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Excitonic Chern insulator and kinetic ferromagnetism in MoTe₂/WSe₂ moiré bilayer

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We propose a new mechanism for the quantum anomalous Hall (QAH) effect in the AB stacked MoTe₂/WSe₂ system. Based on the observation that the inter-layer tunneling is suppressed in the AB stacking, we consider a model with two layers coupled through the Coulomb interaction. The moiré lattices of the two layers are shifted to form a honeycomb lattice. Initially, the system is in a layer-polarized Mott insulator at $\nu_T = 1$. But with a displacement field, an equal number of holes and electrons are doped into the two layers respectively, forming inter-layer exciton condensation. Through mean field theory, we find $p \pm ip$ exciton condensation in a certain parameter regime, which leads to a Chern insulator with Chern number $C = \pm 1$. The valleys are polarized due to the kinetic energy instead of interaction. But the polarization in the two layers can be either the same or opposite. Especially, an inter-valley-coherent (IVC) Chern insulator phase is possible, in agreement with the recent magnetic circular dichroism (MCD) measurement. Our work opens a new direction to search for a topologically non-trivial excitonic insulator with high angular momentum exciton pairing symmetry.

Introduction Chern insulator is one of the famous topologically non-trivial states, which supports the quantum anomalous Hall (QAH) effect with a quantized Hall conductivity at zero magnetic field¹. Although the Chern insulator phase and QAH effect was theoretically proposed to be realized in the Haldane model in 1988², its experimental realization appeared only after 2013. The first realization is on the surface of the three-dimensional topological insulator³. The observations of the QAH effect^{4–10} in truly two-dimensional systems were achieved only a few years ago in the moiré systems^{4–6,9,11–27}.

QAH was first predicted theoretically for the graphene moiré systems²⁸. The moiré superlattice folds the original band into narrow moiré bands in a mini Brillouin Zone (MBZ). Especially the two valleys K and K' form their own moiré bands, which can have non-zero but opposite Chern numbers²⁸. Then the interaction can polarize the valley at filling $\nu_T = 1$ due to the Stoner ferromagnetism mechanism. After that, the Chern band of one valley is fully occupied, leading to a Chern insulator phase. After the experimental observations in several graphene systems^{4–10}, many other theoretical studies^{29–39} confirm this simple picture as the mechanism for the QAH phase in graphene-based moiré systems. In this scenario, interaction is needed for valley ordering, but the band topology already exists at the single particle level. One natural question is then whether a Chern insulator phase is possible without single-particle band topology. We will propose a new mechanism towards QAH with band topology generated by exciton condensation, motivated by the recent experimental measurements of a QAH phase in a moiré system based on the AB stacked transition metal dichalcogenide (TMD) MoTe₂/WSe₂ bilayer^{27,40}.

In the AB stacked MoTe₂/WSe₂ hetero-bilayer^{27,40}, a Mott insulator evolves to a Chern insulator phase at den-

sity $\nu_T = 1$ per moiré unit cell when tuning the displacement field. There is evidence that the Chern insulator phase is in the regime where particles are transferred from the MoTe₂ layer to the WSe₂ layer, so the densities at the two layers are $1 - x$ and x respectively. Previous theories^{41–47} all predict a phase with aligned valley polarizations in the two layers. Because these theories rely on single particle tunneling term to couple the two layers to form a band, the valleys naturally align because the hopping term must be between the same valley due to the momentum conservation. However, the recent experimental measurement of magnetic circular dichroism (MCD) indicates that the valleys in the two layers within the QAH phase are anti-aligned⁴⁰. It is clear that all of the previous theories are inconsistent with this measurement and a new theoretical explanation is needed for this system.

