Inter-valley spiral order in the Mott insulating state of a heterostructure of trilayer graphene-boron nitride

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ABSTRACT
Recent experiment has shown that the ABC-stacked trilayer graphene-boron nitride Moire super-lattice at half-filling is a Mott insulator. Based on symmetry analysis and effective band structure calculation, we propose a valley-contrasting chiral tight-binding model with local Coulomb interaction to describe this Moire super-lattice system. By matching the positions of van Hove points in the low-energy effective bands, the valley-contrasting staggered flux per triangle is determined around $\pi/2$. When the valence band is half-filled, the Fermi surfaces are found to be perfectly nested between the two valleys. Such an effect can induce an inter-valley spiral order with a gap in the charge excitations, indicating that the Mott insulating behavior observed in the trilayer graphene-boron nitride Moire super-lattice results predominantly from the inter-valley scattering.

1. Introduction
The Moire super-lattice in the van der Waals heterostructure composed of multi-layer graphenes and hexagonal boron nitrides (hBN) has recently attracted great interest [1–6]. Both graphenes and hBN have hexagonal lattice structures, but the original lattice periodicity is ruined due to the mismatch between their lattice constants. Nevertheless, the periodicity can be restored on a much larger length scale, i.e., the Moire wave length ($L_M \approx 15$ nm), upon which a triangular Moire super-lattice emerges [2–4]. On the other hand, bilayer graphene with a small twisted angle can also form the Moire band structure [7–10]. In the magic-angle twisted bilayer graphene, the Moire bandwidth is reduced dramatically and the local Coulomb repulsion becomes relatively significant, leading to the observation of the Mott insulating state as well as the unconventional superconductivity around the half-filling [11,12]. Meanwhile, it has been reported that a Mott insulating state also exists in the ABC-stacked trilayer graphene-hBN heterostructure [13]. In this experiment the low energy bandwidth is about 10 meV while a Mott gap $\sim 2$ meV is observed at half filling. The comparable energy scale renders such a system in an intermediate coupling regime, therefore the role of band structure cannot be overemphasized.

In this paper, we will investigate the physical origin of the Mott insulating behavior observed in the trilayer-graphene-hBN heterostructure. Based on the symmetry analysis and effective band structure calculation, we propose a minimal tight-binding model with local Coulomb interaction. This model defined on a triangular lattice characterizes an interacting electron system in a staggered fictitious magnetic field for each of the two degenerate valley degrees of freedom. By matching the van Hove point positions of the effective low-energy bands, the staggered flux of each triangle is close to $\pi/2$. At half-filling, the two valley Fermi surfaces are found to be perfectly nested. Such an effect leads to a novel correlated insulating state with an inter-valley spiral order and a charge excitation gap, giving a natural explanation to the experimental observation.

2. Moire band structure
The ABC-stacked trilayer graphene (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 [14]. 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 can form a triangular Moire super-lattice shown in Fig. 1a, which contains three interlaced regions. The region shaded by blue circles shows the maximal alignment between the TLG and hBN, denoted as the \( \alpha \) zone; and the regions shaded by yellow or green triangles have a larger misalignment between the TLG and hBN, denoted as \( \beta \) and \( \beta' \) zone, respectively. The \( \beta \) zone differs from the \( \beta' \) zone by a sub-lattice exchange, defined by the \( C_6 \) rotation along the \( z \)-axis or the \( M_\alpha \) mirror reflection with respect to the \( x-z \) plane. Each unit cell of the Moire super-lattice includes the \( \alpha \), \( \beta \) and \( \beta' \) zone. The TLG-hBN heterostructure possesses the threefold rotational symmetry along the \( z \)-axis \( C_z \), the mirror reflection symmetry with respect to the \( y-z \) plane \( M_y \), and the time reversal symmetry \( T \).

