Spin-valley antiferromagnetism and topological superconductivity in the trilayer graphene Moire super-lattice

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A recent experiment has shown that exotic correlated insulating phases emerge in the ABC-stacked trilayer graphene-boron nitride Moire super-lattice at both quarter and half-filling. A single-band minimal model with valley contrasting staggered-flux is proposed to capture the relevant band structure of this system, where the conspiracy of perfect Fermi-surface nesting and van Hove singularity strongly enhance the valley fluctuation, leading to inter-valley spiral (IVS) order at half filling. Nevertheless, the weak coupling theory is insufficient to account for the correlated insulating state near quarter filling. In this paper, we consider a strong coupling U(1), × SU(2), symmetric spin-valley model to obtain the correlated insulating state and the pairing instability near quarter filling. A significant ingredient in the strong coupling model is the Dzyaloshinsky-Moriya like interaction inherited from the flux, which breaks not only the valley SU(2), symmetry but also the sub-valley spatial reflection symmetry. We discuss all the possible long-range orders stabilized by the effective spin-valley-exchange interactions, and it turns out that the flux remarkably enhance the ferro-spin inter-valley 120° order, which shares the same valley feature as the IVS order. Upon doping, the leading pairing instability lies in the inter-valley channel with a trigonally warped p ± ip-wave form factor in the presence of the sub-valley reflection symmetry breaking. Depending on the sign of Hund’s coupling, the total pairing state could be either spin singlet or triplet. While the spin singlet chiral topological pairing state (p ± ip)↑↓ − (p ± ip)↓↑ is necessarily chiral, the spin triplet topological pairing state could be chiral (p ± ip)↑↑ + (p ± ip)↓↓, or helical (p ± ip)↑↑ + (p ± ip)↓↓.

I. INTRODUCTION

Valley is a novel low energy degree of freedom commonly studied in graphene-based systems, which may be viewed as an isospin. It carries rather rich topological consequences and been actively explored. However, the valley degree of freedom rarely sets foot in the Mott physics and high Tc superconductivity in the past. The Moire heterostructure gives a new opportunity. In these heterostructures, the original lattice periodicity is broken, and a Moire super-lattice emerges on a larger scale, which efficiently suppresses the kinetic energy scale by folding the bands. As a result, the valley degree of freedom that is highly nonlocal in original lattice is now tamed as a local orbital in the Moire superlattice. Meanwhile, the local interactions that were otherwise weak could possibly come to dominate the kinetics and lead to correlated physics.

Very recently, the experiment of magic-angle twisted bilayer graphene (TBG) successfully demonstrate this scenario. Twisting the graphene bilayer is one efficient way to produce triangular Moire superlattice. Under the tiny magic angle like ∼ 1.08°, the Moire wave-length is about 15 nm, and the low energy bandwidth is most significantly suppressed down to about 10 meV. Meanwhile the local Coulomb interaction was estimated to be of order 10 meV. The experimentalists showed correlated insulating phases with ∼ 0.31 meV gap at half-filling of the valence and conductance bands. Besides, they even observed highly unconventional superconductivity down to ∼ 1.7 K near the half-filling.

Nevertheless, twisting is not the only way to Moire superlattice. The hexagonal boron nitride (hBN) that is commonly used as the substrate for graphene shares almost the same honeycomb lattice with graphene, but with 1.8% larger lattice constant. When the hBN is carefully aligned with a generic multi-layer graphene, this tiny mismatch of the lattice could induce a Moire superlattice with Moire wavelength up to ∼ 15 nm. The band folding and greatly suppression of kinetic energy would equally give the possibility to correlated physics. Indeed, not long after the publication by Pablo’s group, the group led by Feng Wang also reports their discovery of correlated insulating states in the heterostructure of ABC-stacked trilayer graphene (TLG) over hBN. The choice of multi-layer graphene, despite its difficulty in fabrication, is on the purpose of suppressing kinetic energy to the best, as multi-layer results in higher order energy-momentum dispersion near charge neutral point. It turns out that the low energy valence bandwidth is also about 10 meV, and the local Coulomb interaction is also estimated to be of order magnitude ∼ 15 meV. In their experiment, correlated insulating state is observed not only at half-filling but also quarter-filling, with ∼ 2.2 meV gap at half filling and ∼ 0.5 meV gap at quarter filling. And the insulating signal is prominent in valence band but not conductance band.

Now we have two graphene-based Moire superlattice systems in experiments, both of which have similar Moire wave-length, low energy bandwidth, and estimated local Coulomb interaction. However, besides the insulating
gap value, the two systems are qualitatively distinct in the relevant band structure. In TBG, the $C_2 T$ protected Dirac cones glue the valence band and conductance band, and partially filling either bands shows insulating behavior. In contrast, in TLG/hBN, the Dirac cone is no longer protected due to the breaking of $C_2 T$, therefore separating the valence band with the conductance band. Moreover, the experiment shows prominent insulating behavior in partial filling valence band instead of conductance band, suggesting highly particle-hole asymmetry with respect to the charge neutral point. While many people argue for a minimal model capturing the Dirac cones to describe the TBG, it seems that a simpler single band model for the valence band is sufficient to describe the correlated physics in TLG-hBN. Simpler band structure sheds light on the essential correlated physics. Hence we mainly focus on the TLG/hBN system despite the flooding interests in TBG.

Although the Moire bandstructure of generically twisted multi-layer graphene over hBN has already been intensively studied in the past ten years, the newly emerged correlated physics such as Mott phases and unconventional superconductivity is far beyond the description of noninteracting band theories. To reveal the strongly correlated nature, a simplest minimal model capturing the only relevant Moire band and the strong interactions is urgently needed. Indeed, a series of subsequent works concerning this issue have already been proposed soon after the TBG experiments. In a paper by the present authors, the effective bandstructure in TLG/hBN was calculated by the continuum Dirac model, based on which a weak coupling minimal model was proposed to describe the low energy valence band. Among the variety of recent theoretical works, we were the first to demonstrate that the half-filled Fermi-surface (FS) of the two valleys in TLG are exposed to a strong nesting effect and van Hove singularizes, leading to the IVS order. We also pointed out that a similar mechanism is likely to happen in TBG system, except that there would be a triple-Q nesting between valleys instead of single Q nesting. The triple-Q nesting scenario has been firmly demonstrated by a series of subsequent works. From this weak coupling picture, the Mott features in both TBG and TLG/hBN systems near half-filling are mainly attributed to the nesting valley FS. However, the weak coupling nesting scenario is hard to account for both the Mott insulating states at half-filling and quarter-filling. The fact that a gap opens whenever one additional electron is added per site points towards a strong coupling tendency.