In this paper, we propose a completely new route towards QAH in AB-stacked TMD hetero-bilayer. Based on the observation that the inter-layer tunneling is suppressed in the AB stacking because the spin S_z in the two layers for the same valley are opposite (see Fig. 1(a)), we take the view that the most important coupling between the MoTe₂ and WSe₂ layer is the Coulomb interaction. The physics is then similar to the recently proposed^{48,49} and experimentally fabricated^{23,24} Coulomb coupled moiré bilayer. The major difference now is that the lattices of the two layers are shifted to form a honeycomb lattice together instead of being aligned. Then the electron and hole pockets in the two layers can form exciton condensation due to the inter-layer Coulomb interaction. In certain parameter regions, we find that the pairing symmetry of the exciton condensation is $p \pm ip$, similar to the $p \pm ip$ superconductor⁵⁰ after a particle-hole transformation only for one layer. This leads to a topological phase we dub excitonic Chern insu-

lator (ECI). In this case, we can have either intra-valley exciton condensation or inter-valley exciton condensation, which are nearly degenerate. With small valley contrasting flux of appropriate sign, inter-valley exciton condensation is selected and we reach an ECI phase with inter-valley-coherence (IVC), consistent with the MCD measurement⁴⁰. Our theory points out the possibility of a topological excitonic insulator phase with higher angular momentum exciton pairing symmetry, which was not well explored before.

Model For single-layer TMD, due to the Ising spin-orbit coupling, the spin and valley degrees of freedom are locked. For AB stacked MoTe₂/WSe₂ system, the spin-valley locking is opposite in the two layers, as shown in Fig. 1(a). For the WSe₂ or A layer, we have $(\tau_A^z, s_z) \in \{(K_A, \downarrow), (K'_A, \uparrow)\}$. For the MoTe₂ or B layer, we have $(\tau_B^z, s_z) \in \{(K_B, \uparrow), (K'_B, \downarrow)\}$. Here K_a, K'_a labels the two valleys for the layer $a = A, B$. τ_i, s_i with $i = x, y, z$ label Pauli matrices in the valley and spin space respectively. We also use σ_i with $i = x, y, z$ to label Pauli matrices in the layer space. In the following spin index will be ignored and the two valleys K, K' will be labeled as $+, -$. WSe₂ and MoTe₂ layers will be labeled as A, B .

We use a honeycomb lattice model for the MoTe₂/WSe₂ bilayer similar to Ref. 42, where Wannier orbitals from WSe₂ and MoTe₂ live on A and B sublattice sites (see Fig. 1(b)). The essential physics is captured by a simple lattice model:

$$\begin{aligned}
 H = & -t_{AB} \sum_{\langle ij \rangle} (c_{i;\alpha}^\dagger c_{j;\alpha} + h.c.) + D \sum_i (-1)^{\sigma_z(i)} n_i \\
 & - t_A \sum_{\langle\langle ij \rangle\rangle_A} (e^{i\phi_{ij}^A \tau_z} c_{i;\alpha}^\dagger c_{j;\alpha} + h.c.) \\
 & - t_B \sum_{\langle\langle ij \rangle\rangle_B} (e^{i\phi_{ij}^B \tau_z} c_{i;\alpha}^\dagger c_{j;\alpha} + h.c.) \\
 & + \frac{U}{2} \sum_i n_i(n_i - 1) + V \sum_{\langle ij \rangle} n_i n_j + V' \sum_{\langle\langle ij \rangle\rangle} n_i n_j \quad (1)
 \end{aligned}$$

where $\alpha = +, -$ labels the valley index and we assume Einstein summation convention for α . $n_i = \sum_{\alpha=+,-} c_{i;\alpha}^\dagger c_{i;\alpha}$ is the density at site i . $\sigma_z(i) = 1, -1$ for A and B layer respectively. The two unit vectors of the lattice are $\mathbf{a}_1 = (1, 0)$ and $\mathbf{a}_2 = (-\frac{1}{2}, \frac{\sqrt{3}}{2})$. $\phi_{i;i+\mathbf{a}_1}^a = -\phi_{i;i-\mathbf{a}_1}^a = \phi_a$ is the valley contrasting hopping term for layer $a = A, B$. ϕ_{ij}^a of the other bonds are generated by C_3 symmetry. $\Phi_a = 3\phi_a$ is the valley contrasting flux through each triangle of the layer a . In the above $\langle ij \rangle$ labels the nearest neighbor AB bond. $\langle\langle ij \rangle\rangle_a$ labels the intra-layer nearest neighbor bond of the triangular lattice in the layer $a = A, B$. We expect $\phi_A \approx \frac{2\pi}{3}$ and $\phi_B \approx 0$ so that the band bottom of the two layers are at $\mathbf{K}_M$ ($\mathbf{K}'_M$) and $\mathbf{\Gamma}_M$ in the mini Brillouin zone (MBZ). Time reversal symmetry flips the valley τ_z for both layers. Despite the superficial similarity to the model in Ref. 42 and Ref. 45, we will consider the limit $\frac{V}{t_{AB}} \rightarrow \infty$,

