

Site-polarized Mott phases competing with a correlated metal in twisted WSe₂

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Twisted WSe₂ hosts superconductivity, metal-insulator phase transitions, and field-controllable Fermi-liquid to non-Fermi-liquid transport properties. In this work, we use dynamical mean-field theory to provide a coherent understanding of the electronic correlations shaping the twisted WSe₂ phase diagram. We find a correlated metal competing with three distinct site-polarized correlated insulators; the competition is controlled by interlayer potential difference and interaction strength. The insulators are characterized by a strong differentiation between orbitals with respect to carrier concentration and effective correlation strength. Upon doping, a strong particle-hole asymmetry emerges, resulting from a Zaanen-Sawatzky-Allen-type charge-transfer mechanism. The associated charge-transfer physics and proximity to a van Hove singularity in the correlated metal sandwiched between two site-polarized insulators naturally explains the interlayer potential-driven metal-to-insulator transition, particle-hole asymmetry in transport, and the coherence-incoherence crossover in 3.65° twisted WSe₂.

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Twisted transition-metal dichalcogenide homobilayers are emerging platforms for exploring correlation phenomena [1–27] such as superconductivity [16–20,28] and metal-insulator transitions [12,14,21–27]. At the heart of this rich landscape of correlated electron physics is an extraordinarily high level of experimental control. The relative strengths of interaction and kinetic energies are controlled by twist angle θ , while important aspects of the band structure including the energy of the van Hove singularity (vHS) may be tuned *in situ* by “displacement field” (layer dependent electronic potential) E_z arising from a potential difference between top and bottom gates while carrier concentration per unit cell may also be

varied over wide ranges by varying the average of the top and bottom gate potentials [29–31].

The recent discovery of superconductivity and neighboring correlated phases in twisted WSe₂ (tWSe₂) at two distinct twist angles ($\theta = 3.65^\circ$ and 5°) [32,33] has sparked a surge of theoretical interest in this direction [34–45]. The less strongly correlated $\theta = 5^\circ$ system exhibits superconductivity in proximity to a Fermi-surface reconstructed (presumably antiferromagnetic) phase, with both phases occurring only in a small range of incommensurate hole fillings at which the Fermi surface is in proximity to the van Hove singularity [33]. The more strongly correlated $\theta = 3.65^\circ$ system exhibits superconductivity at carrier concentrations very close to the commensurate hole filling of $\nu = 1$ and at very small displacement fields, transitioning at $\nu = 1$ into a correlated insulator upon applying an E_z [32] that is much smaller than the bandwidth, indicating a strong effect beyond a single-particle description. 3.65° tWSe₂ also exhibits unusual transport properties including a maximum at a characteristic temperature $T^* \sim 10$ K in the normal state and a strong particle-hole asymmetry in the magnitude of longitudinal resistance R and

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in the Fermi-liquid (FL) coherence temperature T_{FL} below which $R \propto T^2$ (T denotes temperature). Although the pairing mechanism of superconductivity is yet to be fully understood, its proximity to such correlated phases and unusual transport implies a potential connection between them; see, e.g., Refs. [38,43].