In this paper we come to address the issue of quarter-filling in TLG/hBN from the strong coupling aspects, where the spin-spin repulsion, the valley-valley repulsion and the Hund’s coupling are considered. From the strong coupling limit we derive the effective spin-valley-exchange interactions. The interactions contain the most generic terms allowed by $U(1)_c \times SU(2)_s$ symmetry, including the anti-symmetric Dzyaloshinsky-Moriya (DM) like interaction. While we derive the effective model from the TLG-hBN system, this spin-valley model with generic flux could apply to other graphene-based Moire superlattice. This spin-valley model is qualitatively distinct from the conventionally studied spin-orbital model, due to the exceptional appearance of valley-contrasting flux. A very important consequence is the breaking of sub-valley reflection symmetry that has severe impact on the Mott state and pairing symmetry. We sketch the classical phase diagram for varying parameters by minimizing the energy. The valley-contrasting flux remarkably enlarges the phase space of the ferro-spin inter-valley $120^\circ$ order, which shares the same valley feature as the IVS order from weak coupling theory. When the filling deviates from exact quarter-filling, the leading pairing instability is found to be the inter-valley pairing, whose form factor is a trigonally-warped ($p \pm ip$)-wave. Depending on the sign of the Hund’s coupling, the pairing could favour spin triplet or singlet. The spin singlet pairing is necessarily chiral and breaks $T$. Within the spin triplet channel, the total pairing state could be either chiral superconductor or helical superconductor protected by time reversal symmetry.

This paper is organized as follows. After brief reviewing the Moire band structure and minimal model of the TLG/hBN system, we propose the spin-valley extended Hubbard model in section II. In section III, we go to the strong coupling limit and derive the effective spin-valley-exchange interactions and discuss its ground state order. In section IV, we investigate the leading pairing instability when the Mott insulator is lightly doped. After a brief summary in section V, we’ll compare our theories with others and discuss the experimental signals and some further generalization of our model in the final section VI.

II. MOIRE BAND STRUCTURE AND SPINFUL MINIMAL MODEL

In this section we briefly review the effective bandstructure and minimal model before introducing the complete interaction terms. The ABC-stacked TLG has the same Bravais lattice as in the monolayer graphene. But the electron and hole touching at zero energy support chiral quasiparticles with $3\pi$ Berry phase, generalizing the low-energy band structure of the monolayer and bilayer graphene and characterizing a trigonal warped triple Dirac dispersion in each of the two valleys. The hBN also forms a honeycomb lattice but has a lattice constant about 1.8% larger than that of the graphene. Thus the heterostructure of TLG and hBN breaks the original lattice periodicity and there emerges a large scale triangular Moire super-lattice as shown in Fig.1, which contains three interlaced regions in each Moire unit cell. The TLG/hBN heterostructure possesses the three-fold rotational symmetry along the z-axis $C_3$, the mirror reflection symmetry with respect to the y-z plane $M_x$ and the time reversal symmetry $T$. 
whose Dirac points are close to $K$. Low-energy Moire band structure for the valley into many mini-Brillouin zone by the Moire periodic potential. The original lattice (marked by the purple hexagon) is folded to $C_h$BN, while yellow and green have maximal misalignment and blue one shows the maximal alignment between the TLG and interlaced regions shaded by blue, yellow, and green. The mismatch to 33%. The Moire pattern is composed of three black honeycomb lattice) and hBN (red honeycomb lattice).

FIG. 1: (a) Super-lattice formed by TLG (abstracted by a black honeycomb lattice) and hBN (red honeycomb lattice). For the sake of clearness, we exaggerate the lattice constant mismatch to 33%. The Moire pattern is composed of three interlaced regions shaded by blue, yellow, and green. The blue one shows the maximal alignment between the TLG and hBN, while yellow and green have maximal misalignment and are related by $C_h$. (b) The Brillouin zone of the TLG on the original lattice (marked by the purple hexagon) is folded into many mini-Brillouin zone by the Moire periodic potential. (c) Low-energy Moire band structure for the valley $K$ whose Dirac points are close to $K_s$ in the mini-BZ. The Dirac points near $K_s$ are gapped out by the Moire potential, which is approximated by $V_M \approx 80\, \text{meV}$. (d) Contour plot of the corresponding valence band near the CNP in the mBZ (black honeycomb). The vicinity of $K'_s$ hosts three saddle points where the density of states diverges for this valley band. Color represents energy in unit of meV. (e) The minimal model features a valley-contrasting staggered flux. (f) The Fermi surfaces of two valleys (the red and blue triangles) at half-filling are nested by $Q = (4\pi/3, 0)$ and its equivalents.

The hBN has a large charge gap $\sim 4.6\, \text{eV}$ and therefore contributes only a Moire periodic potential to the low energy dynamics in TLG. Stemming from the difference between boron and nitride atoms, the Moire potential breaks the protecting symmetry of the Dirac points i.e. sub-lattice symmetry $C_2 \mathcal{J}$ (Ref.\textsuperscript{10,25}). Viewed in momentum space, the Moire periodic potential scatters the low energy valleys repeatedly to form a Moire reciprocal lattice. Within the Moire mini-Brillouin zone (mBZ), the dispersion is folded and split into many Moire minibands, whose energy scale decreases from the original bandwidth by orders of magnitude\textsuperscript{20,24}. Notice that as the distance between the two valleys are about 62.5 times longer than the Moire wave-vector, the Moire coupling between the two valleys are exponentially small and can be neglected. As a result, the valleys that were connected within one band are now well separated and compose two degenerate Moire mini-bands. In real space, that means the valley degree of freedom becomes a local orbital in Moire super-lattice, analogous to the physical spin.