while the inter-layer interaction V term was ignored in Ref. 42 and Ref. 45.

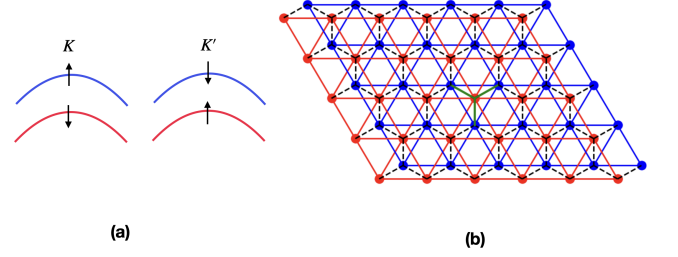


FIG. 1: (a) Illustration of the spin valley locking in MoTe₂/WSe₂ hetero-bilayer. Red and blue lines are the top valence band for the WSe₂ and MoTe₂ layer respectively. In AB stacking, one layer is rotated by 180° compared to AA stacking, which flips the spin-valley locking in this layer. (b) Lattice model on the honeycomb lattice. Red and blue labels A and B sublattice sites, corresponding to WSe₂ and MoTe₂ layer. The solid line is the nearest neighbor bond within each layer. The dashed line is the AB bond connecting the sites of the two layers. In our model, the microscopic hopping t_{AB} is assumed to be zero, but there can be effective inter-layer hopping spontaneously generated from exciton condensation. The exciton condensation order parameter is $\chi_{ij;\alpha\beta} = \langle c_{i;\alpha}^\dagger c_{j;\beta} \rangle$, where $\langle ij \rangle$ is illustrated as the green bond in (b).

In most of the paper, we assume $t_{AB} = 0$ for simplicity and will discuss its effect later. The global symmetry is then $U(1)^4$ at $t_{AB} = 0$, corresponding to the conservation of the four flavors combining layer and valley indices. If $\Phi_a = 0$, then the symmetry is enlarged to $U(1) \times U(2)$, with an $SU(2)$ rotation symmetry in the valley space only for the layer A . When $\Phi_A = \Phi_B = 0$, the symmetry is $U(2) \times U(2)$. When $t_{AB} = 0$, we are free to do a gauge transformation for only one layer to shift ϕ_a by $\pm \frac{2\pi}{3}$. Thus we can focus on the regime with $\phi_A \approx \phi_B \approx 0$. On the other hand, a finite t_{AB} reduces the symmetry to $U(1) \times U(1)$ with only total charge and total valley conservation for generic values of Φ_A, Φ_B . In this case, ϕ_A, ϕ_B can not be gauged away.

Let us focus on the filling with $\nu_T = n_A + n_B = 1$. Although in the experiment the valence band is hole-doped, here we use an electron picture after a particle-hole transformation. We start from large D where all of the electrons are in the B layer and there is a Mott insulator due to large U/t_B . When D is decreased, the B layer is hole-doped and the A layer has an equal number of electrons. A convenient way to deal with the large U term is to project to the Hilbert space without double occupancy and consider a bilayer t-J model:

