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Gate-tunable heavy fermion quantum criticality in a moiré Kondo lattice

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We propose a realization of Kondo-lattice physics in moiré superlattices at the interface between a WX_2 homobilayer and MoX_2 monolayer (X=S,Se). Under appropriate gating conditions, the interface-WX_2-layer forms a triangular lattice of local moments that couple to itinerant electrons in the other WX_2 layer via a gate-tunable Kondo exchange interaction. Using a parton mean field approach we identify a range of twist angles which support a gate-tuned quantum phase transition between a heavy Fermi liquid with large anomalous Hall conductance and a chiral spin liquid coexisting with a light Fermi liquid and describe experimental signatures to distinguish among competing theoretical scenarios.

Crystals containing lanthanide or actinide elements host f-electron local spin moments coupled via Kondo spin-exchange interactions to itinerant conduction (c) electrons. The phase diagrams of such Kondo lattice systems are often extremely rich, and can include magnetic and non-magnetic states, superconductors, and non-Fermi liquids [1–5]. When Kondo screening effects dominate, the f-spins hybridize with c-electrons and contribute to the Luttinger-volume of the Fermi-sea producing exceptionally high (heavy fermion) quasiparticle masses [1–3] arising from the narrow f bandwidth. Alternatively, dominant spin-spin interactions (chiefly c-mediated by RKKY coupling) cause f local moments to order into either i) a magnetic state [6], which is typically an antiferromagnet (AFM) or ii) a quantum spin liquid (QSL) [5, 7] if magnetic order is frustrated.

These Kondo-(un)screened phases are separated by a quantum critical point marked by a fan of non-Fermi liquid transport signatures [2–4, 8] that is often partly occluded by an unconventional superconducting dome. The hallmark of this apparently-continuous critical point is a sudden change in the size of the Fermi surface. Understanding the nature of such Fermi-volume-changing transitions and their relation to non-Fermi liquid behavior is thought to be key to understanding many other strongly correlated systems such as high-temperature cuprate superconductors, and remains a key unsolved problem in condensed-matter physics. Moreover, different experiments and theoretical analyses produce a conflicting picture of the nature of the non-hybridized phase, and its nature remains hotly debated despite decades of intensive study. The complex microscopic structure, many-band character of f-electron materials along with the limited ability to tune carrier-densities and exchanging couplings in bulk $3d$ crystals pose significant obstacles to resolving these mysteries.

In this Letter we propose that progress can be achieved by using recently discovered transition metal dichalcogenide (TMD) moiré material platforms to construct a synthetic Kondo lattice made from relatively simple well-understood components. Crucially, we will show that the Kondo-exchange coupling can be continuously tuned by electrostatic gate tuning, enabling in-situ access to the entire heavy-fermion phase diagram and criticality in a single device. Furthermore, these systems have triangular lattice symmetries that tend to frustrate magnetic order, and can favor fractionalized QSL states. Our main results are summarized in Fig. 1 which contains phase diagrams in the two-dimensional gate-voltage phase space of dual-gated two-dimensional materials. We first identify the twist-angle dependent area within which a Kondo lattice model is realized and then estimate the location of the Kondo-screening quantum critical line. Parton mean-field calculations predict a novel quantum criticality scenario in which local moments in a weak-coupling chiral spin liquid (CSL) state hybridize with conduction electrons to form a strong-coupling heavy Fermi liquid that exhibits an anomalous Hall effect. Previous works on the triangular Kondo lattice model [9, 10] support the existence of such anomalous Hall states due to nonzero spin chirality. The parton mean-field approach used here should be viewed as a way to obtain a regime where quantum criticality is likely to occur. We discuss experimental probes of Kondo lattice physics in moiré materials that discriminate among various competing antiferromagnetic (AFM) and quantum spin-liquid (QSL) weak Kondo-coupling scenarios, including the ones not favored by the parton mean-field theory, highlighting electrical and thermal Hall transport, and electrostatic measurements of entropy. Compared to previous proposals for achieving heavy fermion physics in moiré systems, our setup directly implements a Kondo lattice model using already-demonstrated moiré ingredients; it avoids both topological obstacles [11–14] to local-moment physics of proposed twisted graphene multilayer realizations [15], as well as the need for unusual spontaneous orbital selectivity required to form local moments in electron-doped TMD heterobilayers [16].