Here, we propose a physical mechanism underlying the correlated electron behavior observed in 3.65° tWSe₂ which we validate by applying dynamical mean-field theory (DMFT) [46,47]. While DMFT is formally exact only in the infinite-dimensional limit, it is widely documented to be an accurate and reliable approximation even in two dimensional systems, provided the correlation lengths associated with incipient long-range orders are small. DMFT has been demonstrated to accurately capture self-energy effects deduced from photoemission spectra as well as anisotropic mass renormalization and the crossover into an incoherent regime upon increasing temperatures in quasi-two-dimensional materials including Sr₂RuO₄ [48,49], Fe-based superconductors [50], and, more recently, moiré materials [14,51–53]. Motivated by these established successes, we employ DMFT here to capture the effects of local correlations in a multiorbital model that faithfully captures the band structure and topology [43,54,55]. The model includes three localized orbitals per moiré unit cell, labeled T, H₁, and H₂, centered at positions corresponding to local MM, MX, and XM stackings (where M denotes a transition-metal atom and X a chalcogen atom), respectively, as illustrated in Fig. 1(a). We demonstrate that a Mott MIT occurs at $\nu = 1$ induced either by applying E_z or tuning the dielectric constant ϵ . The Mott insulating state features a large polarization of orbital (or site) occupations with one of the T, H₁, and H₂ sites occupied by approximately one hole and the other two having a very low hole occupancy. The occupancy difference leads to strong orbital differentiation in effective correlation strength and to a strong particle-hole asymmetry upon doping. The doping asymmetry arises from charge transfer physics similar to that discussed by Zaanen, Sawatzky, and Allen (ZSA) [56]. Remarkably, the combination of charge-transfer physics and the proximity to a vHS in a correlated metal sandwiched between two distinct site-polarized insulators naturally explain several key features observed in the experiment.

Our Hamiltonian reads $\mathcal{H} = H_0 + H_{\text{int}}$. Here, H_0 is the kinetic term that accounts for the band structure and includes a layer-dependent potential lE_z (with $l = \pm 1$ denoting the two layers). Because the H₁ and H₂ orbitals are localized on the top and bottom orbitals, respectively, while the wave function of the T orbital is equally divided between the two layers [Fig. 1(a)], a nonzero displacement field splits the on-site energies of the H₁ and H₂ orbitals while leaving the on-site energy of the T orbital unchanged. We construct H_0 via a one-shot Wannierization using an energy-dependent weighting factor applied to the entire Bloch states [57–59]; see the Supplemental Material [60] and Ref. [43]. Figure 1(b) compares the continuum bands (green lines) to the Wannier band structure (black lines) resulting from diagonalization of H_0 at $E_z = 5$ meV. The topmost band, which is the most relevant near $\nu = 1$, is perfectly reproduced, in both dispersion and band topology (not shown here) and the second band is very well reproduced. The third band (whose details are not

