are used, which sets the maximal possible unit cells of the obtained states. The spiral-like states obtained for 2×8 PEPS unit cell at $U = 9$ have smaller actual unit cells, i.e., 1×8 . The 2×2 and 3×6 states for $U = 4, 7$ are collinear. We perform extrapolation to the $D^{-1} = 0$ limit based on $E(D) = E_0 + c/D^\alpha$ ansatz [70].

textures found in HF do not fit well on moderately sized DMRG systems. The DMRG results are therefore consistent with the existence of large-scale spin textures in the thermodynamic limit. The estimate for the critical interaction strength for a homogeneous order such as ferromagnetism, on the other hand, is probably less sensitive to finite-size effects. The finite-size limitations can also already be estimated at the HF level. In HF studies on similar small system sizes as used in DMRG, spin textures that emerge on large systems are also absent. However, the deviation of the critical U for ferromagnetism is smaller than 20% relative to larger system sizes. We note that evidence for ferromagnetism at high U for similar fillings has previously also been obtained in small-system data of a simplified $t - J$ model [41], and of a Hubbard model [21,48,79,80].

Our DMRG data for cylinder geometries show the existence of strong, unidirectional, and apparently incommensurate SDW correlations in the static structure factor [i.e., the only obvious peak is found at some “incommensurate” $k_\perp$ for $k_\parallel = 0$ in Fig. 3(b)]. This, combined with our finding of

a nonzero $\langle S^z \rangle$ [Fig. 3(a)], indicates that the DMRG ground state exhibits canted spin spiral order. Correspondingly, HF calculations on four-leg cylinders only find the spiral phase at moderate U , even though the 3Q orders also fit on this geometry. The DMRG results are thus again in agreement with HF, in that spiral orders are present at intermediate U on the cylinder geometry, albeit in a canted form. We attribute the difference between the results obtained on large tori and the thin cylinder to the intrinsically anisotropic nature of the latter, which does not capture the two-dimensional Fermi-surface nesting.

1/2 electron doping: PEPS. In our infinite PEPS calculations, guided by the HF results, we use the following unit cells: 2×2 (to fit 3Q), 3×6 (to fit 6Q-II and as a proxy for spirals), and 2×8 (to fit MQS and as a proxy for spirals). We consider $4 \leq U \leq 20$. A 2×2 state (3Q-II), large-scale SDWs (6Q-II analog, spiral), and then FP states are found from weak to strong interactions.

By increasing D up to 8 we can systematically lower the variational energy of the PEPS states [Fig. 3(c)]. As a proof of improved accuracy, the $U = 4$ data in Fig. 3(c) show that the energies of the 2×2 and 3×6 PEPS are lower than the HF energies. All states with a 2×2 unit cell obtained with PEPS are collinear 3Q-II states (Table I), but with no clear net magnetization. The 3Q-I tetrahedral state with scalar chirality is not found in our calculations. However, we do not consider this state to be ruled out (see Supplemental Material [70]). HF suggests that 3Q-I and 3Q-II are very close in energy, so we take the PEPS finding of 3Q-II as evidence that one of the two 3Q states is present in the corresponding region of the phase diagram.

At $U = 7$, the optimal 3×6 and 2×2 unit cell PEPS are in close competition. The 3×6 PEPS has a multi-Q order, but it is difficult to establish whether it has a 3×6 or 3×3 collinear unit cell, because the latter can only be strictly realized in the $D \rightarrow \infty$ limit in a $U(1)$ symmetric PEPS due to the LSM obstruction and hence requires a careful scaling analysis with D . With the 2×2 unit cell, we can reach bond dimension $D = 8$, and the energy is well below the exact FP energy. This sets a better lower bound for the FP region ($U = 7$) compared to HF ($U = 5.2$).

At $U = 9$, 2×8 PEPS exhibits spiral-like order with unit cells of size 1×8 . The spiral-like order is uncanted but has a modulated amplitude. This modulation is most likely a finite- D effect [70]. The variational energies are above that of the FP state, but extrapolations in D suggest that they approach the FP energy at large D [see Fig. 3(c)]. For larger U , e.g., $U = 12$, the only PEPS competing states are spirals and FP. The 2×2 (not plotted) states beyond $U = 9$ are nearly fully polarized. We conjecture that by increasing U from 7 to 12, the effective magnetic exchange interaction between electrons has changed from antiferromagnetic to ferromagnetic coupling. The long-wavelength spiral has nearly aligned spins for neighbors and is a compromise between ferromagnetic tendency and band dispersive energies.

1/2 hole doping. The noninteracting band structure at 1/2 hole doping has an almost perfectly circular Fermi surface. Thus, weak-coupling SDWs seem unlikely and are absent in restricted HF [40,42] and our unrestricted HF. Our PEPS

results also indicate the absence of magnetic orders at least for $U \leq 16$ [70].

Discussion. We now connect our results to the effective triangular-lattice systems of bilayer TMDs. Interaction strength in units of the bandwidth can be adjusted by changing the twist angle or choosing different untwisted heterobilayers. The interaction is also long ranged. Restricted HF results [30] indicate that with sufficiently strong long-range Coulomb interaction, insulating stripe phases can emerge for both 1/2 electron and hole doping, with ferromagnetic or antiferromagnetic order. We have performed HF including long-range interaction [$V \neq 0$ in Eq. (1)] and find that for the SDW region, a reasonable strength of V and screening length is not sufficient to stabilize stripes or fluxed phases [81], i.e., the existence of large-scale SDW remains unaffected by the long-range interactions. In the experiments of Refs. [12,17], no insulating phases are observed near 1/2 electron and hole doping, indicating that neither the 3Q-I spin-tetrahedral state nor stripe states are realized. The tunable metal-insulator transition at neutrality indicates that the interaction strength can be estimated as $U \sim 6 - 9$ (which roughly corresponds to $U \sim 2.5 - 3.5$ in HF), which is within the parameter regime that we have considered in this work. For experimental investigations of SDWs, the spin-valley locking in TMDs has two important consequences: in-plane spin order will result

in atomic-scale order (due to the associated intervalley coherence), and perturbations to the TLHM break the $SU(2)$ spin rotation symmetry down to $\mathbb{Z}_2 \times U(1)$, allowing for finite-temperature long-range order.

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