A synthetic Kondo-Lattice – We propose a realization of Kondo lattice physics in TMD trilayers in which a triangular lattice moiré pattern is formed between an atomically aligned WX_2 bilayer twisted by a small angle θ relative to an MoX_2 monolayer, where X=S,Se is a

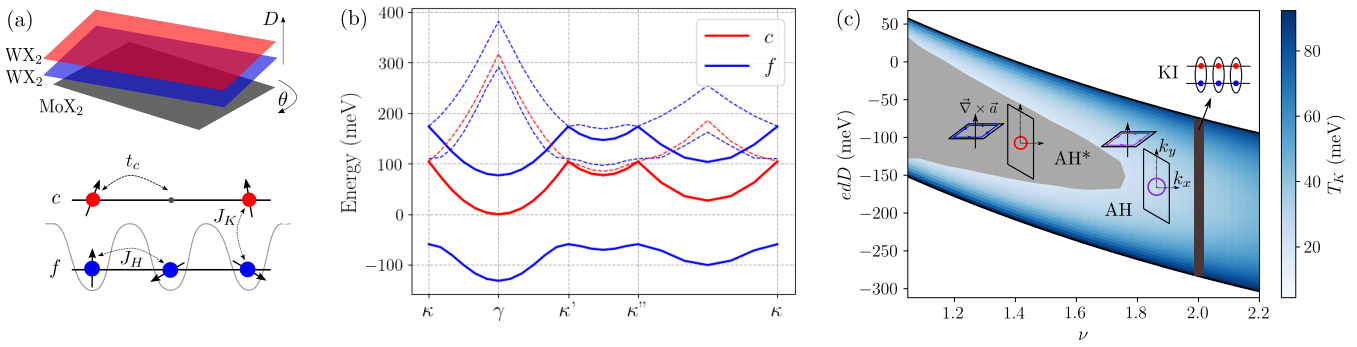

FIG. 1. **Kondo Lattice Simulator:** (a) Schematic of a TMD trilayer Kondo lattice system. The two active TMD layers experience moiré modulation potentials with different strengths so that one layer has flat bands and one has dispersive bands. When the density in the flat band layer is tuned to one electron per moiré period, and the total density to a higher value, a triangular lattice Kondo model is realized. (b) Gated self-consistent Hartree-Fock band structures in the middle of the Kondo lattice model regime at $\theta = 4.5°$, with filling factors $\nu_f = 1$ and $\nu_c = 0$. The solid blue bands are the upper and lower Hubbard bands in the f-layer. The dashed blue bands are the second lowest energy f-band in each spin. The red bands are the first (solid) and second (dashed) lowest energy (spin-degenerate) bands in the c-layer with bandwidth 104.49meV. (c) Parton mean-field phase diagram as a function of the moiré band filling factor ν and the external displacement field D, which are linearly related to the top and bottom gate voltages. The black lines bound the Kondo lattice regime within-which the mean-field filling factor is 1 in the f-layer and $\nu_c = \nu - 1$ in the c-layer. In the AH* phase, a Fermi surface of c-electrons (red) is decoupled from a chiral spin liquid (CSL) of spinons f. In the AH phase the Fermi surface (purple) has mixed c and f character. The Kondo insulator (KI) phase at $\nu = 2$ is adiabatically connected to direct product of singlets between c and f electrons on each site. The color scale indicates the Kondo temperature of the AH states.

chalcogen. We note that one could alternatively form a moiré potential by choosing different chalcogens for the WX_2 and MX_2 layers to force a lattice mismatch (though we do not model that case here). The active valence-band degrees of freedom reside in the WX_2 layers and come from momenta near the K and K' points [17] in the triangular-lattice Brillouin zone, where strong spin-orbit coupling locks the spin and valley degrees of freedom into a single effective spin-1/2 moment (with approximate SU(2)-symmetric interactions [18, 19]) that we will refer to simply as spin. Because the bottom WX_2 layer (closest to the MoX_2) experiences a stronger moiré modulation potential [19], its triangular-lattice symmetric moiré minibands will be narrower, enhancing correlations in this layer. Temporarily neglecting interlayer tunneling, the goal will be to half-fill the lowest moiré miniband of the lower (f) layer to form a Mott insulator of well-formed local moments, and partially fill the upper layer (c) to form a conducting Fermi surface. Mott insulators in related TMD bilayers have been achieved in several recent experiments [20–30], indicating the feasibility of this setup. Next, one must adjust the top-gate electric field E_t so that hole chemical potential, μ_c, lies inside the charge gap of the f-layer, $0 < \mu_c < U$ where $\mu_c = \partial\epsilon_c/\partial n_c$ is the chemical potential and ϵ_c is the energy per area of the c-layer, U is the charge gap of the f-layer, and we measure energy relative to the bottom of the f-layer gap. If correlations are neglected in the c-layer

