Excitonic density wave and spin-valley superfluid in bilayer transition metal dichalcogenide

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Recently, twisted bilayer WSe$_2$[1] emerged as a new robust moiré system hosting a correlated insulator at moiré half-filling over a range of twist angle and displacement field. In this work, we present a theory of this insulating state as an excitonic density wave due to intervalley electron-hole pairing. We show that exciton condensation is strongly enhanced by a van Hove singularity near the Fermi level. Our theory explains the remarkable sensitivity of the insulating state to the displacement field as well as the lack thereof to the Zeeman field in terms of pair-breaking versus non-pair-breaking perturbations. We further predict superfluid spin transport in this electrical insulator, which can be detected by optical spin injection and spatial-temporal imaging.

Introduction – Moiré superlattices based on transition metal dichalcogenides[1–6] have recently emerged as a new host of strongly correlated phases of matter. In a very recent experiment on WSe$_2$ homobilayers[1] with twist angles $\theta \in [4^\circ, 5.1^\circ]$, an insulating dome was found at low temperature within a narrow range of displacement field, when the topmost valence miniband is half filled. The activation energy gap is $\sim 3$meV, which is remarkably large among moiré systems[7–12]. However, the insulating gap is smaller by an order of magnitude than either the miniband width (about $60 – 100$meV for $\theta = 4^\circ \sim 5^\circ$) or the characteristic Coulomb interaction energy $e^2/\epsilon L$ ($\sim 30$meV for a dielectric constant $\epsilon = 10$ at $4^\circ$). The smallness of insulating gap compared to the interaction energy and bandwidth and its sensitivity to the displacement field speak against the scenario of a Mott insulator and call for a new theoretical understanding.

In this Letter, we predict that the insulator in twisted TMD at half filling is an excitonic spin density wave formed by the pairing of electrons and holes in minibands at different valleys. This electron-hole pairing is strongly enhanced by a van Hove singularity[13–20] (VHS) near the Fermi level. We show that the detuning of the displacement field has a pair breaking effect on the excitonic insulator, while the Zeeman coupling to an out-of-plane magnetic field is non-pair-breaking at leading order. We introduce a low-energy theory for twisted TMD and calculate the phase diagram as a function of displacement field, temperature and magnetic field, finding good agreement with the experiment [1]. Interestingly, due to spin-valley locking in TMD [21, 22], our excitonic insulator is also a spin superfluid and thus enables coherent spin transportation over long distances. We propose an all-optical setup for spin injection and spatial-temporal imaging of spin transport.

At small twist angle, the two sets of moiré bands in bilayer TMD originating from $K$ and $K'$ valleys are decoupled at the single-particle level and treated separately hereafter. It is important to distinguish two different stacking configurations denoted as AA and AB, which differ by a $180^\circ$ rotation of the top layer. The two have very different moiré band structures due to spin-valley locking in TMD. In AA stacking, the $K$ valleys on two layers with the same spin polarization are nearly aligned, so that interlayer tunneling is allowed and creates layer-hybridized moiré bands, as shown in Fig. 1 (a). In contrast, in AB stacking, the $K$ valley on one layer is nearly aligned with the $K'$ valley on the other layer with the opposite spin polarization, so that interlayer tunneling is forbidden and the resulting moiré bands have additional spin/layer degeneracy [2, 23]. Throughout this work we consider AA stacking, where the full filling for the topmost moiré bands corresponds to 2 electrons per supercell.

At low carrier density, the dominant interaction is Coulomb repulsion within the same valley and between two valleys: $H_{int} = \sum_{i,j} \int d\mathbf{r} d\mathbf{r}' V_{ij}(\mathbf{r} - \mathbf{r}') n_{i,r} n_{j,r'}$, where $i, j = \pm$ is valley index and $n_{i,r} = c_{i,r}^\dagger c_{i,r}$ is density operator of a given valley. The interacting Hamiltonian is reminiscent of bilayer quantum Hall system[24], where the layer degree of freedom plays the role of valley. At half-filling of each layer, Coulomb repulsion leads to interlayer exciton condensation. However, unlike Landau levels, the moiré band in twisted bilayer TMDs considered here has a sizable bandwidth comparable to or larger than the interaction energy, which is far from the flat band limit.

We calculate the moiré band structures using the continuum model developed in Ref.[2] with parameters extracted from first principle calculations[39]. Interestingly, we find that the moiré band dispersion is highly tunable by the displacement field, $D$. For a certain range of displacement field, our band structure calculation (Fig. 1 (b), (c)) shows that the Fermi level at half filling is close to a van Hove singularity (VHS) in density of states, resulting from saddle points near the corners of the mini-Brillouin zone, $\mathbf{K}_M$ and $\mathbf{K}'_M$. The proximity to VHS is supported by the observed sign change of Hall coefficient near half-filling [1].

