Mean-Field Continuum Model of the Twisted Kitaev Bilayer

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Kitaev quantum spin liquids have been the focus of intense research effort thanks to the discovery of various materials (e.g., RuCl$_3$) that approximate their intriguing physics. In this paper we construct a mean-field approximation for a moiré superlattice emerging in twisted Kitaev bilayers in terms of solutions of commensurate bilayers. We show that the band structure of deconfined spinons, defined on the mini-Brillouin zone of the superlattice, is greatly modified. Bands which are almost perfectly flat appear at energies above the lowest gap. Including intralayer modulation, such bands become isolated from other dispersive ones. Intriguingly, flat-band eigenstates exhibit a localization akin to wavefunctions of Kagome lattices.

Introduction—Van-der-Waals heterostructures [1, 2], composed of two-dimensional (2D) materials held together by weak dipolar forces, have recently experienced a surge of interest after the discovery of strongly-correlated phases in transition-metal halides [3–9] and twisted stacks [10–16]. Transition-metal halides are a family of exfoliable materials which exhibit several types of ordered phases down to monolayer thicknesses [3–9]. Among such materials, RuCl$_3$ features Kitaev quantum spin liquid (QSL) phases activated by either temperature or magnetic field [17–25]. RuCl$_3$ is part of a broader class of strongly-correlated materials (which includes, e.g., the iridates [26–30]) that, thanks to a combination of spin-orbit coupling, crystal symmetry and interference between superexchange paths [25, 31–33], are predicted to exhibit a dominant Kitaev coupling [25, 34–38] and QSL behavior [35] (ideally down to absolute zero, though in real materials order is observed at low temperatures [17–25]).

QSLs, whose excitations can feature a nontrivial statistics [35], are a form of topological order [38]. Their behavior is at odds with that of conventional magnets, whose excitations (the magnons) are usually bosons. On the contrary, Kitaev QSLs can support excitations (called “spinons”) which behave as fermions or Majorana particles [35]. Neutron-scattering [19, 20] and Raman [21] measurements support this picture. In the presence of a magnetic field, the spectrum of excitations becomes gapped and Majorana particles are predicted to localize at sample edges or defects. This is compatible with the pronounced thermal Hall effect observed [39–44] in RuCl$_3$. The magnetic field also allows a distinct QSL phase, whose nature is the subject of an intense debate in the literature [45–54], to emerge.

Long-periodicity moiré superlattices, obtained by twisting layers with identical lattice structures, can greatly modify the properties of the heterostructure [16, 55–62] (see also Refs. [63, 64] and references therein). For example, they can turn two weakly-correlated sheets of graphene into a strongly-correlated heterostruct-
nal quantum spin-1/2 system with bond dependent Ising-like interactions. This model provides a qualitative understanding of the low-energy physics [32] of RuCl$_3$. In fact, the lattice of RuCl$_3$ is composed by transition-metal (Ru) ions encased in halogen (Cl) octahedra organized in a hexagonal lattice. The strong spin-orbit coupling and crystal field split the $t_{2g}$ multiplets of Ru atoms, leaving effective spin-1/2 quantum variables coupled (via superexchange through Cl atoms) with a bond-dependent interaction of the Kitaev type [32].

Commensurate Kitaev bilayers, obtained by coupling two hexagonal lattices, have been addressed in Refs. [70–72]. In particular, in Ref. [70] bilayers with perfect atom-on-atom stacking have been studied within a mean-field approximation. There, it is shown that both the intralayer and interlayer hopping amplitudes of Majorana excitations depend on the interlayer spin coupling $J$. For small values of $J$, the two layers are exactly decoupled, i.e. spinons cannot tunnel between the two. On the contrary as $J$ is increased (to about $\sim 0.44$ times the intralayer Kitaev coupling, $K$), interlayer hopping turns on and grows with it. Notably, when this happens the intralayer hopping amplitude decreases, i.e. spinons are less mobile in each layer, and vanishes for $J \simeq 0.58K$. For larger values of $J$, the two layers are strongly coupled and spinons cannot propagate within a given layer. We find that in an AB bilayers, where only half of the atoms of one layer sit on top of atoms of the other layer, the boundaries between the three phases differ from the corresponding commensurate stacking. At small twist angles, the pattern is approximately periodic, and it is possible to identify a well-defined moiré supercell. However, since our calculations for commensurate stacking configurations. In the following we consider a moiré heterostructure consisting of two Kitaev monolayers, with a relative twist between them.