Using the effective two-component Hamiltonian for the TLG\textsuperscript{19} we have calculated the band structures with the first harmonic component of Moire scattering potential $V_M$ assumed to act only on the bottom graphene layer\textsuperscript{19,25}. The low energy band dispersion is shown in Fig. 1c and 1d. Indeed, the kinetic energy scale is quenched from about 1 eV to around 20 meV. As the Dirac points are further gapped out by the Moire potential, the valence band is well isolated. Besides the flatness, there are two most significant features in this bandstructure. First, the half-filled Fermi-surface (FS) is subjected to remarkably good nesting instability between the two valleys. Second, the splitting of Dirac cones leaves triple van Hove points along zone boundary in the vicinity of $K'_s$. The stronger Moire potential, the closer the van Hove points merge towards $K'_s$.

Upon Fourier transformation, we can parametrize the isolated valence band by a tight binding model in real space triangular Moire super-lattice. Note that due to the singleness of the band, we directly Fourier transform the band dispersion and the hopping integrals obtained in this way is unique, regardless of the gauge of the Bloch wave-function and the choice of Wannier orbitals.

| $V_M$ | 80   | 100  | 200  |
|-------|------|------|------|
| $t_1$ | $2.1266\, e^{0.1128\pi}$ | $1.7702\, e^{0.1209\pi}$ | $0.9013\, e^{0.1419\pi}$ |
| $t_2$ | $0.1344\, e^{i0\pi}$ | $0.1160\, e^{i0\pi}$ | $0.0620\, e^{i0\pi}$ |
| $t_3$ | $0.0411\, e^{-i0.4560\pi}$ | $0.0129\, e^{-i0.5639\pi}$ | $0.0099\, e^{i0.2288\pi}$ |

As is shown in the Table,\textsuperscript{22} it turns out that the hopping integral on the triangular lattice decays very fast: the next-nearest neighbor hopping is smaller than the nearest neighbor hopping by one order of magnitude, and the third nearest neighbor hopping is further smaller by two order of magnitude. Therefore, as a simplest minimal model, we take only the nearest neighbor hopping, which is estimated to be about 1 meV in order to produce a bandwidth of about 10 meV. The most important thing is the presence of the complex phase of the nearest neighbor hopping, which breaks the inversion symmetry.
inside the valley. And since the two valleys are related by $M_z$, the valley rotation symmetry is broken down to the valley conservation symmetry i.e. $\text{SU}(2)_{v} \to \text{U}(1)_v$. The complex phase shapes the band structure dramatically and ranges slightly different depending on the different Moire potential $V_{M}$. However, as the Moire potential $V_{M}$ grows stronger, the band dispersion acquires an asymptotic particle-hole symmetry. In fact, when the phase factor becomes $\pi/6$, there emerges a particle hole symmetry and the Fermi-surface at half-filling is exposed to perfect nesting instability.

For simplicity, we'll just consider the ideal phase $\pi/6$ in the following discussion and argue that it explains the essential physics in the real materials in the absence of perpendicular electric field. Taking into account the symmetries $C_3$, $M_x$, and $T$, the hopping integral distribution in the triangular super-lattice is shown in Fig. 1. We can see that there exists a valley-contrasting staggered flux in every elementary triangles, and the flux is $\pm 3\phi = \pm \pi/2$ depending on the valley and the triangles. Thus the minimal tight-binding model for the valence band of the TLG-hBN heterostructure is given by the Hamiltonian

$$H_t = \sum_{r,\nu,\sigma} \sum_{\delta} \left( -\tau \epsilon \phi \sum_{\nu} \delta_{r+\delta,\nu,\sigma} c_{r,\nu,\sigma} c_{r,\nu,\sigma} + h.c. \right) - \mu \sum_{r,\nu,\sigma},$$

where $\delta = (1, 0)$ and $(-1/2, \pm \sqrt{3}/2)$ are the nearest neighboring vectors of the primitive unit cell. The band dispersion of valley $\pm$ is given by $\epsilon_{\pm k} = -2t \sum_{k} \cos(k \cdot \delta) + \phi$, which varies with the phase $\phi$. In general, the valley band dispersion is trigonally warped by the nonzero flux that breaks the sub-valley reflection symmetry $M_x \tau_z$ or the sub-valley six-fold rotation $C_6 \tau_z$, along with which shift the van Hove singularity points.

In the previous paper[1], we mainly discuss the instability of FS from the weak coupling scenario, where the perfect nesting condition leads to a logarithmic divergence of inter-valley susceptibility and strongly enhance the valley interaction. This justifies why we neglect the spin interaction but focus on the valley interaction. Now in this paper, we take a different starting point, namely, the strong coupling limit and treat the valley and spin interaction on equal footing. This is motivated by the experimental signature that even the 1/4 filling exhibits insulting behavior. Given the large Moire unit cell, we assume that only on-site interactions dominate and neglect the long range interactions. Restricted by the symmetries, the on-site interactions contain the spin-spin Hubbard repulsion and valley-valley Hubbard repulsion as well as the Hund’s coupling:

$$H_{int} = V \sum_{r,\sigma,\sigma'} n_{r,+,\sigma} n_{r,-,\sigma'} + U \sum_{r,\nu} n_{r,\nu,t} n_{r,\nu,t} - 2 J_H \sum_{r} \left( S_{r,+} \cdot S_{r,-} + \frac{n_{r,+} + n_{r,-}}{4} \right),$$

where we define the spin operator on each valley as $S_{r,\nu}$ and correspondingly we can define the valley-isospin operator on each site in terms of Pauli matrix as $T_{r,\sigma}$:

$$S_{r,\nu} = \frac{1}{2} \sum_{a,\beta} c_{r,\nu,a}^{\dagger} \sigma_{a\beta} c_{r,\nu,b} (a = x, y, z),$$

$$T_{r,\sigma} = \frac{1}{2} \sum_{a,\beta} c_{r,\sigma,a}^{\dagger} \tau_{a\beta} c_{r,\sigma,b} (a = x, y, z).$$

So the total spin and valley operator on each site is $S_{r} = \sum_{\nu} S_{r,\nu}$ and $T_{r} = \sum_{\sigma} T_{r,\sigma} (a = x, y, z)$. Note also that the pair hopping term from one valley to another is neglected. For the usual Hund’s coupling, $J_H > 0$, which guarantees that the on-site orbital singlet (spin triplet) gains energy from Coulomb repulsion. However, in general there is no forbidding anti-Hund’s coupling $J_H < 0$ that favours the orbital triplet instead.