$$\mu_c = \left(\frac{2\pi e^2}{\epsilon A_M}(\nu_c - 1) + eD\right) d + \epsilon_F(\nu_c) + U, \qquad (1)$$

where the displacement field $D \equiv (E_t + E_b)/2$, ϵ is the background dielectric constant, $E_{(t,b)}$ are the gate electric fields above the c-layer and below the f-layer respectively, d is the separation between WX_2 layers, ν_c is the number of holes per moiré period in the top layer, and $\epsilon_F(\nu_c)$ is the c-layer Fermi energy. Since the total carrier density is related only to the difference in gate fields via the Poisson equation: $4\pi e(1 + \nu_c)/(\epsilon A_M) = E_t - E_b$, ν_c and D can be controlled separately. For a given value of ν_c, μ_c increases monotonically with D and passes through the $(0, U)$ Kondo-lattice interval. As illustrated in Fig. 1(c), $\mu_c(D)$ increases with ν_c at fixed D and both boundaries of the Kondo lattice interval move to smaller D.

Kondo Lattice Model – Since the on-site repulsion in the f-layer, U, greatly exceeds the f-layer hopping and f/c interlayer tunneling amplitudes, within the Kondo lattice regime highlighted in Fig. 1c, we model the system by a Kondo-lattice Hamiltonian that retains only spin degrees of freedom in the f-layer:

$$H_{\rm KH} = -t_c \sum_{\langle \boldsymbol{r}\boldsymbol{r}'\rangle} c^\dagger_{\boldsymbol{r}\alpha} c_{\boldsymbol{r}'\alpha} + \frac{J_K}{2} \sum_{\boldsymbol{r}} \boldsymbol{S}_{\boldsymbol{r}} \cdot c^\dagger_{\boldsymbol{r}\alpha} \boldsymbol{\sigma}_{\alpha\beta} c_{\boldsymbol{r}\beta} - \mu_c \sum_{\boldsymbol{r}} c^\dagger_{\boldsymbol{r}\alpha} c_{\boldsymbol{r}\alpha} + J_H \sum_{\langle \boldsymbol{r}\boldsymbol{r}'\rangle} \boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}'}. \qquad (2)$$

Here $\boldsymbol{S}_{\boldsymbol{r}}$ is a spin operator at a triangular lattice site $\boldsymbol{r}$ in the f-layer, $c_{\boldsymbol{r}\alpha}$ annihilates a c-layer electron of spin α at site $\boldsymbol{r}$ and the repeated spin labels are implicitly summed.

We extract the conduction band hopping (t_c) and

Kondo coupling (J_K) parameters from self-consistent Hartree-Fock (SCHF) calculations in which the small hybridization, Γ, between the WX_2 layers is temporarily neglected (see Supplemental Material [31] for details). We later reintroduce Γ perturbatively to compute spin-exchange couplings.

Inside the Kondo lattice regime the solutions of the SCHF equations are characterized by a mean-field state that has a small itinerant electron Fermi surface, a fully occupied triangular-lattice [19–22, 32] majority-spin local moment band, and an empty minority-spin local moment band. SCHF calculations are performed using a continuum moiré model for holes with moiré modulation potentials in the two WX_2 layers that attract holes to lattice sites and are represented by a Fourier expansion $\sum_{\boldsymbol{b}} V_m(\boldsymbol{b})\exp(i\boldsymbol{b}\cdot\boldsymbol{r})$ involving only the first shell of reciprocal lattice vectors $\boldsymbol{b}$ [19]. We use a moiré potential strength $V_m = 30$ meV for the f-layer, based on recent experimental estimates [33], and a WSe_2 effective mass $m = 0.35m_e$ [19], where m_e is the mass of an electron. We see in Fig.1(b) that the c-layer and the minority spin f-layer mean-field bands are nearly-free-electron-like, with very small band-gaps to higher mini-bands at the zone boundary. This arises due to screening effects that result in approximate cancellation between attraction by the moiré potential and repulsion from the f-layer holes. Despite the small energetic separation between the first and second c-layer mini-bands, in the Supplemental Material [31] we show that tunneling between the f-orbital and the second c-band is suppressed, justifying the use of a single-band model for both layers.