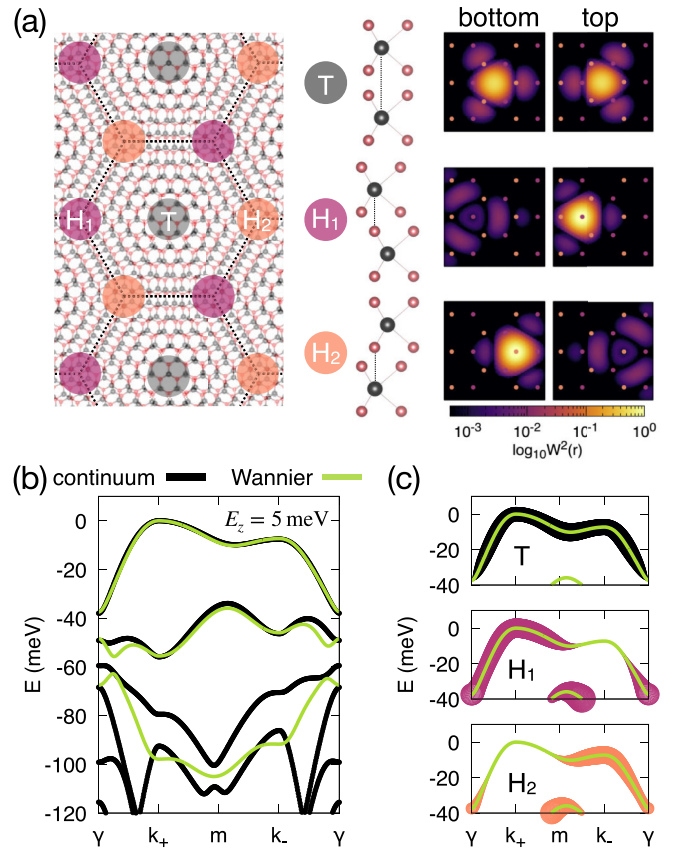


FIG. 1. (a) Sketch of the local MM (T), MX (H₁), and XM (H₂) stacking regions in the moiré lattice of tWSe₂ and the amplitude of the corresponding Wannier functions on the top layer ($l = +1$) and the bottom layer ($l = -1$) at $E_z = 0$. (b) Three-orbital Wannier bands for one valley/spin ($\sigma = \uparrow$) overlaid with continuum model bands at $E_z = 5$ meV along with (c) the corresponding spectral weight of Wannier orbitals in the topmost band. The opposite valley/spin ($\sigma = \downarrow$) states are obtained via time-reversal conjugation of the $\uparrow$ states.

relevant to our calculations) represents an average over the spectral weight of the remote continuum bands, which must be included to have a model with three exponentially localized Wannier orbitals with a vanishing total Chern number [43,55]. All three orbitals have substantial spectral weight in the topmost band [Fig. 1(c)], with the H₁ (H₂) orbital contributing predominantly near the k_+ (k_-) point, reflecting the nearly complete layer polarization of the corresponding Wannier function [Fig. 1(a)]. At $\nu = 1$ and $E_z = 0$, the orbital-resolved hole occupations are $\nu_T \simeq 0.5$ and $\nu_{H_1} = \nu_{H_2} \simeq 0.25$.

The interaction term H_{int} accounts for local and nonlocal density-density Coulomb interactions, which reads

$$H_{\text{int}} = \sum_{i,\eta} \frac{U_{\eta}}{\epsilon} n_{i\eta\uparrow} n_{i\eta\downarrow} + \frac{1}{2} \sum_{i,j,\eta\eta',\sigma\sigma'} \frac{U_{i\eta,j\eta'}}{\epsilon} n_{i\eta\sigma} n_{j\eta'\sigma'}, \quad (1)$$

where i, j denote Bravais lattice cells, $\eta, \eta' \in \{T, H_1, H_2\}$ the orbital, and $\sigma, \sigma' \in \{\uparrow, \downarrow\}$ the spin locked to the valley degree of freedom [21,29]. $n_{i\eta\sigma}$ is the associated hole number operator. $U_{i\eta,j\eta'}$ refers to Coulomb interaction between orbitals η and η' located in Bravais lattice cells i and j , respectively,

obtained from evaluating double-gate Coulomb interaction in the Wannier basis. $U_{i\eta,j\eta'}$ vary by at most $\sim 10\%$ upon applying a large E_z of $E_z = 20$ meV, so we use $U_{i\eta,j\eta'}$ obtained at $E_z = 0$ for all our calculations; see the Supplemental Material [60]. $U_\eta \equiv U_{i\eta,i\eta}$ and $U_H \equiv U_{H_1} = U_{H_2}$. We find that $U_H \simeq 1.7U_T$ due to the stronger localization of H_1 and H_2 Wannier orbitals compared to that of the T orbital. We rescale Coulomb interactions by introducing an adjustable parameter ε , which acts as a dielectric constant; see the Supplemental Material [60]. Non-density-density interactions are at least an order of magnitude smaller and are thus neglected.

Our DMFT calculations treat local interactions [the first term in Eq. (1)] with the numerically exact hybridization-expansion continuous-time quantum Monte Carlo method [61–63] and nonlocal interactions [the second term in Eq. (1)] with the Hartree approximation. The Fock self-energy is neglected to prevent bandwidth overestimation [64]. The model is solved in the hole representation (see the Supplemental Material [60]). Since there are three sites per unit cell, three impurity problems must be solved in every DMFT self-consistency loop. Where necessary, the self-energy is analytically continued from Matsubara to real frequency (ω) axis using the maximum entropy method [60,65,66].