$$\begin{aligned}
H = & -t_A \sum_{\langle\langle ij \rangle\rangle_A} (e^{i\phi_{ij}^A \tau_z} c_{i;\alpha}^\dagger c_{j;\alpha} + h.c.) + D \sum_{i \in A} n_i \\
& - t_B \sum_{\langle\langle ij \rangle\rangle_B} (e^{i\phi_{ij}^B \tau_z} P c_{i;\alpha}^\dagger c_{j;\alpha} P + h.c.) - D \sum_{i \in B} n_i \\
& + J_B \sum_{\langle\langle ij \rangle\rangle_B} \tau_i^\mu R_z^{\mu\nu} (2\phi_{ij}^B) \tau_j^\nu + (V' - \frac{J_B}{4}) \sum_{\langle\langle ij \rangle\rangle_B} n_i n_j \\
& + V \sum_{\langle ij \rangle} n_i n_j + V' \sum_{\langle\langle ij \rangle\rangle_A} n_i n_j \quad (2)
\end{aligned}$$

where P is the projection operator to forbid double occupancy. $R_z^{\mu\nu}(\phi)$ is the matrix representing rotation around z axis (see the supplementary⁵¹). For $U/t_B = 50$ ⁴⁵, $J_B = \frac{4t_B^2}{U} = 0.08t_B$. The effect of J_B is negligible. For simplicity, we do not impose a double-occupancy constraint for the A layer which is appropriate in the small n_A limit.

Phase diagram We can obtain a phase diagram by Hartree-Fock calculation directly for the Hubbard model in Eq. 1. We find valley polarization at an intermediate value of D and an excitonic Chern insulator (ECI) in a certain parameter regime. To make the mechanism of the magnetic order transparent, it is more convenient to work on the bilayer t-J model in Eq. 2 and use the Schwinger boson parton construction for the B layer: $c_{i;\alpha} = b_{i;\alpha} f_i$ with the constraint $\sum_\alpha b_{i;\alpha}^\dagger b_{i;\alpha} = f_i^\dagger f_i$ at each site $i \in B$. $b_{i;\alpha}$ is Schwinger boson carrying the valley degree of freedom while f_i is the spinless holon operator. The magnetic order at layer B is decided by the condensation of the Schwinger boson: $\left(\begin{smallmatrix} \langle b_{i;+} \rangle \\ \langle b_{i;-} \rangle \end{smallmatrix} \right) = F_i$. For valley ferromagnetic (FM) state with $+$ valley, we use $F_i = \sqrt{n_a} \begin{pmatrix} 1 \\ 0 \end{pmatrix}$.

On the other hand, if there is a 120° anti-ferromagnetic (AF) order, we use $F_i = -\frac{\sqrt{n_a}}{\sqrt{2}} e^{\frac{i}{2}(\mathbf{Q} \cdot \mathbf{r}_i) \tau_z} \begin{pmatrix} 1 \\ 1 \end{pmatrix}$, where $\mathbf{Q} = \mathbf{K}_M$ or $-\mathbf{K}_M$. For the A layer, we assume full valley polarization as found in the Hartree Fock calculation. Then there is only spinless charge degree of freedom which we also label as f_i for $i \in A$.

After the magnetic orders are decided for both layers, the charge degree of freedom is conveniently captured by the following spinless model:

$$\begin{aligned}
H_f &= H_0^f + H_V^f \\
H_0^f &= - \sum_{a=A,B} \sum_{\langle\langle ij \rangle\rangle_a} t_{ij;a}^f f_i^\dagger f_j + D \sum_{i \in A} n_i - D \sum_{i \in B} n_i \\
H_V^f &= V \sum_{\langle ij \rangle} n_i n_j + \sum_{a=A,B} \sum_{\langle\langle ij \rangle\rangle_a} V' n_i n_j \quad (3)
\end{aligned}$$

where $t_{ij;B}^f = t_B (F_i^\dagger e^{i\phi_{ij;B} \tau_z} F_j)$ and $t_{ij;A}^f = t_A e^{i\phi_{ij;A} \tau_z}$. $n_i = f_i^\dagger f_i$ is constrained to be the physical density. The total filling is $n_A + n_B = 1$ so there are electron and

hole pockets in the two layers respectively. Due to the V term, we can decouple the following mean field:

$$H_M^f = -V \sum_{\langle ij \rangle} \chi_{ji} f_i^\dagger f_j \quad (4)$$

where $\chi_{ij} = \langle f_i^\dagger f_j \rangle$ is the exciton order parameter. The above term is basically a spontaneously generated hopping term on the nearest neighbor AB bond of the honeycomb lattice.