The mean-field Hartree potential at each total moiré band filling factor depends on the gate-controlled external displacement field D. The phase boundaries in Fig. 1(c), calculated for the twist angle $\theta = 4.5^\circ$ case, were constructed by identifying the area in the gate-voltage phase space over which the top layer Fermi level lies in the bottom layer charge gap. Data for other twist angles is summarized in the Supplemental Material [31].

To estimate the Kondo coupling constant, we perturbatively reintroduce the weak interlayer hybridization, which we take to be momentum-independent (see Supplemental Material [31]), via a Schrieffer-Wolff transformation to obtain:

$$J_K = 2\Gamma^2\left(\frac{1}{U + E_F^f - E_F^c} - \frac{1}{E_F^f - E_F^c}\right), \qquad (3)$$

where $E_F^{f/c}$ are the respective Fermi levels. Crucially, we see that J_K can be enhanced with a displacement field that brings the c Fermi level closer to resonance with the upper or lower Hubbard band of the f-layer. The estimated Γ is much weaker than the Hubbard gap – for $\theta = 4.5^\circ$, $\Gamma \approx 21.6$ meV and $U \approx 209.06$ meV. As long as the applied displacement field is sufficiently away from the edges of the Kondo lattice regime shown in Fig. 1(c), where the above perturbative formula is valid, the system is well described a Kondo lattice, and a crossover to a mixed-valence regime is expected closer to the edges. Below, we will show that, for suitable twist angles, this allows gate-tuning across a Kondo-screening quantum critical point (KS-QCP).

The Heisenberg exchange coupling, J_H between the f-spins arises both through super-exchange $\sim 4t_f^2/U$ between the f-layer spins, and via RKKY interactions mediated by the conduction electrons [34]: $J_{\rm RKKY}(\boldsymbol{r}) = (\nu_c/\sqrt{3}t_c)J_K^2\left[\mathcal{J}_0(k_F r)\mathcal{Y}_0(k_F r) + \mathcal{J}_1(k_F r)\mathcal{Y}_1(k_F r)\right]$ where k_F is the conduction electron Fermi momentum, $\mathcal{J}_n, \mathcal{Y}_n$ are respectively Bessel functions of the first and second kinds[35].

Gate-tuned Kondo physics – The moiré Kondo lattice system has two especially attractive features: i) it is possible to tune the system between the strong J_K heavy fermion regime and the more complex weak J_K regime electrostatically by moving through the phase diagram with gates, and ii) the intrinsic triangular lattice geometry frustrates magnetic order and favors the more interesting fractionalized states that are thought to be a strong possibility in the weak J_K regime. To identify where these states are most likely to occur we employ a parton[36] mean-field theory [5, 37], in which we fractionalize the f-layer spins into neutral spinons $\boldsymbol{S}_{\boldsymbol{r}} = \frac{1}{2}f^\dagger_{\boldsymbol{r}\alpha}\boldsymbol{\sigma}_{\alpha\beta}f_{\boldsymbol{r}\beta}$ which introduces a local-$U(1)$ gauge redundancy $f_{\boldsymbol{r}\alpha} \to e^{i\chi_{\boldsymbol{r}}}f_{\boldsymbol{r}\alpha}$. In an exact treatment this results in an emergent dynamical $U(1)$ gauge field, a_μ, which is approximated as a static background field in the mean-field theory (fluctuations beyond mean-field are discussed below where important). Previous parton mean-field studies applied to Kondo systems find spurious finite-temperature phase transitions, which smoothen into crossovers when fluctuations about the mean-field are included [5, 38, 39]. Nevertheless, the zero temperature quantum phase transition remains sharp [5, 39, 40] and forms the focus of our following analyses.

In the parton framework Kondo screening is captured by a non-vanishing value of a (non-local) hybridization order-parameter $\Delta_{\boldsymbol{r}} = \langle c^\dagger_{\boldsymbol{r}\alpha}f_{\boldsymbol{r}\alpha}\rangle$. For $|\Delta| > 0$ the neutral f-spinons hybridize with the charged c-electrons to become ordinary charged electrons. When the average background gauge flux a_μ is zero, the system then has both a small Fermi surface of c-electrons with weak f character, and a large heavy Fermi-surface of f-electrons with weak c character; we denote this heavy Fermi liquid phase as FL. By contrast when the hybridization vanishes, $\Delta = 0$, the spinons can form a time-reversal invariant QSL with a neutral Fermi surface of spinon excitations that coexists with the small Fermi surface of c-electrons. We denote this phase as FL* following Ref. [5], which shows how gauge fluctuations beyond mean-field give strong non-Fermi liquid corrections to observable properties of the FL* phase and KS-QCP.