The monolayer Kitaev model is defined on a hexagonal lattice, whose unit cell is spanned by a pair of vectors $a_{1,2} = (1/2, \pm \sqrt{3}/2)$. The vectors $\tau_c = (a_1 - a_2)/3$, $\tau_x = (2a_2 + a_1)/3$, $\tau_y = -(a_2 + 2a_1)/3$ connect elements of the A sublattice to their nearest neighbors on the B sublattice [see Fig. 1(b)]. The reciprocal lattice is spanned by the vectors $g_{1,2} = b(\sqrt{3}/2, \pm 1/2)$, where $b = 4\pi/\sqrt{3}$. These define the maximally-symmetric hexagonal Brillouin zone (BZ) with corners at the inequivalent points $K_{1,2} = \pm (g_1 + g_2)/3$. Spin-1/2 quantum variables are located at each site and are coupled with a bond-dependent Ising interaction. The $x$, $y$ and $z$ components of the spins are coupled along the blue, green and red directions shown in Fig. 1(b), respectively. The solution of the Kitaev models passes through the fractionalization of each spin variable in terms of four Majorana particles [35]. Of these, three are paired between neighboring sites to form a gauge field on top of which the fourth (deconfined) one propagates (more details are given below). Since the gauge potential commutes with the Hamiltonian and with itself on different bonds, the model reduces to a quadratic Hamiltonian for deconfined particles, which can thus be solved exactly [35]. The energy dispersion is very similar to graphene’s, i.e. it is gapless and exhibits Dirac cones at the points $K_{1,2}$.

The twisted heterostructure is generated from a commensurate bilayer structure by applying a rigid rotation $R(\pm \theta/2)$ to the top and bottom layers respectively. At zero twist angle, bonds of equal type [i.e. $x$, $y$, $z$ or red, green, blue in Fig. 1(b)] are chosen to be exactly aligned on top of each other. The resulting moiré pattern has an associated reduced moiré Brillouin zone (MBZ) in momentum space [Fig. 1(c)], while its reciprocal lattice is spanned by vectors $G_{1,2} = g_{1,2}^t - g_{1,2}^b$, where $g_{a,b}^t \equiv R(\pm \theta/2)g_a$, are the rotated vectors of the top/bottom layers [Fig. 1(d)]. The real-space superlattice is thus characterized by vectors $L_{1,2}$ [Fig. 1(a)] which satisfy $L_i \cdot G_j = 2\pi \delta_{ij}$. For small angles one can approximate each region of the heterostructure by the corresponding commensurate stacking.

Commensurate bilayers are uniquely characterised by the in-plane displacement $d$ of the top layer relative to the bottom one. AB stacking occurs where $d = \tau_c$, such that the A sublattice in the top layer is aligned with the B sublattice in the bottom layer, while B atoms of the top layer lie in the middle of hexagons of the bottom layer. Similarly, BA stacking occurs where $d = -\tau_c$, and...
where \( i \) labels the unit cell, and \( K_\alpha \) is a bond-dependent Kitaev coupling. Vectors with superscript \( l \) are rotated according to their layer as \( v^{l,b} = R(\pm \theta/2)v \), while \( r_i \) is a coordinate defined in a global reference frame. Each spin may be represented by a set of four Majorana fermions \( \{ c, b^\sigma, b^\sigma, b^\sigma \} \) such that \( S^\alpha = i b^\alpha c \). Moving to the Majorana fermion representation and performing a mean-field decoupling, one then obtains

\[
H^l_K = -\frac{i}{2} \sum_{\alpha} \sum_i K_\alpha \langle u^\alpha_i(r_i) \rangle c^\dagger_{\alpha,\ell,\ell} c_{\alpha,\ell,\ell} + u^l_{0,\alpha}(r_i) b^\alpha_{\ell,\ell} b^\alpha_{\ell,\ell},
\]

where the mean-field components are defined as \( u^\alpha_i(r_i) = \langle i b^\alpha_{r_i,\ell,\ell} b^\alpha_{r_i,\ell,\ell} \rangle \) and \( u^l_{0,\alpha}(r_i) = \langle i c^\dagger_{r_i,\ell,\ell} c_{\alpha,\ell,\ell} + \tau^\alpha_{\ell,\ell} \rangle \) respectively. Following Ref. [70], we choose not to include mean fields mixing \( b \)- and \( c \)-Majorana particles at the same lattice site. These can be used to characterize classical order, which is what we want to avoid here.