For both TLG and hBN, the honeycomb lattice can be bipartitioned into two triangular sub-lattices. A Dirac cone is generated in the electronic structure at the charge neutral point (CNP). The Dirac fermions become massive when the sublattice symmetry \( C_2 \cdot T \) which relates the two sub-lattices is broken \([15,16]\). In the hBN, boron and nitrogen atoms each form one of the sub-lattices, which breaks the symmetry between these two sub-lattices. This leads to a large energy gap (about 2.3 eV) in the low-lying excitations \([15]\). In contrast, the TLG itself is invariant under the sublattice symmetry, which protects the triple Dirac points near the Brillouin zone (BZ) corners. Thus the low-energy physics is dominated by the TLG, while hBN just contributes to a Moire scattering potential under second order perturbation. Such a Moire potential modulation folds the bands in the original BZ of the graphene layers into many mini-bands in the mini-Brillouin zones (mBZ), as displayed in Fig. 1b. As the mBZ is smaller by four order of magnitude compared to the scale of the folded bands, because the valley distance in the original BZ is significantly suppressed. And the mini-bands near the charge neutral point (CNP) mainly originate from the low-energy valleys \( (K' \text{ and } K'') \) shown in Fig. 1b in the original TLG \([17]\).

There are two crucial points about the Moire modulation of the band structure. First, the two valleys originally connected within one band are now significantly separated into two degenerate bands, because the valley distance in the original BZ is significantly longer than the characteristic wave vector of the Moire potential. This is the reason why the valley degree of freedom enters into the superlattice as the internal degrees of freedom of electrons. Second, as the triple Dirac cones are in fact split by trigonal warping process in the TLG \([14]\), the splitting distance is relatively small in the original BZ, but quite comparable to the scale of the folded mBZ (Fig. 2a and b). As a result, the flat dispersion between the Dirac cones dominate most area of the mBZ, which further suppresses the kinetic energy. Moreover, the Dirac point is gapped out by the interplay between the hBN and TLG, which breaks the sub-lattice symmetry. A valence band is thus separated from the other mini-bands by the Moire band gap (Fig. 2c), which has fourfold degenerate associated with the spin and valley degrees of freedom. Electrons around the valleys \( K' \) and \( K'' \) are related to each other by either one of the following transformations: the time-reversal symmetry \( T \), mirror reflection \( M_y \), and \( C_6 \) rotation.

Using the effective two-component Hamiltonian for the TLG \([14]\), we have calculated the band structures with the Moire scattering potential \( V_M \) assumed to act only on the bottom graphene layer \([13]\). Since the two valley bands are connected through the mirror transformation \( M_y \), we can just focus on the \( K \) valley. In Fig. 2c, the electronic structure for the bands of valley \( K \) is displayed. The contour plot of the corresponding valence band near the CNP is also shown in Fig. 2d. The three Dirac points originally at \( K' \) are separated to locate along the boundary of the mBZ towards \( K_d \), reducing the energy dispersion and inducing the triple van Hove singularity near \( K_d \). When the Dirac points are further gapped out, the remaining triple van Hove points are the most remarkable fingerprint of the Moire band structure. More precisely, three van Hove points actually line along the mBZ boundary and center around the zone corner \( K'_d \). Increasing the value of \( V_M \) pushes the three van Hove points towards \( K'_d \). Above all, due to the Moire scattering and the Dirac physics, the kinetic energy scale is quenched from 1 to 20 meV. Because the valence band is separated from the other bands, we are able to write a one-band minimal tight-binding model with valley and spin degeneracy.