When ring-exchange effects are small, as is the case for the twist angles we consider, the parton mean-field-theory has been shown to instead favor mean-field solutions [41] with π emergent magnetic flux per unit cell, which divide unequally between up and down triangular plaquettes (illustrated in the Supplemental Material [31]), spontaneously breaking time-reversal and lattice translation symmetries. The emergent-magnetic field gives the spinon bands a net Chern number $C = \pm 1$, to form a chiral spin liquid (CSL). We note that, in this scenario, gauge-fluctuations are topologically-gapped due to a Chern-Simons term for a_μ induced by the spinon Chern bands. This mean-field prediction is further supported by recent 2d-DMRG [42] calculations, that show regions of CSL phase arise in triangular-lattice Hubbard models near the Mott transition. Without hybridization ($\Delta = 0$), CSL of f-spins coexists with the c-electron Fermi surface which we refer to as an Anomalous Hall* (AH*) phase. Kondo hybridization ($|\Delta| > 0$), transmits the Berry curvature of the CSL-spins to the hybridized electrons resulting in a time-reversal-symmetry broken metal with a non-zero (but not quantized) anomalous Hall (AH) conductance.

The parton mean-field phase diagram is shown in Fig. 1(c) (see also Supplemental Material [31]) for twist angle $\theta = 4.5°$. In all regimes sufficiently away from $\nu = 2$ [where a Kondo Insulator (KI) arises], AH(*) phases are favored over flux-free FL(*) phases. We have also checked for magnetic mean-field solutions with magnetization, $\langle \boldsymbol{S_r} \rangle \neq 0$, forming the 120° pattern favored by the triangular lattice Heisenberg model, but did not find any regions where AFM order either coexists with- or supplants- the AH or AH* phases. Crucially, the KS-QCP (here between AH and AH* phases) is accessible by gate tuning. We estimate that the Kondo temperature, the characteristic temperature near which Kondo screening emerges, as $T_K \gtrapprox 100K$ using $T_K = \Lambda e^{-1/(2J_K \rho_c)}$ [6] where Λ and ρ_c are respectively the c-electron bandwidth and the density of states at the Fermi-level.

Similar gate-field phase diagrams that include Kondo-screening quantum criticality lines arise over a range of twist-angles: $3.5° \lesssim \theta \lesssim 6°$. For larger θ, the f-layer is no longer a Mott insulator, and for smaller θ, the Kondo coupling is large compared to t_c so that only hybridized phases arise. We note that, if desired, the quantum criticality could potentially be accessed at smaller twist angles by suppressing the interlayer tunneling, Γ between WX_2 layers, for example, by introducing a hexagonal boron nitride (hBN) spacer layer.

Experimental signatures – Existing measurement techniques for $2d$ moiré materials offer experimental tests that can both identify Kondo-screening quantum criticality line and cleanly distinguish among the various proposed QSL and AFM scenarios for the non-hybridized phase. We focus on i) the entropy density $s(\nu)$, which

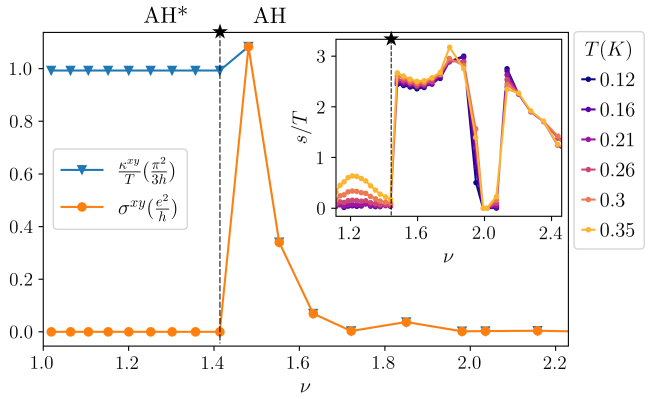

FIG. 2. Thermal (κ^{xy}) and electrical (σ^{xy}) Hall conductivities per spin, calculated using the parton mean-field bands for $\theta = 4.5°$ and $J_K = 22.2$ meV. In the AH* phase, the spinons are in a Chern insulating state with quantized κ^{xy}/T and a gap of ≈ 0.26 meV at $\nu \approx 1$. σ^{xy} jumps to the κ^{xy}/T value in the AH phase as the spinons acquire electronic character. The inset shows s/T per spin versus ν at temperatures specified by line color. The jump near the KS-QCP is due to effective mass enhancement in the AH phase when the hybridization exceeds the Chern insulator gap.