The DMFT phase diagram in the ε - E_z plane for $\nu = 1$ is shown in Fig. 2(a). We first identify a correlated metallic phase near $\varepsilon \sim 10$ and in the small $|E_z|$ regime. It is characterized by dominant T-orbital hole occupation and a small quasiparticle weight Z at $\omega = 0$, accompanied by incoherent spectral weight at higher energies [cf. Fig. 2(b)]. Interestingly, this metal undergoes first-order Mott transitions to three distinct correlated insulating phases upon a slight tuning of either E_z or ε .

The three correlated insulators are characterized by a strong orbital polarization in the hole occupation, which results in localization of holes at the sites hosting the relevant orbital, as depicted in the bottom panels of Fig. 2(c). This polarization is influenced by E_z , which introduces energy difference between the top and bottom layers: E_z shifts the hole energy level of H_1 (H_2) orbital approximately by $-E_z$ ($+E_z$). Correspondingly, we find H_1 - and H_2 -polarized insulators for $E_z > 0$ and $E_z < 0$ regimes, respectively. This transition is facilitated by Coulomb interactions, so the phase boundaries between metal and the insulators are shifted toward smaller $|E_z|$ as ε decreases. This underscores the decisive role of Coulomb interactions and naturally explains how a small E_z can induce the MIT as observed in experiment [32]. Here, nonlocal interactions between orbitals of different character, as well as the local interactions, promote site-polarized localization by penalizing a homogeneous charge distribution among orbitals. Below a critical ε , Coulomb interactions for the T orbital also become large enough to localize T-orbital holes, resulting in a Mott transition in the T-orbital sector as shown in Fig. 2(a). In this regime, the T-polarized insulator preempts the formation of H_1 - and H_2 -polarized insulators.

Further insight can be obtained by examining spectral functions shown in Fig. 2(c). Note that the spectral functions are defined here for electrons, so that increasing the hole occupancy ν corresponds to populating fewer states as a function of ω . For the three distinct site-polarized insulators, a Mott gap opens in the nearly half-filled orbital sector,