Let’s take a check on the symmetries of this Hamiltonian. The presence of valley-contrasting flux breaks valley SU(2)$_v$ down to U(1)$_v$ and breaks the sub-valley reflection symmetries. The Hund’s coupling breaks the independent spin rotation on each valley leaving a total spin rotation symmetry SU(2)$_+$ x SU(2)$_-$ → SU(2)$_v$. As a result, the strong coupling Hamiltonian should be generically invariant under U(1)$_v$ x U(1)$_v$ x SU(2)$_v$. Moreover, when it comes to the spatial symmetries, the sub-valley reflection symmetry $M_x \tau_z$ is broken, as well as the sub-valley six-fold rotation $C_6 \tau_z$. As we’ll show later, the breaking of this sub-valley reflection symmetry changes the Mott state and pairing symmetry dramatically.

### III. STRONG COUPLING EFFECTIVE THEORY AT QUARTER FILLING

At quarter filling, there is one electron on each site on average. The limit $U, V \gg t$ would expel any double occupied states and freeze the charge, giving rise to a Mott insulator as observed by the experiment. Nevertheless, the spin and valley degree of freedoms are mobile and can win the energy from the virtual hopping process. To obtain the leading effective interaction, let’s first consider two neighboring sites, $r$ and $r + \delta$. The phase factor of the hopping can then be equivalently treated as gauging the phase of electron on $r$: $c_{r,\nu} \to e^{i \phi} c_{r+\delta,\nu}$ and $c_{r+\delta,\nu} \to e^{-i \phi} c_{r,\nu}$. The 16 possible states of the two electrons can be labeled by their total spin and valley quantum number $|S, S_{z}; T, T_{z}|$. However, the anti-symmetric states have the privilege of virtual hopping and lowering the energy. The lowered energy can be simply calculated by treating the hopping terms as perturbation and performing second order perturbation. The anti-symmetric condition locks the total spin and valley quantum number for the states with low lying energy. The result is shown in Table 1.

By means of projector onto each level labeled by good quantum number we could immediately write down the
TABLE II: Low lying states of the two sites labeled by the spin and valley quantum number.

| Energy levels | channels | deg |
|---------------|----------|-----|
| 0             | $|S = 0; T = 0\rangle \& |S = 1; T = 1\rangle$ | 10  |
| $\frac{4t^2}{V-J_{\perp}}$ | $|S = 0; T = 1, T_z = \pm 1\rangle$ | 2   |
| $\frac{4t^2}{V-J_{\parallel}}$ | $|S = 0; T = 1, T_z = 0\rangle$ | 1   |
| $-\frac{4t^2}{V-J_{\parallel}}$ | $|S = 1; T = 0\rangle$ | 3   |

The normal valley-exchange interaction is modified by the local spin-valley-exchange interactions dressed by flux is given be

$$h_{i,j} = \frac{1}{4} (J_1 + 2J_2 - J_0) S_i \cdot S_j + \left[ (J_0 + J_1) S_i \cdot S_j + \frac{3J_0 - J_1}{4} \right] T_i \cdot \left( e^{i T^x z \phi} T e^{-i T^x z \phi} \right)_j - 2 (J_1 - J_2) \left( S_i \cdot S_j - \frac{1}{4} \right) T_i^z T_j^z,$$

where we have denoted the energy gain from virtual hopping of intra-valley spin singlet channel, the inter-valley spin singlet channel and the inter-valley spin triplet channel respectively as $J_2 \equiv \frac{4t^2}{V-J_{\perp}}, J_1 \equiv \frac{4t^2}{V+J_{\parallel}}$ and $J_0 \equiv \frac{4t^2}{V-J_{\parallel}}$.

The normal valley-exchange interaction is modified by the flux as

$$T_r \cdot \left( e^{i T^x z \phi} T e^{-i T^x z \phi} \right)_r = \cos(\phi) T_r \cdot T_{r+\delta} + (1 - \cos(\phi)) T^z_r T^z_{r+\delta} - (\sin(\phi)) (T_r \times T_{r+\delta}) \cdot \hat{z},$$

which introduces additional easy-plane interaction as well as the anti-symmetric DM interaction. Therefore, the valley SU(2)$_v$ has been broken, and the DM interaction further breaks the sub-valley reflection symmetry $M_x \tau_x$.

Since the exchange interactions are overall repulsive, the system is expected to exhibit overall antiferromagnetism, which can be contributed by either the spin or the valley. Without the knowledge of the accurate parameters, we’ll try to understand this spin-valley model in a broad range of parameters. While it is hard to accurately solve the quantum model exactly, as a first step we treat the model in the classical limit, where the spin and valley-isospin are treated as vectors and the ground state energy is minimized by minimizing the energy on each bond.

Recall that on a triangular lattice with antiferromagnetic Heisenberg interaction, the Neel order along $S^z$ direction is frustrated by the lattice, and the dipole spins are compromised to form a classical coplanar 120\degree order[14][15]. The spin 120\degree order exhibits alternating spin chirality around the elementary triangle plaquettes. Therefore this order has two degenerate configurations, that differ by the spin chirality on a given plaquette, as shown in Fig. 2a. For both classical configurations at zero temperature, the expectation value of exchange term of each bond saturates to the same value: $\langle S_r \cdot S_{r+\delta} \rangle = \frac{1}{2}$. This is due to the reflection symmetry $M_x$ that relates the two configurations. In contrast, when it comes to the valley-isospin, the two valley-isospin 120\degree configurations are related by the sub-valley reflection symmetry $M_x \tau_x$ instead. Therefore, the breaking of $M_x \tau_x$ lifts the degeneracy between the two valley-isospin 120\degree configurations:

$$\langle T_r \cdot \left( e^{i T^x z \phi} T e^{-i T^x z \phi} \right)_r \rangle = \frac{1}{4} \cos(\frac{2}{3} \pi \pm 2 \phi),$$

in which the ± sign depends on the relative sign between the isospin chirality and flux around a given plaquette, as shown in Fig. 2b. In our situation, $\phi = \pi/6$. Hence for the configuration where positive sign of isospin chirality matches positive sign of flux on a plaquette, $\langle T_r \cdot \left( e^{i T^x z \phi} T e^{-i T^x z \phi} \right)_r \rangle = -1/4$; for the configuration where the sign of the isospin chirality and flux does not match, $\langle T_r \cdot \left( e^{i T^x z \phi} T e^{-i T^x z \phi} \right)_r \rangle = 1/8$. In this sense, the flux stabilizes one of the valley 120\degree configurations but repels the other one.