Since a diverging DOS near the Fermi level enhances correlation effects, the detuning of VHS by the displacement field is expected to affect the metal-insulator tran-
FIG. 1: (a) The moiré band structure for one valley of twisted bilayer WS2. At a critical displacement field $D_s$, the $K_M$ becomes a saddle point of the dispersion. (b) The density of states (with various $D$ near $D_s$) at twist angle $\theta = 4^\circ$. (c) Evolution of energy contours as function of the displacement field (the dark line corresponds to half-filling). At $D_s$, the system hosts a higher order van Hove singularity at $K_M$.

sition. Motivated by this consideration, we now develop a low-energy theory by expanding the moiré band from valley $K$ ($K'$) around $K_M$ ($K_M'$). Based on the lattice symmetries, the Taylor expansion up to third order in momentum takes the general form

$$\varepsilon_{\pm}(k) = \alpha k^2 \pm \xi(k), \quad \xi(k) = k_y^3 - 3k_yk_z^2$$

where $\pm$ is the valley index related by time reversal symmetry $\mathcal{T}: c_{k,+} \rightarrow c_{-k,-}, c_{k,-} \rightarrow c_{-k,+}$.

The coefficient $\alpha$ depends on the displacement field $D$. It is useful to first consider a critical displacement field $D_s$ where $\alpha = 0$. Then, $k = 0$ becomes a monkey saddle point where three energy contour lines interact [18, 25], resulting in a high order van Hove singularities (hVHS) with a power law divergent density of states [16–18]

$$\rho_{\alpha=0}(E) \sim 1/|E|^{1/3},$$

as shown in Fig. 1(c).

At $\alpha = 0$, the low-energy dispersion has a perfect nesting condition: $\varepsilon_{+}(k) = -\varepsilon_{-}(k)$, so that within the topmost moiré band occupied states of one valley map onto unoccupied states of the other valley under a shift of momentum by a nesting wavevector $Q = K_M - K_M'$. As a result of perfect nesting, Coulomb repulsion $V$ immediately leads to an intervalley exciton condensate with an order parameter

$$\Delta \sim -\sum_k c_{k,+}^\dagger c_{k,-}.$$

The ordered state is fully gapped and electrically insulating. It spontaneously breaks the spin-valley $U(1)_s$ symmetry, time reversal symmetry, and translational symmetry. Therefore, our excitonic insulator is both an electron-hole superfluid at finite commensurate momentum and a spin density wave.

We can draw an analogy between the intervalley exciton condensate and the BCS superconductivity. After a particle-hole transformation on one of the valleys, the intervalley exciton order parameter becomes precisely an s-wave superconductivity with the valleys playing the roles of spins. Despite the similarities with the BCS problem, the proximity of hVHS introduces interesting new features for the exciton condensate.

Consider the case with $\alpha = 0$. In the weak coupling regime, we can analytically solve the gap equation as follows,

$$\frac{1}{V} \approx \int_{-\infty}^{\infty} dE \rho(E) \frac{1}{2\sqrt{E^2 + \Delta^2}} \sim \int_{0}^{\infty} dE \frac{1}{|E|^2} \frac{1}{2\sqrt{E^2 + \Delta^2}} \sim 1/\Delta^{1/3}$$

where we have used the power law density of state near the hVHS. We find the intervalley exciton order parameter scales as $\Delta \sim V^3$. For weak coupling, the critical temperature $T_c$ is proportional to the gap, therefore $T_c \sim V^3$. This power-law scaling of the critical temperature is distinctly different from the BCS formula. We also numerically compute the zero temperature order parameter $\Delta_0$ and find $\Delta_0/T_c \approx 1.88$, larger than the standard BCS value.

Next we consider the strong coupling regime where the electron and hole form tightly bound pairs, analogous to the BEC limit of Fermi gas. Denoting the UV cutoff in reciprocal space $\Lambda$ and the corresponding bandwidth $2\Lambda^3 = W$, we solve the gap equation for interaction strength $V \sim O(W)$ and find the order parameter scales as $\Delta \sim V$. However, instead of being determined by the pairing gap, the transition temperature is set by the BEC temperature of the exciton gas, which scales as $T_c \sim W^2/V$, proportional to the inverse mass of the excitons. Viewed from real space, our system in the strong-coupling regime is a Mott insulator with a charge gap $\sim V$ and an antiferromagnetic exchange interaction $\sim W^2/V$ (see Fig. 2(a)). An interesting feature

| Order Parameter | Superconductivity | Intervalley Exciton |
|-----------------|------------------|---------------------|
| $c_{s,c,c}^\dagger$ | $c_{s,c,c}^\dagger$ | $c_{s,c,c}^\dagger$ |
| Broken Symmetry | $U(1)$ charge | $U(1)$ $S_\pm$ valley |
| Pair-breaking | $B$ | $\mu$ & $\alpha$ |
| Non-pair-breaking | $\mu$ | $B_\perp$ |
| Density | Superfluid density | $S_{\pm}$ polarization |