The obtained phase diagram is shown in Fig. 2(a), similar to the Hartree Fock results (see the supplementary). We find the following phases: (I) MI: layer polarized Mott insulator with 120° AFM in layer B; (II) FL: Fermi liquid with electron and hole pockets in A and B layer; (III) AF EI: excitonic insulator with AFM order in the B layer and τ_z FM order in the A layer. Chern number is $C = 0$; (IV) NEI: nematic excitonic insulator with τ_z FM in both layers. Chern number is $C = 0$; (V) ECI: excitonic Chern insulator with $|C| = 1$ and τ_z FM order in both layers. The transition between the trivial AF-EI and the topological ECI phase is first order with a small jump of the charge gap Δ_c .

Exciton order parameter and Chern number We allow various translational symmetry breaking patterns in the $\sqrt{3} \times \sqrt{3}$ unit cell and the exciton order parameter is in the form:

$$\chi_{ij} = \sum_{Q,L \in Z} \chi_{Q,L} e^{iQ\mathbf{K}_M \cdot \mathbf{r}_i} e^{iL\theta(\mathbf{r}_j - \mathbf{r}_i)} \quad (5)$$

where $i \in A$ and $j \in B$. $\chi_{Q,L}$ is the amplitude. $\theta(\mathbf{r}_j - \mathbf{r}_i)$ is the angle of $\vec{r}_j - \vec{r}_i$ relative to the $\hat{x}$ direction. Here Q and L are associated with the center of mass momentum and angular momentum of the exciton.

Both Q and L are not gauge-invariant. One can simply shift the momentum of each layer by a gauge transformation $f_{i;a} \rightarrow f_{i;a} e^{\pm i\mathbf{K}_M \cdot \mathbf{r}_i}$ with $a = A, B$ to shift Q and L . We define Q and L by going to the gauge which shifts the electron and hole pockets of the two layers to the Γ_M point of the MBZ⁵². In this gauge, the electron-hole problem can be mapped to the familiar Bardeen-Cooper-Schrieffer (BCS) theory of superconductor. Then we find there is only a $\mathbf{Q} = 0$ component, meaning that the exciton order can efficiently hybridize the electron and hole pockets at Γ_M point. The AF-EI has $L = 0$ or s wave exciton order. Its resulting band is trivial. The ECI phase is dominated by the $L = 1$ or $L = -1$ component and the exciton order is of $p \pm ip$ symmetry. This leads to a Chern band with $C = \pm 1$ and a QAH effect. In the NEI phase, the mixing between s ($L = 0$) and p ($L = \pm 1$) wave for the exciton order breaks the C_3 symmetry. The Chern number is zero. The exciton angular momentum is plotted in Fig. 2(b).

Let us also provide some intuitive explanations for why $p \pm ip$ exciton condensation is favored over the s wave. In the supplementary, we show that the competition between s wave and p wave exciton instability is decided by the magnitude of the form factors