Meanwhile, the flux also lifts the degeneracy between the planar and Ising ferromagnetic states of valley-isospin. For convenience, we denote in the following the energetically favorable valley-isospin 120\degree order as T-AF, and the spin 120\degree order as S-AF. The Ising ferromagnetism of valley is abbreviated as T-Fz, which shows expectation

$$\langle T_r \cdot \left( e^{i T^x z \phi} T e^{-i T^x z \phi} \right)_r \rangle = \langle T^z_r T^z_{r+\delta} \rangle = 1/4.$$

On the other hand, the planar ferromagnetic state of valley has

$$\langle T_r \cdot \left( e^{i T^x z \phi} T e^{-i T^x z \phi} \right)_r \rangle = (T^z_r \cdot T^z_{r+\delta}) = 1/8, (T^z_r \cdot T^z_{r+\delta}) = 0$$

and this ordered state is abbreviated as T-Fxy. Due to the SU(2)$_x$ spin rotation symmetry, the ferromagnetic
state of spin does not discriminate the Ising and planar ferromagnetism, and can be simply denoted as S-F, which saturates the expectation value \( \langle S_\uparrow \cdot S_{\uparrow+} \rangle = 1/4 \). The energy of the several classical order candidates is shown in Table III.

TABLE III: The energy of the classical orders as combination of the spin and valley (anti-)ferromagnetism. We draw a comparison between the model with zero flux and with nonzero flux.

| classical orders | energy \((\phi = 0)\) | energy \((\phi = \pi/6)\) |
|------------------|----------------|---------------------|
| I(S-F, T-AF)     | \(-\frac{1}{2}J_0\) | \(-\frac{1}{2}J_0\) |
| II(S-AF, T-AF)   | \(-\frac{3}{8}(\frac{J_1}{2} + \frac{J_2}{2} + \frac{5J_0}{8})\) | \(-\frac{3}{8}(\frac{J_1}{2} + \frac{5J_0}{48})\) |
| III(S-AF, T-Fz)  | \(-\frac{3}{8}J_2\) | \(-\frac{3}{8}J_2\) |
| IV(S-AF, T-Fxy)  | \(-\frac{3}{8}(\frac{J_1}{2} + \frac{J_1}{2})\) | \(-\frac{3}{8}(\frac{J_1}{2} + \frac{3J_1}{8} + \frac{5J_0}{24})\) |

By minimizing the average bond energy, we sketch the phase diagram shown in Fig. 3. Before discussing the case with flux, let’s first shut down the flux and look at the more conventional classical phase diagram in Fig. 3. The diagonal line \((J_1 = J_2)\) in this phase diagram corresponds to the SU(2)\(_x\)×SU(2)\(_z\) symmetric spin-orbital model. According to the previous results, the SU(2)\(_x\)×SU(2)\(_z\) symmetric model under \(J_0 = J_1\) exhibits gapless ordered states robust against quantum fluctuation, partially justifying our assumption of ordering in our two-dimensional model. The limit of \(J_1 = J_2 = 0\) leaves only \(J_0\) term that stabilizes the S-F and T-AF order, and the other limit with \(J_1 = J_2 \gg J_0\) stabilizes the S-AF and T-F instead. In between the more nontrivial gapless phase exists to display the SU(4) symmetry point \(J_0 = J_1 = J_2\). The Schwinger boson mean-field approach shows S-AF and T-AF long range order for this high symmetry model. When \(J_1\) deviates from \(J_2\), the anisotropy of the orbital space occurs and leads to different ferromagnetic state of valley-isospin.

Now let’s turn on the flux and see what’s happening. As shown in Fig. 3, the phase space of the S-F and T-AF is remarkably enlarged. Within a considerable phase space \(J_1 < 5J_0/3, J_2 < 5J_0/3\), the valley-isospin shows coplanar 120° order. This is no wonder when being reminded that the flux lowers the energy and stabilize the valley antiferromagnetic state. Near the phase boundaries where more than one classical orders are degenerate and competing, the quantum fluctuation would play an important role and possibly lead to nontrivial physics such as spin liquid. But far away from the phase boundaries, the quantum fluctuation is supposed to render only tiny correction to the energy but not change the nature of the ordered state.

Finally, we give a bit comment on the finite temperature behavior. As the system is a clean 2-dimensional lattice, the Mermin-Wagner theorem defies continuous symmetry breaking under any finite temperature. Therefore, the thermal fluctuation would resist the spontaneous breaking of spin SU(2)\(_x\) symmetry and the formation of spin long range order. Instead, the spin order parameter fluctuates in real space and can be stabilized by small magnetic field that explicitly breaks the symmetry. Nevertheless, the breaking of valley U(1)\(_y\) symmetry can occur through the Kosterlitz-Thouless transition under finite temperature, establishing the quasi-long range valley order.

### IV. INTER-VALLEY PAIRING INSTABILITY

When the filling deviates slightly from quarter filling i.e. doping the Mott insulator, the effective Hamiltonian would additionally involves a hopping term of the charge carriers under occupancy constraint:

\[
H_{\text{eff}} = \mathcal{P} (H_t + H_J) \mathcal{P}
\]

where \(\mathcal{P} = \prod_r \frac{1}{2} n_r (2 - n_r) (3 - n_r) (4 - n_r)\) is the projector that projects onto the single or null occupancy on each site. The spin-valley-exchange interaction is under the projection of single occupation.