TABLE I: The correspondence between intervalley exciton condensation and the BCS superconductivity.
is that the crossover scale between weak and strong coupling behaviors is relatively small in this model due to the presence of the hVHS.

Perturbations – The energy of van Hove singularities relative to the Fermi level at half-filling $\mu$ and the quadratic term in energy dispersion $\alpha$ can be viewed as perturbations to the ideal limit of perfect nesting. Experimentally the displacement field $D$ tunes both $\alpha$ and $\mu$. Both perturbations have pair-breaking effects on exciton condensation as they lift the degeneracy between electrons in one valley and holes in the other. In particular, under the aforementioned particle-hole transformation, $\mu$ precisely maps to the Zeeman field in a superconductor. We also consider an out-of-plane magnetic field that splits the spin/valley degeneracy by Zeeman energy [26],

$$H_B = \pm B_{\perp} \int_k c_{k,\pm}^\dagger c_{k,\pm}.$$  

In contrast to $\alpha$ and $\mu$, the out-of-plane magnetic field $B_{\perp}$ maps to the chemical potential in a superconductor and its effect is non-pair-breaking. The correspondence between exciton condensate and superconductor is summarized in Tab. I.

In the following, we consider a mean field theory for the intervalley exciton condensate that includes these perturbations. The mean field hamiltonian reads

$$H_{MF} = \sum_{k,\nu=\pm} (\varepsilon_\nu(k)+\nu B_{\perp}-\mu)c_{\nu k,\pm}^\dagger c_{\nu k,\pm} + \Delta c_{\downarrow,\pm}^\dagger c_{\uparrow,\pm} + h.c. + \frac{\Delta^2}{V},$$  

where $\Delta$ is the order parameter for the exciton condensate and $V$ is the effective interaction strength in the $s$-wave channel. The quasi-particle spectrum is given by

$$E_{\pm}(k) = \alpha k^2 - \mu \pm \sqrt{(\xi(k)+B_{\perp})^2+|\Delta|^2}. $$

The quasiparticle gap between the two bands is given by $\Delta_g = 2\Delta - \alpha \Delta^2$, which is an indirect gap for $\alpha \neq 0$.

We calculate the mean field free energy and vary it with respect to $\Delta$ to get the following gap equation,

$$\frac{1}{V} = \sum_k \frac{1}{2\sqrt{(\xi(k)+B_{\perp})^2+|\Delta|^2}} (n_F(E_-(k))-n_F(E_+(k))),$$  

where $n_F(E)$ is the Fermi-Dirac distribution. Fig. 2 (b)-(d) show the quasiparticle gap $\Delta_g$ as a function of $\mu$, $T$ and $B_{\perp}$ for various $\alpha$ at weak coupling $V = 0.1W$. See supplementary materials for results with $V = 0.5W$. Notice the gap equation is invariant under $\alpha \rightarrow -\alpha$, $\mu \rightarrow -\mu$. Thus, we only plot for $\alpha > 0$.

First, Fig. 2(b) shows the quasiparticle gap as a function of $\mu$ at $T = 0$ and $B_{\perp} = 0$. For a narrow range of $\mu$, the system develops the intervalley exciton order which gives rise to a full gap. With the realistic bandwidth $W = 100$meV, the maximal mean field quasiparticle gap is $\Delta_g \approx 3.5$meV, which is close to the activation gap fitted from transport experiments [1].

Next, we plot the quasiparticle gap as a function of temperature $T$ at half-filling in Fig. 2(c). The finite temperature metal-insulator phase transitions are continuous. For $W = 100$meV, the maximal critical temperature is $T_c \approx 10K$, which is consistent with the onset temperature of insulating behavior [1].

Remarkably, the excitonic insulator only exists when the van Hove singularity is tuned close to the Fermi level by the displacement field. For $V = 0.1W$, a small detuning in $\mu$ of about 0.01$W$ is sufficient to drive the excitonic insulator to a normal metal through a first-order phase transition, which precisely corresponds to the pair-breaking transition of an $s$-wave superconductor driven by the Zeeman field.