In Eq. (2) we have effectively separated the full quartic Hamiltonian into four separate (quadratic) tight-binding problems, one for each Majorana species. The effective Hamiltonians of itinerant Majorana fermions in a given layer, \( H^l \), is given by the first line of Eq. (2). Direct calculation of \( u^\alpha_i(r_i) \) is impractical, except in the simplest commensurate cases where translation invariance may be utilized to take them to be independent of \( r_i \), thus dramatically reducing the number of independent mean-field components. Fig. 2 shows mean-fields \( u^l_{\alpha,S} \) and \( u^l_{\alpha,B} \) calculated as in Ref. [70] for two stacking configurations, \( S = AA \) and \( S = AB \). The boundaries between different phases appears shifted, for the AB stacking, to values of \( J/K \) which are roughly twice of those found for the AA configuration. This finding can be interpreted as the result of the halved number of interlayer nearest-neighbours in AB bilayers. In each commensurate configuration, the signs of mean fields can be flipped via a gauge transformation. therefore, only their moduli have a physical meaning. On the other hand, in a moiré superlattice one needs to fix these signs in such a way that, in certain limiting condition, the behavior of the structure recovers the expected one. We set \( u^l_{\alpha,B} = u^l_{\alpha,AB} \), i.e. mean-fields have the same sign in AB and BA regions. This choice is dictated by the requirement that, in the limit of zero interlayer coupling, one recovers the Hamiltonian describing two independent Kitaev layers.

We construct the twisted stack mean-fields by interpolating those of commensurate bilayers as \( u^l_{\alpha}(r_i) = \sum_{S} u^l_{\alpha,S} T(r_i - r_S) \), where \( r_S \) denotes the positions of the three stacking configurations in the moiré unit cell [Fig. 1(a)]. We choose the periodic function \( T(r) = (1/9) \sum_{n=1}^3 (1 + e^{iG_n \cdot r} + e^{-iG_n \cdot r}) \), since it preserves the threefold rotational symmetry of the superlattice while matching the periodicity of the supercell. \( G_{1,2} \) are the previously defined reciprocal lattice vectors for the Moiré superlattice, and \( G_3 = G_2 - G_1 \). \( T(r) = 1 \) \( \{ T(r) = 0 \} \) for \( r = r_{AA} \) \( \{ r = r_{AB}, r_{BA} \} \). Therefore, \( u^l_{\alpha}(r) \) represents the simplest real-valued periodic function on the superlattice which smoothly interpolates between the values of the mean-field parameters in regions approximated by AA, AB, and BA commensurate stacking configurations respectively. The intralayer term may then be written in momentum space representation as

\[
H^l = -\frac{i}{6} \sum_{k} \sum_{n=1}^3 \left( f^l(k,0) c^\dagger_{k,\ell,\ell} c_{k,\ell,\ell} + f^l(k,-G_n) c^\dagger_{k,\ell,\ell} c_{k-G_n,\ell,\ell} + f^l(k,G_n) c^\dagger_{k,\ell,\ell} c_{k+G_n,\ell,\ell} \right),
\]

where \( k \) is measured from the Brillouin zone center and \( f^l(k,G) = (1/3) \sum_{n=1}^3 K^\alpha u^l_{\alpha,S} e^{iG_r S_r} e^{i(k+G) \cdot \tau^\alpha_n} \).
tween itinerant Majorana fermions

\[ H_{\perp} = -iJ \sum_{n=0}^{2} \sum_{k,\mu} w_{\alpha,\mu} c_{\alpha}^{\dagger} (g_n e^{i\tau_n} - g_n^{\dagger} e^{-i\tau_n}) c_{\alpha} \equiv \mathcal{G}_n \tau \cdot \mathbf{r} \]

where \( \mathcal{G}_n \equiv G_n - \Delta K \), \( g_0 = G_0 = 0 \), and \( \Delta K = (G_1 + G_2)/3 \). \( w_{\alpha,AA} \), \( w_{\alpha,AB} \), and \( w_{\alpha,BA} \) are parameters to be derived from the Majorana mean-field theory of AA, AB, and BA stacked bilayers respectively, and \( J \) is the Heisenberg exchange. In analogy with intralayer mean fields, we have \( w^{l}_{AB} = w^{l}_{BA} \). We also take \( w_{\alpha,BA} = w_{\alpha,AA} \). \( k \) is measured from the Brillouin zone center despite (4) being valid only around the Dirac points, so as to maintain consistency with the intralayer terms. In all calculations we work in a neighbourhood around \( (K_1^0 + K_1^1)/2 \). Both intra- and interlayer terms simplify considerably in the isotropic limit where \( K_x = K_y = K_z = K \); the mean-field components \( w^{l}_{\alpha,S} = w^{l}_{S} \), and \( w^{l}_{\alpha,S} = w^{l}_{S} \) become independent of bond direction. We note that although these mean-fields enter as free parameters in our equations, their values are determined by the (physical) spin couplings \( J \) and \( K \) via the solution of the commensurate bilayers.

Results—The \( k \)-dependent Hamiltonian is expanded in a plane-wave basis [83, 84] and diagonalised to yield the itinerant Majorana fermion spectrum. We calculate the spectrum across the first MBZ around \( (K_1^0 + K_1^1)/2 \), and study the isotropic limit for simplicity. The band structure is sensitive to the ratio between Heisenberg and Kitaev coupling, \( J/K \). In fact, as shown in Fig. 2(a), the mean fields (and therefore intra- and inter-layer hopping amplitudes) depend on this quantity.