To reduce redundancy we can decompose the fermion degree of freedom into the holon and spin-valleyon \(c^\dagger_{r\nu\sigma} = h^\dagger_{r\nu} f^\dagger_{r\nu\sigma}, \) under the constraint \(\sum_{\nu, \sigma} f^\dagger_{r\nu\sigma} f_{r\nu\sigma} = 1\). In this way, after some simple derivation the spin-valley-exchange interaction under constraint \(\mathcal{P} H_J \mathcal{P}\) can be exactly expressed in terms of the spin-valleyon without redundancy. We can decompose the exchange interaction into 6 anti-symmetric pairing channels which are energetically favourable than the...
other 10 symmetric pairing channels:

\[
\mathcal{P} \mathcal{H} \mathcal{P} = -\frac{1}{4} \sum_{r,\delta} \left\{ J_2 \tilde{\Delta}_2(r, r + \delta) \cdot \tilde{\Delta}_2(r, r + \delta) \\
+ J_1 \Delta_1(r, r + \delta) \Delta_1(r, r + \delta) \\
+ J_0 \tilde{\Delta}_0(r, r + \delta) \cdot \tilde{\Delta}_0(r, r + \delta) \right\},
\]

(10)

where the spin singlet intra-valley channel, spin singlet inter-valley channel and spin triplet inter-valley channel are denoted respectively as:

\[
\tilde{\Delta}_2(i, j) \equiv \psi_j e^{-ir_i \phi} \sigma_y \gamma_j (\tau_x, \tau_y) \psi_i,
\]

\[
\Delta_1(i, j) \equiv \psi_j e^{-ir_i \phi} \sigma_y \gamma_x \psi_i,
\]

\[
\tilde{\Delta}_0(i, j) \equiv \psi_j e^{-ir_i \phi} \gamma_y \psi_i.
\]

The spin-valleyon basis is compactly expressed as \( \psi_r \equiv (f_{r,+}, f_{r,-}, f_{r,\uparrow}, f_{r,\downarrow}) \). The above expression is exact without approximation, as long as the particle constraint is rigorously kept.

For further discussion we’re going to do the approximation and treat the holon in mean-field level \( \langle \hat{h} \rangle = \langle \hat{h}^\dagger \rangle = \sqrt{\tau} \), with \( x \) being the charge carrier density away from one quarter. Thereby we obtain a renormalized kinetic hopping term for the spin-valleyon:

\[
H_t \rightarrow \int \sum_{k, \nu, \sigma} (\tilde{\epsilon}_{\nu k} - \mu) f_{k, \nu, \sigma}^\dagger f_{k, \nu, \sigma},
\]

(12)

in which the hopping amplitude is renormalized by the charge carrier density: \( t \rightarrow \tilde{t} = tx \). The kinetic term determines the spin-valleyon FSs, which have spin degeneracy but differ by the valleys. The FS of two valleys are inversion-related triangular warped pocket filling about 1/4 of the BZ (Fig. 4). Due to the sub-valley reflection symmetry breaking, the single valley FS lacks inversion symmetry and therefore it is frustrating for two spin-valleyons on the same valley FS to form a Cooper pair with constant center-of-mass momentum. Therefore we argue that it is difficult for the intra-valley pair to condense. In the following we mainly compare the spin singlet and spin triplet inter-valley pairing channels.

Before introducing the pairing order parameter, we can rewrite the interaction in the pairing hopping form in momentum space:

\[
\mathcal{P} \mathcal{H} \mathcal{P} \rightarrow \sum_{k, k'} \eta_{k, k'} \left( \psi_{k, \downarrow}^\dagger \gamma_y \psi_{k', \downarrow}^\dagger \cdot \psi_{k, \downarrow} \gamma_y \psi_{k', \downarrow} \right) \cdot \left( \psi_{k, \uparrow}^\dagger \gamma_y \psi_{k', \uparrow}^\dagger \cdot \psi_{k, \uparrow} \gamma_y \psi_{k', \uparrow} \right) + \int d \phi J_0 \eta_{k, k'} \left( \psi_{k, \downarrow}^\dagger \gamma_y \psi_{k', \downarrow}^\dagger \cdot \psi_{k, \downarrow} \gamma_y \psi_{k', \downarrow} \right)
\]

(13)

where the hopping form factor \( \eta_{k, k'} \equiv -\sum_\delta \cos(k \cdot \delta + \phi) \cos(k' \cdot \delta + \phi) \). From this we can see that the interactions fall in two competing pairing instability, the spin singlet pairing driven by \( J_1 \) and the spin triplet channel driven by \( J_0 \) respectively.

\[
\eta_{k, k'} = -\frac{1}{3} \left( D_k D_{k'} + D_k^* D_{k'} + S_k S_{k'} \right),
\]

(14)

\[
S_k \equiv \sum_\delta \cos(k \cdot \delta + \phi),
\]

\[
D_k \equiv \cos(k_x + \phi) + e^{i2\pi/3} \cos(k_x/2 - \sqrt{3}k_y/2 - \phi) + e^{-i2\pi/3} \cos(k_x/2 + \sqrt{3}k_y/2 - \phi).
\]

(15)

The first channel is s-wave that can be excluded, and the latter two are T related chiral representations. The chiral representation indeed satisfies the sign changing condition across the FS and are energetically favorable. Under \( T_c \), the Cooper pair would spontaneously pick up one of the chiral form factor.

We can examine the pairing symmetry associated with \( D_k \) more carefully. Near the mBZ center \( \Gamma_s \), the pairing form factor can be expanded as \( D_k = -\frac{1}{3} (k_x + i k_y) + \ldots \).
E/t
-1
-2
0
1
2

FIG. 5: (a) and (b) show the distribution of $\vec{d}_k$ vector defined by form factor $D_k$ in mBZ, for valley $+$ (left figure) and valley $-$ (right figure) respectively. The trigonally-warped FS is highlighted by blue/red lines for valley $\pm$ respectively. The black hexagon marks the first mBZ. The small black circles inside the figure denote vortex cores while the black crosses denote anti-vortex cores, both of which are zeros of the pairing condensate. The vortex cores are located on $\Gamma$ and its $C_3$ counterparts. The FSs are fully gapped and the pairing condensate winds $2\pi$ around them. (c) and (d) show the Bogoliubov spectra for one of the spin triplet channel being placed on an open cylinder with smooth edges, for valley $+$ and valley $-$ respectively. The momentum $k$ follows the edges along $x$ direction. The blue and red line denote the chiral edge modes on the two edges respectively, indicating a chiral Bogoliubov edge mode. The chemical potential $\mu \approx -1.58t$ to ensure filling near 1/4 filling, and the pairing order parameter is chosen as $\Delta = 0.5 t$ for a clearer demonstration, whose value does not affect the topology.