Finally, we plot the gap as function of the out-of-plane magnetic field $B_{\perp}$ at $T = 0$ and $\mu = 0$ shown in Fig.
2(d). Notice the critical field for small $\alpha$ (i.e., good nesting) is much larger than the critical chemical potential in Fig. 2(b). In addition, for all $\alpha$, the quasiparticle potential decreases slowly with $B_\perp$ initially, which indicates the effect of out-of-plane magnetic field is non-pair-breaking to the leading order. For larger $B_\perp$, the quasiparticle gap is reduced in an approximately linear way. For a nearly high-order van Hove singularity (i.e., small $\alpha$), the upper critical magnetic field is significantly larger than the critical displacement field, when measured in terms of the Zeeman energy and detuning energy respectively.

In summary, our theory based on a van Hove singularity near the Fermi level gives insulating gap and critical temperature comparable to the experimental values, and explains the remarkable sensitivity of the gap to the displacement field as well as the lack thereof to the Zeeman field in terms of pair-breaking versus non-pair breaking perturbation to exciton condensation. In the following, we propose an experiment to directly probe the macroscopic intervalley coherence in the insulating state.

**Optical spin injection and spin superflow** – In our theory, the half-filling insulator in TMD homobilayer spontaneously breaks the spin/valley $S_z$ conservation in a similar way that a superconductor spontaneously breaks charge conservation. Therefore it can be regarded as a spin superfluid. The possibility of spin supercurrent has been theoretically predicted in magnetic insulators with easy-plane anistropy [27–31]. Its signature has been reported in recent electrical measurements on quantum Hall state in graphene and antiferromagnetic insulator Cr$_2$O$_3$ [32–34], where spin Hall effect is used to generate and detect a non-equilibrium spin accumulation.

A key advantage of 2D TMDs is that the spin polarization can be easily generated and detected by purely optical means. It has been shown that a circularly polarized light can efficiently generate spin polarizations in TMDs due to the spin-valley locking and valley-selective coupling to chiral photons [35–37]. The local spin polarization can be read out by measuring the difference in reflectance of right- and left-circularly polarized lights, i.e., the circular dichroism spectroscopy [5, 38]. Therefore, TMDs provide an ideal platform for studying spin transport with a fully optical setup.

We propose the following experiments (see Fig. 3(a)) to detect spin superfluidity in the insulating state of bilayer TMDs. We first create a local spin polarization by circularly polarized light, and then use the spatial-temporal resolved circular dichroism spectroscopy [5, 38] to monitor its propagation as a function of time. In the spin superfluid state, the local spin polarization will propagate ballistically and coherently via collective modes (in the absence of dissipation, see below) [27]. Such ballistic spin transport is a key feature of our intervalley excitonic insulator. In contrast, spins should have diffusive dynamics [27] if the half-filling insulator is valley polarized.

In a spin superfluid, the spin transport equation involves the superfluid phase $\varphi$ that specifies the angle of the in-plane magnetic order parameter [27],

$$\frac{dM_z}{dt} = -\nabla \cdot J_z - \frac{M_z}{\tau},$$

$$\frac{d\varphi}{dt} = -\frac{1}{K}M_z + \ldots,$$

where $M_z$ is the magnetization along $z$-direction and a conjugate variable to $\varphi$. The spin current $J_z$ is given by $J_z = \rho \nabla \varphi$, where $\rho$ is the superfluid stiffness. The parameters $\rho$ and $K$ can be obtained by considering the effective low energy of the Goldstone mode; see supplementary materials. $\tau$ is the spin relaxation time, which is expected to be long in TMDs since a spin-flip requires intervalley scattering due to spin-valley locking. In a spin superfluid, the spin-valley wavepacket shows ballistic propagation at the spin wave velocity $v = \sqrt{\rho/K}$. In contrast, in the absence of spin superfluidity, the spin dynamics is diffusive. The two cases yield completely different transport behaviors as shown in Fig. 3(b).

We also note that recent experiments found a highly insulating state at half filling in WSe$_2$/WS$_2$ heterobilayer [5]. A likely candidate for its magnetic order is the 120° antiferromagnetism on the triangular lattice. This state has the same symmetry breaking as our excitonic insula-
tor in twisted WSe₂ bilayer. Thus our prediction of spin superfluid transport may also apply to WSe₂/WSe₂.