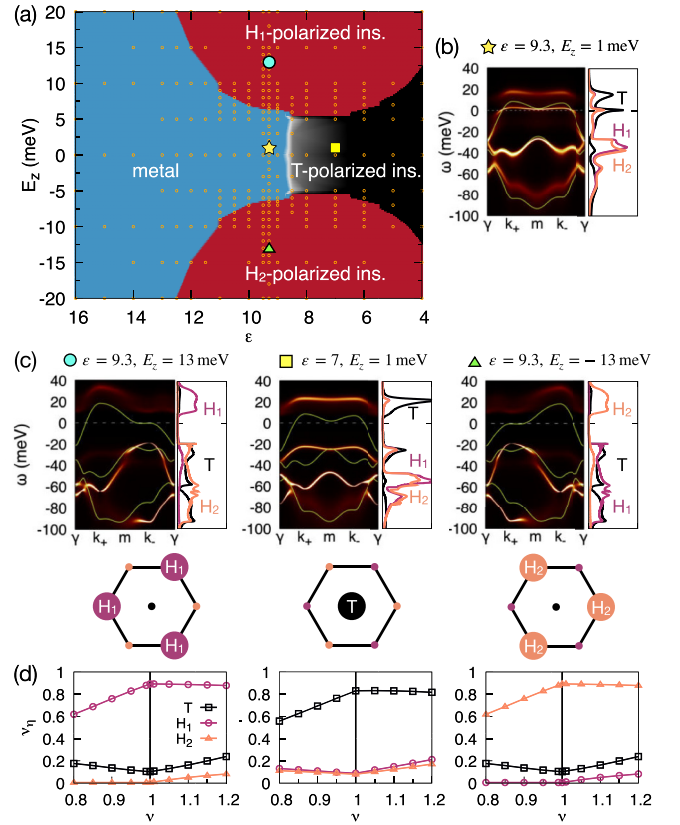


FIG. 2. (a) Phase diagram in the ε - E_z plane at hole filling $\nu = 1$ and $T = 5.8$ K. (b), (c) Momentum-resolved and momentum-integrated spectral functions for $\sigma = \uparrow$ and $\nu = 1$ obtained at the (ε, E_z) values marked by colored symbols in the phase diagram. The continuum-model bands (green) are overlaid. The schematics in the bottom panels of (c) illustrate the hole occupation at each site, represented by the size of the circles for the H_1 -polarized (left), T-polarized (middle), and H_2 -polarized (right) insulators. (d) The orbital-resolved hole occupancy ν_η as a function of total occupation ν for the same (ε, E_z) sets as in the upper panels in (c). The symmetry of the system under $H_1 \rightarrow H_2$ upon $E_z \rightarrow -E_z$ leads to a corresponding symmetry of the spectral functions (c) and of the orbital occupancies (d).

whereas the spectral weight of the remaining orbitals lies below the chemical potential—indicating the absence of hole occupation—in sharp contrast to the noninteracting case [cf. Fig. 1(c)]. This polarization leads to strong site differentiation in effective correlation strength, as defined by the imaginary part of the DMFT self-energy: the nearly half-filled orbital exhibits strong correlations subject to a Mott transition (large self-energy), while the nearly empty (i.e., no holes) orbitals are weakly correlated (small self-energy).

A clear dichotomy emerges between electron-doped and hole-doped sides of these insulators near $\nu = 1$. As inferred from the spectral functions in Fig. 2(c), electrons are doped into strongly correlated orbitals, whereas holes are doped into weakly correlated ones. This is corroborated by the orbital-resolved hole occupancy ν_η obtained from DMFT around $\nu = 1$ presented in Fig. 2(d). Namely, for $\nu > 1$, holes are doped into the weakly correlated orbitals, whereas ν_η for the strongly correlated orbitals remains nearly unchanged. In this