$O (k^2)$, which contains the $p + ip$ pairing form to the leading order. Indeed, we investigate the pairing gap on FSs close to quarter filling (Fig 3), which exhibits amplitude anisotropy and winding of phase by $2\pi$. The gap maxima are located near the FS corners while the gap minima are at the middle point of each arc of the near-triangle-shaped FS. To gain a better insight into this, we explicitly map out the vector field of the complex pairing form factor $D_k$ in the mBZ: $\vec{d}_k = \Delta (\text{Re}D_k, \text{Im}D_k)$. As shown in Fig 5, in each mBZ there are three vortex cores residing on the $\Gamma_\pm = (0, 0)$, $K_\pm = (-4\pi/3, 0)$, and $K'_\pm = (4\pi/3, 0)$ respectively. Besides, three anti-vortex cores are located at $-Q/2$, i.e. $(-2\pi/3, 0)$ and its $C_3$ counterparts. These anti-vortices result in sign changing from $k$ to $-k - Q$. The hole doped valley FSs avoid all the zeros and hence are fully gapped, but each FS encloses single vortex core residing at $\Gamma_\pm$, which explains the phase winding by $2\pi$. This indicates that it is adiabatically equivalent to the chiral $p + ip$ pairing condensate, except that the form factor is trigonally warped by the flux. The mean-field superconducting Hamiltonian can be easily proved to yield a Bogoliubov de-Gennes topological Chern number for each valley:

$$C_\nu = \frac{1}{4\pi} \int_k \langle \hat{h}_k \cdot (\partial_x \hat{h}_k \times \partial_y \hat{h}_k) \rangle = 1,$$

where $\hat{h}_k$ is the unit vector of $\hat{h}_k \equiv (d^\dagger_k, d^0_k, \hat{c}_k)$, and $\hat{c}_k$ is the renormalized effective kinetic term.

To see that the topological state does support gapless edge modes, we perform exact diagonalization for the pairing state placed on a cylinder with smooth edges. As is shown in Fig. 5a and 5b, the Bogoliubov spectra of Bogoliubov quasi-particles of both valleys support chiral gapless Bogoliubov edge modes.

The above discussion is limited in a selected channel in the spin space. Now we come back to address the spin space part of the pairing symmetry. There are three possible scenarios depending on the parameters.

1. When $J_1 > J_0$, the spin singlet pairing is more energetically favourable. Since there is only one component in the spin singlet channel, it is necessarily a chiral state. We could introduce the chiral spin singlet pairing order

$$\Delta_{\text{chiral}} = -\frac{J_0}{3} \int_k \langle \psi_{s\downarrow s\downarrow} \rangle D_k^*,$$

$$H_{\text{MF}}^s = \Delta_{\text{chiral}} \int_k D_k \left( f^\dagger_{k,+} f^\dagger_{-k,-} - f^\dagger_{k,-} f^\dagger_{-k,+} \right) + h.c.$$ (18)

The edge supports a chiral spinful complex fermion mode associated with total BdG Chern number $C = 4$. The spin $SU(2)$ symmetry is respected in this case.

2. When $J_1 < J_0$, the spin triplet pairing is more energetically favourable. Different from the spin singlet case, the spin triplet pairing has multi-component and allows room for two $T$-related pairing condensates. Therefore it falls into two degenerate situations.

First, the chiral spin triplet pairing state. We could introduce the vector order parameter in the $SU(2)$ spin triplet space for the chiral spin triplet pairing:

$$\tilde{\Delta}_{\text{chiral}} = -\frac{J_1}{3} \int_k \langle \psi_{s\downarrow s\downarrow} \rangle D_k^*,$$ (19)

which corresponds to the chiral pair condensate in real space

$$\frac{J_{\text{SILO}}}{2} \langle \psi_{s\downarrow s\downarrow} e^{-i \tau_3 \phi} \sigma_y \tau_y \psi_{s\downarrow} \rangle = \tilde{\Delta}_{\text{chiral}} e^{i \frac{\pi}{2} \delta}.$$ (20)

Note that $\tilde{\Delta}_{\text{chiral}}$ is assumed to be real vector so that all directions can be related by $SU(2)_s$ rotation. In contrast, when beyond real vector, an example like $\tilde{\Delta}_{\text{chiral}} = \Delta_{\text{chiral}} (\frac{1}{2}, \frac{1}{2}, 0)$ fails to gap out the spin up FS and is
not the most energetically favourable choice. Below $T_c$ the vector order parameter spontaneously picks one direction and breaks SU(2)$_s$ → U(1)$_s$, meanwhile the chiral pair condensate breaks time reversal symmetry $T$. To illustrate the mean-field Hamiltonian after superconductivity sets in, we might as well pick up a direction like $\Delta^t_{\text{chiral}}(0, 1, 0)$ and picks up the $D_k$ without loss of generality:

$$H^t_{\text{MF}} = \Delta^t_{\text{chiral}} \int_k D_k \left( f^\dagger_{k,+,+} f^\dagger_{k,-,-} + f^\dagger_{k,+,+} f^\dagger_{k,-,-} \right) + h.c.$$  

(21)

This mean-field pairing term describes a chiral pair condensate for both spin up and spin down pairing. The edge supports a spin degenerate complex fermion edge mode, corresponding to the total BdG Chern number $C = 4$.

Second, there is the spin triplet helical pairing state. When the orbital form factors of the Cooper pairs with opposite spin are counter-chiral and related by $T$, the $T$ is preserved. Without loss of generality, we could write down the helical order parameter up to a choice of spin basis:

$$\Delta_{\text{helical}} = -\frac{2J_1}{3} \int_k \langle f_{k,-,+} f_{k,+,-} \rangle D^*_k,$$

(22)

and the interaction term is decoupled after superconductivity sets in:

$$H^t_{\text{MF}} = \Delta_{\text{helical}} \int_k D_k f^\dagger_{k,+,-} f^\dagger_{k,-,+,} + D^*_k f_{k,+,-} f^\dagger_{k,-,+,} + h.c.$$  

(23)

which is degenerate with the chiral candidate due to a fictitious symmetry that mirror reflects the momentum of only the spin down electron. In this case, the BdG Chern numbers of different spin components are opposite so that the total BdG Chern number is zero but there exists a spin BdG Chern number $C_s = 4$ in the presence of $T$. Correspondingly, the edge supports a helical complex fermion mode: the spin up component propagates counter-clockwise with the spin down component.