Given the Moire mini-band structure, the minimal model should satisfy all the symmetries mentioned above, and reproduce the key feature of mini-valence-band: the triple van Hove points and ultra-flat dispersion. In the triangular Moire lattice sites
van Hove points. For the valley
the hopping parameter $t$ these van Hove points approach to
/symmetry. Right at $U$
indices, the fluxes alternate between the $b$
pair of pseudo-spin denoted by the Pauli matrices
Fig. 3. Proposed minimal lattice model, Fermi surfaces and their band structure. (a)
The TLG-hBN moire superlattice composed of three different zones labelled by $x$, $y$, and $z$, and the pattern of a valley contrasting staggered flux allowed by $C_3$, $M_z$, and
$T$ symmetries. The $x$ zones form the effective triangular lattice sites of the moire
superlattice. (b) 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 connected by reciprocal
unit vectors. (c) Band structures of the two valleys related by the $M_z$-symmetry. The
$m$
ectors of the primitive unit cell,
where $o$
messentially tunes those three
$\varepsilon_{\text{HV}}$ which reveals the essential correlated physics in the TLG-hBN
heterostructure.

3. Inter-valley spiral order in the half-filled Mott insulator

When $\Phi = \pi/2$ at half-filling, the Fermi surface becomes a perfect
triangle that touches the mBZ corners, as shown in Fig. 3b. In
this case, the two Fermi sheets are perfectly nested and linked by
three vectors $Q = (4\pi/3, 0)$ and $(-2\pi/3, \pm 2\pi/\sqrt{3})$. However, these
three nesting vectors are equivalent to each other, because they are
simply related by the reciprocal vector of the mBZ. More explicitly,
because of the particle-hole symmetry $\varepsilon_{\text{HF}} = -c_{\text{HF}, Q, +}$, we have the relation $\varepsilon_{\text{HF}} = -c_{\text{HF}, Q, -}$. In such a circumstance, the on-site Coulomb
interactions become important. Taking into account the most
relevant on-site Coulomb repulsion between valleys, we propose a
valley version of the Hubbard model:

$$H_V = \sum_{\Gamma} n_{\Gamma \uparrow} + n_{\Gamma \downarrow},$$

where $n_{\Gamma \sigma} = \sum_{\text{k}} c_{\Gamma \sigma, \text{k}}^\dagger c_{\Gamma \sigma, \text{k}}$ is the local electron density operator. The
perfect Fermi surface nesting motivates us to introduce the following
inter-valley order parameter

$$\Delta_Q = \sum_{\text{k}} (\varepsilon_{\text{k}, +}^{\sigma} + \varepsilon_{\text{k}, -}^{\sigma}),$$

to decouple the Coulomb interaction $V$-term in Eq. (4) as

$$H_V \simeq -\sum_{\text{k}} \left( \Delta_Q^2 + \varepsilon_{\text{k}, +}^{\sigma} + \varepsilon_{\text{k}, -}^{\sigma} \right),$$

where $\Delta_Q$ is a spatial uniform order parameter. Under the mean-field
approximation, the above model Hamiltonian can be diagonalized,
and the order parameter is determined by the self-consistent
equation

$$\int_{\text{mBZ}} \frac{\sqrt{3} d\text{k}_x d\text{k}_y}{16\pi^2} \frac{V}{\sqrt{\varepsilon_{\text{k}, +}^{\sigma} + \Delta_Q^2}} = 1,$$

which is similar to the BCS gap equation. If we further assume that
the overall mBZ contribution is dominated by a narrow shelf of
width $D$ around the Fermi energy, the solution to the above
equation is then given

$$\Delta_Q \simeq D e^{-\pi N(0)},$$

where $N(0)$ is the density of states at the Fermi level. At half-filling,
$N(0)$ diverges, and an infinitesimal interaction $V$ can induce a finite
inter-valley long-range order and gap out the Fermi surfaces
completely. This has been confirmed by the numerical solution to the
self-consistent equation, as shown in Fig. 4a. Actually this is a very
peculiar insulating state induced by the inter-valley scattering $V$. In
real space, $\Delta_Q$ describes an inter-valley spiral long-range order of the
valley pseudo-spin:
played in Fig. 4b. Therefore, it is this inter-valley spiral phase that describes the Mott insulating phase observed by the experiment [13].