can be extracted from electrostatic measurements of $\partial\mu/\partial T = -\partial s/\partial\nu$ [43–46], and ii) electrical (σ) and thermal (κ) longitudinal and Hall conductivities, and present detailed parton calculations of these observables in the Supplemental Material [31]. Since thermal transport properties can be challenging to measure, in the Supplemental Material [31], we also describe all-electrical probes of the spinon transport properties of QSL scenarios using a combination of AC measurements and separate contact of the c- and f- layers.

In each scenario, the c-electrons contribute Fermi-liquid like behavior with $s \sim T$ and $\sigma \sim$ constant, that add to contributions of (neutral) f-spinons,with the exception of $\nu = 2$ where we find a featureless KI with activated thermodynamic and transport signatures with gap ≈ 1.14 meV. In each case, the Kondo-hybridization results in a discontinuous jump in electrical conductivities due to the incorporation of f-spins into the conducting Fermi-sea.

In the AH*/AH scenario favored by our parton calculations, the CSL of f-electrons in the AH* phase contribute a quantized thermal Hall conductivity $\kappa_f^{xy} = \frac{\pi^2}{3h}T$ per spin, and thermally-activated entropy $s \sim e^{-E_g/T}$ with E_g the spinon-gap. Kondo hybridization produces a quantized jump $|\Delta\sigma^{xy}| = e^2/h$ per spin, but smoothly evolving κ^{xy} across the KS-QCP. Note that the previous statements hold for $T \ll E_g$. Alternatively, in a time-reversal symmetric FL*/FL scenario, the f-spinons form a gapless state with a Fermi-surface. Here, due to gauge-fluctuations beyond the parton mean-field treatment, the spinon entropy contribution varies from non-

Fermi liquid-like $s \sim T^{2/3}$ [5, 47] in the FL* phase, marginal FL $s \sim T \log 1/T$ [40] at the KS-QCP, and an ordinary $s \sim T$ in the FL phase. Lastly, in a more conventional AFM/FL scenario, the spinons are confined in the AFM phase where and the f-layer spin-waves would contribute $s \sim T^2$, as well as a jump in the $\sim T$ coefficient, Hall density, and conductivity across the KS-QCP. We note that the jump in electrical conductivities across the KS-QCP is expected to be much larger in the AFM/FL and FL*/FL scenarios than the AH*/AH scenario since the spinons are gapped at the KS-QCP in the latter. Scattering of conduction electrons by spin-waves could also lead to characteristic transport signatures, as pointed out recently in Ref. [48]. The experimental signatures discussed so far for the various phases hold in a regime of temperatures much below a quantum critical crossover temperature corresponding to the condensation of Δ, and the quantum critical signatures hold above this temperature [5, 40, 64].

Discussion – Several extensions of our theoretical analysis could be fruitful avenues for future investigation. Unfrustrated orthorhombic Kondo lattice systems, like the prototypical heavy fermion compound $CeCoIn_5$, often exhibit superconductivity in the vicinity of the KS-QCP. It remains an open question whether this superconducting tendency persists in frustrated triangular lattice geometries. Second, large moire unit cells imply large magnetic fluxes per unit cell [49], and therefore significant scalar-chirality in local moment interactions, at relatively weak magnetic field B. These could favor CSL phases [50, 51] at finite B even if they were not favored at $B = 0$. The ability to apply large flux per unit cell could effectively induce orbital emergent gauge fields in the spinon Fermi surface state, leading to quantum oscillatory phenomena with $1/\kappa B$-periodicity, where κ is an $O(1)$ gauge-susceptibility [52]. However, these effects may be challenging to observe in current TMD moiré samples, due to impurity suppression which so far obscures quantum oscillations even in simple Fermi-liquid states. Another promising direction would be to explore the consequences of modifying the electronic structure by imposing relative twists between the two W-layers [53–55].

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AK and NCH contributed equally to this work.

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