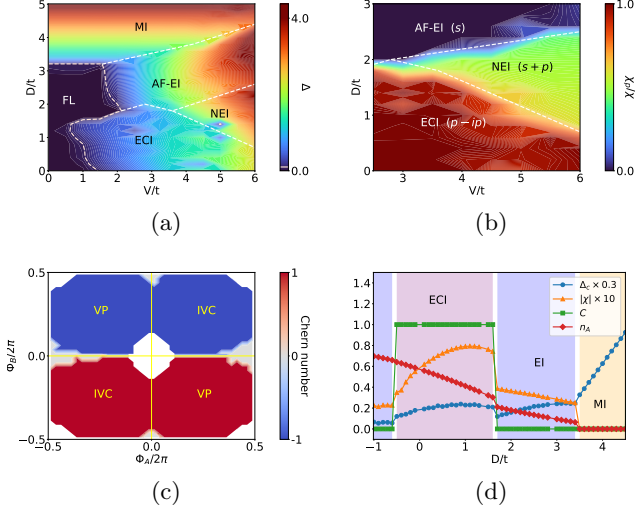


FIG. 2: (a) Schwinger boson mean field phase diagram in $D - V$ plane. The color bar shows the single electron charge gap Δ_c . Various phases are marked: Mott Insulator (MI), Fermi liquid (FL), trivial exciton insulator with nematicity (NEI), trivial exciton insulator with 120° order in B layer (AF-EI), excitonic Chern insulator with τ_z ferromagnetic order (ECI). In all of the three excitonic insulator phases, the A layer is valley polarized. (b) Evolution of exciton angular momentum. Colors show the weight of $p - ip$ (or $p + ip$) component. The ECI is dominated by $p \pm ip$ symmetry. NEI has a mixture of s and $p - ip$ waves so it breaks C_3 symmetry. (c) $\Phi_A - \Phi_B$ phase diagram for the Chern number. We used $D = 0.5, V = 4$. Metallic regions are whitened out. The model has time-reversal symmetry. We have restricted to the state with $\langle \tau_A^z \rangle > 0$. The corresponding valley-polarized (VP) and inter-valley-coherent (IVC) regions are marked with text. (d) Displacement field driven evolution along a line cut with $V = 2.5$. The transition from trivial to topological exciton insulating state is first order. Exciton order χ and charge gap Δ_c jumps on the phase boundary. The ECI phase onsets when n_A is above a critical density around 0.25. In the calculation, τ_A^z is fixed to be polarized to the $+$ valley. We use the following parameters: $U = 50, t_A = t_B = 1, \phi_A = \pi/4, \phi_B = \pi/12, V = 2V' = 4$. The results in (a) and (b) are calculated using Schwinger boson mean field theory, while (c) and (d) are obtained from Hartree-Fock mean field theory.

$F_l(\mathbf{k}) = \sum_{j=0,1,2} e^{i\frac{2\pi}{3}lj} e^{i\mathbf{k} \cdot \delta\mathbf{r}_j}$ near Fermi surfaces of the electron and hole pockets. Here $l = 0, 1, -1$ is the angular momentum of the exciton. $j = 0, 1, 2$ is summed over three nearest neighbor bonds and $\delta\mathbf{r}_j$ is the displacement vector of the nearest neighbor bond. When $n_A > 0.25$, we find that $|F_{l=\pm 1}(\mathbf{k})| > |F_{l=0}(\mathbf{k})|$ for $\mathbf{k}$ around the Fermi surfaces of the electron and hole pockets. This agrees with our phase diagram in Fig. 2(a) where the

ECI phase onsets after a critical exciton density. We also find that a small valley flux Φ_A or Φ_B is needed to stabilize the ECI phase. For the $\Phi_A = \Phi_B = 0$ point, the ECI phase shrinks to a quite small regime in (D, V) parameter space (see the supplementary).

Kinetic magnetism and symmetry selection The ECI phase in the above is fully valley polarized in both layers. Unlike the quantum Hall ferromagnetism, there is no Hund's coupling or band topology in our model. Instead, the ferromagnetism is kinetically driven: with the valley polarization, the hopping of the holon f_i is larger, favored by the kinetic energy. This is similar to the celebrated Nagaoka ferromagnetism⁵³ in a hole-doped Mott insulator.