By assuming\(\frac{w}{C_0}=\frac{v^2}{C_0}\), the valley-pseudo-spin operators have been replaced by the spin-singlet antiferro-valley 120° ordering state. When the spin degrees of freedom is retrieved, however, the corresponding insulating phase at quarter-filling is given by the spin-polarized antiferro-valley 120° ordering state, while the half-filling insulating phase will be replaced by the spin-singlet antiferro-valley 120° ordering state.

5. Discussion and conclusion

Compared with the magic-angle twisted bilayer graphene [11], the kinetic energy of both systems is suppressed by the band folding, resulting in a similar Moire super-lattice and the mBZ. The Dirac cones of the twisted bilayer graphene are separated and hybridized, yielding the van Hove singularity at \(M_1\) point and flat dispersion in between. Similarly, in the TLG-hBN, the triple Dirac cones from the ABC stacked trilayer are separated by the trigonal warping with a strong hybridization, yielding the triple van Hove points and flat dispersion in between. The drastic distinction between these two systems is reflected in their symmetries and Fermi surface structures of the Moire bands. More precisely, with respect to the same Moire triangular super-lattice, the TLG-hBN is invariant under the symmetry \(M_{1}\) while the twisted bilayer preserves the symmetry \(M_{1}M_{2}\) instead. Consequently, the \(C_{1}\) symmetric Fermi surfaces in the latter are distinct from that of the former by 30 degree rotation [10]. If the Fermi surfaces have a nesting effect, the three nesting vectors are no longer connected by the reciprocal lattice vector. Then the twisted bilayer graphene will be subjected to an inter-valley triple-\(\mathbf{Q}\) nesting, and the inter-valley Coulomb repulsion induces a drastically distinct Mott insulating phase.

What experimental signature could be observed for this order? In the large length scale of Moire super-lattice characteristic of \(\lambda_M = 15\) nm, where the valley degrees of freedom are treated as the internal degrees of freedom inside each Moire supercell, the order with long-range wave vector \(\mathbf{Q}\) exhibits an in-plane spiral feature of the valley pseudo-spins, but does not involve a density spatial modulation running over the Moire superlattice. Nevertheless, when zooming into the small length-scale of the original graphene lattice characteristic of \(\lambda = 0.246\) nm, the valley degrees of freedom retain their orbital character. The nesting between the two valleys in the original BZ gives rise to a short range nesting wave vector, corresponding to a \(\sqrt{3}a\)\(\times\)\(\sqrt{3}a\) charge modulation pattern in the microscopic lattice.

Moreover, in the magic-angle twisted bilayer graphene, unconventional superconductivity was also observed slightly away from...
the half-filling [12]. Naturally, one would ask whether this TLG-hBN heterostructure could also become a superconductor by doping away from the half-filling. If yes, what is the most probable pairing symmetry. From our above analysis, the inter-valley scattering should still be the most important channel of pairing interactions, because the intra-valley pairing is not energetically favored due to the peculiar Fermi surface structures. If only the inter-valley Coulomb repulsion is considered, there is no privilege between spin singlet and spin triplet pairing. However, the inter-valley Hund’s rule coupling favors a spin-triplet pairing state. Therefore, the superconducting state in the TLG-hBN is expected to be in the inter-valley spin-triplet pairing channel. A detailed discussion on this will be given in a separate paper.

In conclusion, we have proposed a minimal tight-binding model to describe the low-energy states of the TLG-hBN super-lattice. Compared to the low-energy effective bands, the valley-contrasting staggered flux of each Moire triangle acquires the value of $\pi/2$. At half-filling, the Fermi surfaces are perfectly nested between the two valleys. This leads to a strong inter-valley scattering and the system becomes unstable against an inter-valley spiral order. We believe that this inter-valley spiral ordered phase is just the Mott insulating phase observed in the experiments [13].

Note added: while in the preparation of this work, we noticed that two preprints [18,19] on the model for magic-angle twisted bilayer graphene appear. One of them [18] proposed a similar tight-binding model for the TLG-hBN heterostructure.

**Conflict of interest**

The authors declare that they have no conflict of interest.

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