All in all, the relative strength between $J_0 = 4t^2/(V - J_H)$ and $J_1 = 4t^2/(V + J_H)$ depends on the sign of Hund’s coupling. To conclude, depending on the sign of the Hund’s coupling, the favourable pairing state can be spin singlet or triplet, as stabilized by $J_1$ or $J_0$. Within the spin triplet channel the pairing could fall into chiral or helical degenerate candidates, as shown in Table. [V]

| Condition | Type | Pairing Form | Symmetries |
|-----------|------|--------------|------------|
| $J_H > 0$ | chiral | $(p \pm ip)_{\uparrow\uparrow} + (p \pm ip)_{\downarrow\downarrow}$ | U(1)$_s$ × U(1)$_s$ |
|           | helical | $(p \pm ip)_{\uparrow\uparrow} + (p \mp ip)_{\downarrow\downarrow}$ | U(1)$_e$ × $T$ |
| $J_H < 0$ | chiral | $(p \pm ip)_{\downarrow\uparrow} - (p \pm ip)_{\uparrow\downarrow}$ | U(1)$_e$ × SU(2)$_s$ |

V. DISCUSSION AND SUMMARY

The perpendicular electric field is not explicitly considered in this paper by far. However, in the graphene-based multi-layer-hBN system, it is very easy and natural in experiment to tune this electric field and change the band structure completely. In fact, as are shown in Fig. 6 the valence band structure in the presence of generically nonzero perpendicular electric field essentially manifest in the moving of van Hove points, which can be captured by the flux changing as shown in Table. [V]

![FIG. 6: Valence band structure in the presence of nonzero perpendicular electric field $\Delta$ and $V_M=80$ meV. (a) $\Delta=20$ meV, (b) $\Delta=40$ meV, (c) $\Delta=60$ meV. The most important feature is the moving of the van Hove points with increasing $\Delta$.](image)

TABLE V: Parametrize the valence band of valley $+$ with tight binding model on triangular Moire superlattice. The parameters differ by different perpendicular field $\Delta$. All are in unit of meV. $V_M=80$ meV.

| $\Delta$ | 20 | 40 | 60 |
|---------|----|----|----|
| $t_1$   | 2.8648 $e^{0.3588\pi}$ | 4.9168 $e^{0.4887\pi}$ | 7.0883 $e^{0.5301\pi}$ |
| $t_2$   | 0.1385 $e^{0.0\pi}$ | -0.1053 $e^{0.0\pi}$ | -0.4166 $e^{0.0\pi}$ |
| $t_3$   | 0.1526 $e^{0.1561\pi}$ | 0.1719 $e^{0.3067\pi}$ | 0.1660 $e^{-0.9297\pi}$ |

As we’ve elaborated in the previous and this paper, the valley-contrasting flux is crucial in determining the correlated physics. Actually, the flux changing would modify the spin-valley exchange interaction and therefore could result in different spin-valley orders for the Mott insulators in a rich phase diagram. Moreover, the flux changing could also change the leading pairing symmetry dramatically. For example, when the flux is gradually turned off, the anti-vortices of the pair condensate would approach towards the $\Gamma_s$ across the FS, which drives a topological phase transition from $p \pm ip$ to $d \mp id$ pairing symmetry with $C = \mp 2$ instead. Actually, the $d \mp id$ pairing symmetry is much more commonly studied in the triangular lattice driven by repulsive interaction. [23][53] We note that the exceptional possibility of $p \pm ip$ pairing symmetry in this system is due to the nonzero valley-contrasting flux. The flux explicitly breaks the sub-valley reflection symmetry and mixes the Cooper pairs with different parity.

There are some possible experimental consequences of our Mott insulating orders and the topological superconductivity. First, the Mott states at 1/4 filling is likely
to exhibit the inter-valley 120° order. This order entails valley coherence, which may be detected by an optical experiment as the two valleys are in correspondence to the left and right circular-polarized light. Besides, concerning the orbital character of the valley, a spatial charge modulation would occur in the microscopic graphene lattice. As the two valleys in the original BZ are located on the opposite corners, the momentum interference induces $\sqrt{3}a \times \sqrt{3}a$ charge pattern in the microscopic graphene lattice. Second, since the $p+ip$ pairing superconductivity is degenerate with that of $p-ixp$, in realistic material there are likely to form domains between these two superconducting phases below the critical temperature. While the domain is supposed to show fully superconducting gap, on the domain walls there are expected to be topologically protected gapless fermion modes. These signatures are amenable to STM probe. On the other hand, our theory exhibits clear pairing gap anisotropy, which may possibly be confirmed by laser ARPES measurement with high resolution.

Compared with some recent works related with the Mott states or superconductivity of TLG/hBN Moire superlattice, our theory is different from theirs in the following aspects. While Xu and Balents study the pairing symmetry driven by SU(4)-symmetric spin-valley-exchange interaction in the presence of Hund’s coupling, our strong coupling model is shown to feature valley-contrasting flux and SU(2)$_s \times$U(1)$_v$ exchange interaction instead. The breaking of sub-valley reflection symmetry is the crucial ingredient of our theories, which leads to totally different pairing symmetries. Senthil et al. also presented a valley-contrasting flux model and discussed its strong coupling effective model, but they did not specify and discuss the flux effects. Unlike ours, their leading spin-valley exchange interaction preserves SU(4) symmetry and therefore would not yield our result. In Senthil’s later paper, he and his coworkers show that the Moire valence band could have nonzero valley Chern number. They argue SU(4) ferromagnetism and each Hubbard band is spin-polarized and valley-contrasted. However, for the spin triplet pairing state commonly studied in triangular lattice with trigonally warped $d$-wave, distinct from the $d \mp id$ pairing state commonly studied in triangular lattice with antiferromagnetic interaction. The fact that $p \pm ip$ takes the place of $d \mp id$ is due to the flux, which breaks the sub-valley reflection symmetry $M_x \tau_x$ and sub-valley sixfold rotation symmetry $C_6 \tau_x$. Therefore the Cooper pair of even and odd parity can be mixed, and the orbital angular momentum of Cooper pair is discriminative only in a modulo 3 fashion. As a result, beside $d \mp id$ pairing the $p \pm ip$ pairing state could also occur. The tringular distortion of the $p \pm ip$ pairing form factor can be viewed as the result of mixing with $d \mp id$. When the flux is artificially turned off, the pairing symmetry restores the conventional $d \mp id$. Therefore the flux is the most crucial ingredient of new physics in this system. The nonzero orbital angular momentum of the Cooper pair could in principle break $T$. However, for the spin triplet pairing case, it allows room for the formation of $T$ related Cooper pairs.

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