As shown in Fig. 2(a), for \( J/K \lesssim 0.44 \), both \( w_{AA} \), \( w_{AB} = 0 \), and the system consists of two effectively decoupled monolayers with maximal intralayer hoppings \( w^{l}_{S} = \pm 0.5 \). We refer to the values of the mean-fields in this regime as the Kitaev QSL values. We stress that, in this case, the mean-field solution is exact. The band structure obtained in this regime is shown in light gray in Figs. 2(b) and (c) as baseline for those of coupled bilayers. It corresponds to the folding of Dirac cones of isolated layers within the mini-Brillouin zone of a twisted (decoupled) bilayer. The band structure presents a linear low-energy dispersion around the corners of this smaller zone \( (K \) and \( K') \), and it is degenerate at higher energies at both the \( M \) (middle point of the hexagonal mini-Brillouin zone) and \( \Gamma \) points. In particular, higher bands are six-fold degenerate at the latter.

For \( 0.44 \lesssim J/K \lesssim 0.58 \), \( w^{+}_{AA} \) and \( w^{+}_{AA} \) deviate from their QSL values, while the mean-fields for the AB stacking remain unchanged, i.e. the interlayer couplings \( w_{AB} = w_{BA} \) vanish exactly. Under these conditions the interlayer coupling becomes finite over the entire supercell, but is reduced in the neighborhoods of \( r_{AB} \) and \( r_{BA} \), and vanishes exactly at those points. Conversely, the intralayer hopping is minimum around AA regions. As \( J/K \) enters in this range of values, the sixfold degeneracy at \( \Gamma \) breaks, and bands split off and begin to flatten. There is no low-energy gap opening at the points \( K \) and \( K' \) even for large \( J/K \), although the lowest band becomes increasingly energetically isolated from the rest.

Notably, very flat higher-energy bands appear close to other dispersive bands for \( J/K \gtrsim 0.5 \). In the absence of modulation of intralayer hopping amplitudes, flat and dispersive bands are degenerate at the \( \Gamma \) point. The degeneracy is broken when the non-uniformity of intralayer terms is included. Visually, the group of bands above the lowest one in Fig. 2(c) strongly resembles the spectrum of a Kagome lattice. The latter tri-partite model exhibits two dispersive bands touching a perfectly-flat low-energy band at the \( \Gamma \) point. To verify whether Kagome physics emerges in TKBs, we analyze the wavefunction of the flat band. Fig. 3 shows the probability density of the flat-band eigenstate at the \( \Gamma \) point for \( J/K = 0.5 \). Intriguingly, the wavefunction projected on the \( A \) sublattice [Fig. 3(b)] produces a clear Kagome pattern in real space. It is also instructive to look at the phase of the wavefunction. This presents vortices of winding number

![Fig. 2](image-url)

Panel (a) Mean-field solutions for AA (top) and AB (bottom) stacked bilayers as a function of \( J/K \) in the isotropic limit. Panel (b) In blue, the spectrum of itinerant Majorana fermions for \( J/K = 0.45 \). Here, the twist angle is \( \theta = 1.1^\circ \). The grey background represents the bandstructure of decoupled twisted Kitaev bilayer \( (J/K = 0) \). Panel (c) Same as in Panel (b), but for \( J/K = 0.5 \). \( K \) and \( K' \) refer to \( K_1^1 \) and \( K_1^0 \) of Fig. 1(c) respectively.
Here, $J/K$ centered about the $A$ (left) and $B$ (right) site of the top layer. Images are shown that such terms allow one to isolate flat from dispersive bands.

In the Supplemental Material [85] we show band structures and wavefunctions calculated for higher values of $J/K$, which exhibit a higher degree of flattening. There, we also show results obtained for unphysical choices of $\pm 1$ at the AB and BA stacking configurations (the corners of the hexagonal superlattice unit cell), where the wavefunction itself vanishes exactly.

Conclusion—By considering the variations in local stacking across a moiré heterostructure and their effect on mean-field parameters, we have been able to build a mean-field approximation for a twisted bilayer of Kitaev quantum spin liquids. We have constructed a working approximation for intra- and inter-layer tight-binding hopping amplitudes of deconfined Majorana particles within a continuum model in terms of solutions of mean-field theories on different commensurately stacked bilayers. Accounting for these effects yields a complex band structure, which exhibits flat bands and features resembling Kagome lattices.

While preparing this manuscript, we became aware of a related work [86]. While our results for small values of $J/K$ agree with Ref. [86], we successfully capture the effect of the varying stacking arrangement on the intralayer hopping, which was neglected in other works. We have shown that such terms allow one to isolate flat from dispersive bands.

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