Acknowledgement – We thank Abhay Pasupathy, Augusto Ghiotto and Cory Dean for sharing their experimental results prior to publication, and Yang Zhang for collaboration on related work. We thank Ran Cheng, Feng Wang, Noah F. Q. Yuan, Yi-Zhuang You and Shu Zhang for useful discussion. This work is supported by DOE Office of Basic Energy Sciences under Award DESC0018945. ZB is supported by the Pappalardo fellowship at MIT and partially by KITP program on topological quantum matter under Grant No. NSF PHY-1748958. LF is partly supported by Simons Investigator Award from the Simons Foundation.
in the mean field phase diagram. For small $\alpha$ to a normal metal through an intermediate phase separated half-filling state. In the weak coupling case, as we introduce finite $\alpha$, the phase diagram has a strong particle-hole asymmetry in place. Further increasing $\alpha$ can drive the system from an excitonic insulator to a normal metal through an intermediate exciton metal phase.

**SUPPLEMENTARY MATERIALS**

**Mean field phase diagram**

First let us derive the mean field gap equation. We can obtain the mean field hamiltonian Eq. 6 by Hubbard-Stratonovich transformation of the interacting hamiltonian. Setting the variation to zero, we get precisely the gap equation shown below.

$$F_{MF} = \sum_{k,\nu=\pm} -\frac{1}{\beta} \log(1 + e^{-\beta E_\nu(k)}) + \frac{\Delta^2}{V}, \quad (11)$$

where $E_\pm$ are given in Eq. 7. We can take $\Delta$ to be real and vary the free energy with respect to it,

$$\frac{\partial F_{MF}}{\partial \Delta} = \frac{2\Delta}{V} + \sum_{k,\nu=\pm} \frac{\Delta}{(\xi_k + B)^2 + \Delta^2} \frac{\nu}{1 + e^{\beta E_\nu}}. \quad (12)$$

Setting the variation to zero, we get precisely the gap equation in Eq. 8.

Now we plot the zero temperature mean field phase diagram as function of $\mu$ and $\alpha$ for $V = 0.1W$ and $V = 0.5W$ in Fig. 4. There are in general three phases in the mean field phase diagram. For small $\alpha$ and $\mu$, we get the exciton insulator, which has non-zero exciton condensate and a fully gapped spectrum. For larger $\alpha$, the system develops fermi surfaces while maintaining the exciton order, which we denote as the exciton metal phase. Further increasing $\alpha$ or $\mu$ destroys the exciton order and leaves the system a normal metal. Notice the phase diagram has a strong particle-hole asymmetry induced by finite $\alpha$. We can focus our attention to the half-filling state. In the weak coupling case, as we increase $\alpha$, the system will go from an excitonic insulator to a normal metal through an intermediate phase separation regime. In the strong coupling limit, raising $\alpha$ can drive the system from an excitonic insulator to a normal metal through an intermediate exciton metal phase.

**Low energy effective theory**

We derive the effective theory for the phase fluctuation of the exciton order parameter in the weak coupling limit. The effective theory will give us the velocity of the spin wave excitations. After a Hubbard-Stratonovich transformation, the interacting fermion theory can be brought into the following form (in imaginary time),

$$\mathcal{L} = \sum_{\nu=\pm} \psi_\nu^\dagger (-i\omega + H_{k,\nu}) \psi_\nu + \Delta \psi_\nu^\dagger \psi_\nu + h.c. + |\Delta|^2/V. \quad (13)$$

Let us assume the exciton order is fixed at $\Delta = |\Delta|e^{i\varphi}$ with $\varphi = 0$. The fermion green’s function is given by

$$G_\psi = (-i\omega + H_k + |\Delta|^2)^{-1}. \quad (14)$$

where $\tau$’s are the pauli matrices in valley space. Consider the effective action for phase fluctuations. The self energy for $\varphi$ is given by the following bubble diagram,
approximation, the green’s function for \( \varphi \) is given by
\[
G_b(i\Omega, p) = \frac{V/|\Delta|^2}{1 + V/|\Delta|^2 \Pi(i\Omega, p)} = -\frac{1}{c_1|\Delta|^{-1/3}\Omega^2 + c_2|\Delta|p^2}.
\] (17)

Therefore the effective action for \( \varphi \) is
\[
\mathcal{L}_{\text{eff}} = \int_{x,\tau} K(\partial_\tau \varphi)^2 + \rho(\partial_x \varphi)^2,
\] (18)

where \( K = c_1|\Delta|^{-1/3} \) and \( \rho = c_2|\Delta| \). The superfluid velocity is given by \( v = \sqrt{\rho/K} = \sqrt{c_2/c_1}|\Delta|^{2/3} \). The case of \( \alpha \neq 0 \) is more involved and we leave it to future work. However, the form of effective action in Eq. 18 is generally applicable in any superfluid state.