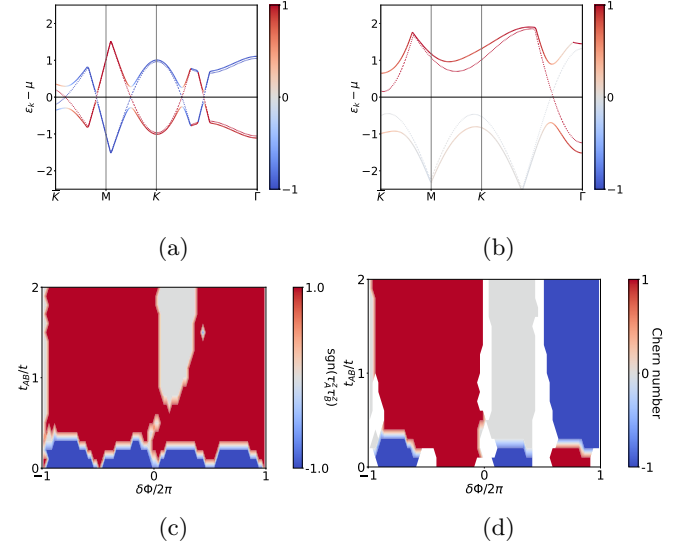


FIG. 3: Exciton gap opening and phase diagrams in $t_{AB} - \delta\Phi$ plane. (a), (b) Mean-field electron band structure in ECI and EI phases. Dotted and solid curves stand for band structures before and after turning on exciton order parameters. Colors show the valley polarization. (a) and (b) are obtained with $V = 1.5, D = 0.5$ and $V = 3, D = 3$ respectively. (c) The sign of $\tau_A^z \tau_B^z$ when it is nonzero. (d) Chern number. The gapless region is whitened out. In presence of t_{AB} , $\delta\Phi \rightarrow \delta\Phi + 2\pi$ is no longer a gauge transform. These phase diagrams are calculated using Hartree-Fock mean field for $t_A = t_B = 1, \Phi_A = 2\pi + \delta\Phi, \Phi_B = \delta\Phi, U = 50, V = 2V' = 4$ and $D = 0.5$.

The valley polarizations in two layers can have the same or opposite signs. At $\Phi_B = 0$ (or $\Phi_A = 0$), the valley SU(2) symmetry of the B (or A) layer guarantees the degeneracy of intra-valley and inter-valley exciton condensation. With a finite Φ_B , one of them is selected, while the other one is selected for an opposite sign of Φ_B because we can apply the transformation $i\tau_x$ only for layer B to flip Φ_B and τ_B^z together. This selection is shown in Fig. 2(c). For $\Phi_A \Phi_B > 0$, one finds that inter-valley exciton is favored and the QAH phase has

inter-valley-coherence(IVC) with $\tau_A^z \tau_B^z < 0$, consistent with the MCD measurement⁴⁰.

A finite t_{AB} can also lift the degeneracy and select the $\tau_A^z \tau_B^z > 0$ phase, as shown in Fig. 3(c). We need a finite but small valley flux Φ_A or Φ_B (as small as $\frac{\pi}{12}$) to stabilize the ECI phase. Deep inside the region with $\Phi_A \Phi_B > 0$, a sizable t_{AB} as large as 0.35 t is consistent with an ECI phase with opposite valley polarizations.

Lastly, our Schwinger boson formulation has the nice feature of spin-charge separation. In the supplementary, we show that increasing the superexchange J_B term can lead to an excitonic Chern insulator with canted spin texture and only partial valley polarization. This may be one explanation for the non-saturation of the MCD at a small magnetic field in the recent experiment⁴⁰. However, we note that partial valley polarization can also be trivially explained by domain wall formation driven by disorder effects.

Summary In conclusion we propose a new mechanism for the QAH effect in the Coulomb coupled moiré bilayer and apply it to the MoTe₂/WSe₂ system. In our model, the inter-layer hopping is spontaneously generated from inter-layer exciton condensation, which can have $p \pm ip$ symmetry and lead to an excitonic Chern

insulator (ECI). The ECI phase has valley ferromagnetism arising from kinetic energy, in contrast to the quantum Hall ferromagnetism picture for the QAH effect in graphene moiré systems^{28,31}. The valley polarizations in the two layers can be the same or opposite, depending on small perturbations away from a symmetric point where they are degenerate. Our theory opens a new direction to search for interaction-driven topological phases without single-particle band topology, for example in moiré bilayer with an hBN barrier in the middle. Excitonic insulators with $p \pm ip$ pairing symmetry may also be realized in other systems outside of the moiré materials.

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Note added When finalizing this manuscript, we become aware of a recent preprint⁵⁴ which also proposes inter-valley $p \pm ip$ exciton order for MoTe₂/WSe₂ system. But the mechanism to select the inter-valley exciton order over intra-valley exciton order in Ref. 54 relies on a large Hund's coupling and is different from our theory.

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