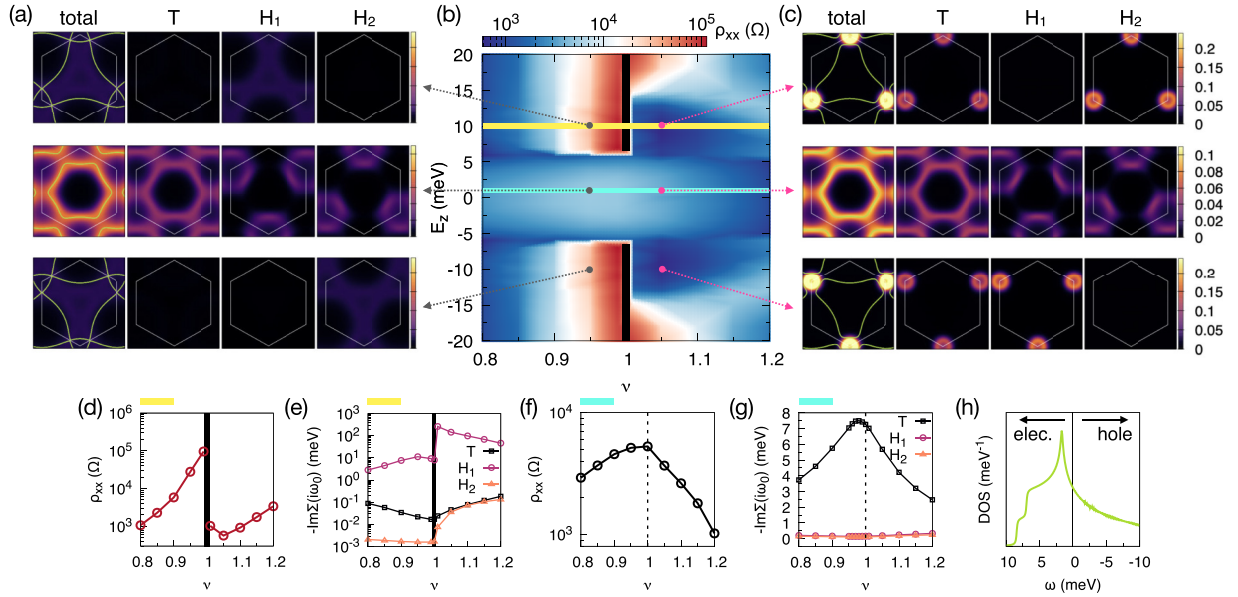


FIG. 3. (a),(c) k -resolved spectral functions of spin- $\uparrow$ character at the Fermi level in the Brillouin zone for (a) electron-doped and (c) hole-doped regions, approximated by the DMFT Green's function $-G(\mathbf{k}, \tau = \frac{1}{2T})/(\pi T)$ with τ imaginary time. Noninteracting Fermi surface (green line) is overlaid. (b) Calculated resistivity ρ_{xx} using the DMFT self-energy as a function of ν and E_z at $\varepsilon = 9.3$ and 5.8 K. The black bars indicate the insulating regions. (d), (f) ρ_{xx} along the yellow and cyan lines indicated in (b). (e), (g) Scattering rate Γ which is approximated by the imaginary part of the self-energy at the lowest Matsubara frequency $\omega_0 = \pi T$, $\Gamma = -\text{Im}\Sigma(i\omega_0 = i\pi T)$, calculated along the yellow and cyan lines in (b). (h) Noninteracting density of states at $E_z = 1$ meV. The vertical black solid line at $\omega = 0$ indicates the Fermi level at $\nu = 1$.

respect, these site-polarized insulators can be classified as charge-transfer insulators in the ZSA scheme [56].

Motivated by the recent experiment by Xia *et al.* [32], we next focus on the transport properties. In DMFT, the conductivity can be obtained as [47]

$$\sigma_{\alpha\beta}^{\text{dc}} = \frac{\pi e^2}{\hbar S N} \sum_{\sigma k} \int d\omega \left(-\frac{\partial f}{\partial \omega} \right) \text{Tr} [v_{\sigma k}^{\alpha} A_{\sigma k}(\omega) v_{\sigma k}^{\beta} A_{\sigma k}(\omega)], \quad (2)$$

where S denotes the area of the unit cell, N the number of k points in the first Brillouin zone, and f the Fermi-Dirac function. $v_{\sigma k}^{\alpha}$ is the Fermi velocity incorporating intraunit cell terms in direction $\alpha \in \{x, y\}$ [67] and $A_{\sigma k}(\omega)$ the momentum-resolved spectral function, both in the orbital basis; see the Supplemental Material [60]. Tr means the trace over orbital indices. Equation (2) neglects vertex corrections. This is valid in DMFT for single-band systems and extends to multiorbital systems under assumptions discussed in Ref. [68].

Figure 3(b) shows the resistivity $\rho_{xx} = [\sigma^{\text{dc}}]_{xx}^{-1}$ around $\nu = 1$ at $\varepsilon = 9.3$ and 5.8 K. In good agreement with the experiment [32], two strongly resistive regions sandwich the metallic phase at $\nu = 1$, along with a pronounced asymmetry in ρ_{xx} , which is larger on the electron-doped side ($\nu < 1$) than on the hole-doped side ($\nu > 1$). As discussed earlier, these two strongly resistive regions correspond to the H_1 - and H_2 -polarized insulators for $E_z > 0$ and $E_z < 0$ regimes, respectively. The asymmetry in ρ_{xx} for these H_1 - and H_2 -polarized insulators originates from the charge-transfer physics, which induces a strong dichotomy in the orbital character of the low-energy spectral weight between the

electron-doped and the hole-doped sides. This is clearly manifested in the k -resolved spectral function at the Fermi level $A_k(0)$. On the electron-doped side [$\nu < 1$, Fig. 3(a)], only the strongly correlated orbital, namely, H_1 for $E_z > 0$ (top row) or H_2 for $E_z < 0$ (bottom row), constitutes the spectral weight at the Fermi level. This results in an incoherent Fermi surface due to a large scattering rate, and thus bad conductivity; see also Fig. 3(d). On the other hand, on the hole-doped side [$\nu > 1$, Fig. 3(c)], the strongly correlated orbital is gapped out and holes are doped to weakly correlated orbitals due to the charge-transfer nature of the insulator [cf. Fig. 2(d)]. This leads to a coherent Fermi surface and good conductivity, and explains why the electron-doped side shows higher resistivity than the hole-doped side, despite the strongly correlated orbital showing a much larger scattering rate on the latter [cf. Fig. 3(e)]. As more holes are doped, however, scattering rates of T and H_2 are gradually enhanced followed by an increase in resistivity; see the $\nu > 1.05$ side of Fig. 3(d). Note that charge redistribution among the orbitals (due to the site-polarized Mott transition at $\nu = 1$) makes the Fermi surface substantially different compared to the noninteracting one indicated by the green line in Fig. 3(c).

Interestingly, we also find an asymmetry of ρ_{xx} in the small $|E_z|$ regime, despite the absence of a site-polarized Mott transition at $\nu = 1$; see Fig. 3(f). In this regime, the T orbital dominates the spectral weight at the Fermi level over the H orbitals as shown in the middle row of Figs. 3(a) and 3(c), and the DMFT Fermi surface closely resembles the noninteracting one, with some smearing due to the large scattering rate Γ ($> T$) of the dominant T orbital, as shown in Fig. 3(g). In principle, both a weakened precursor of the orbital polarization

scenario or a mechanism distinct from that near the insulating regime [Fig. 3(d)] could be responsible for the resistivity asymmetry.

Regarding the latter, a likely contributor to the asymmetry here is the proximity to the vHS inherent to the noninteracting band structure. The noninteracting density of states (DOS) $D(\omega)$ presented in Fig. 3(h) clearly exhibits the vHS on the electron-doped side. This corresponds to smaller velocities on the electron-doped side and also suppresses the intersite hybridization. This effect is reflected in the local hybridization function $\Delta_0(i\omega)$ at low frequencies, obtained from the noninteracting local Green's function $G_0(i\omega) = \text{Im}\Delta_0(i0^+) = \text{Im}G_0(i0^+)/[\text{Re}G_0(i0^+)^2 + \text{Im}G_0(i0^+)^2] \simeq -1/[\pi D(0)]$, where $D(0) = -\text{Im}G_0(i0^+)/\pi$ [48]. Thus a larger DOS suppresses the effective coupling of an orbital (site) with its neighbors, leading to stronger correlations [48,69–75]. It is indeed evidenced by the T-orbital scattering-rate asymmetry which exhibits larger values near $\nu = 1$ on the electron-doped side than on the hole-doped side as shown in Fig. 3(g). This effect also prevails in a single-orbital model using the topmost band only (see the Supplemental Material [60]).

The experiment of Ref. [32] shows a reentrant metallic phase at $\nu = 1$ in the regime of very large $|E_z|$ that our results do not capture. There can be different reasons for this: as one possibility, in the very large- E_z regime, the two layers can be effectively decoupled such that the moiré potential becomes weakened and the ansatz of the three Wannier functions sketched in Fig. 1(a) remains no longer valid. As a second possibility an interplay of enhanced screening, ordering, and nonlocal correlations could be at play. Indeed, both fRG [43] and Hartree-Fock [45] studies of the same model albeit assuming enhanced dielectric screening find ordering tendencies at small to intermediate E_z that are suppressed at very large E_z .

We now investigate the temperature dependence of ρ_{xx} in the metallic phase. In the experiment by Xia *et al.* [32], superconductivity emerges within a narrow filling range around $\nu = 1$ in the metallic phase with an onset temperature of ~ 200 mK. At higher temperatures in the normal state, a characteristic temperature scale $T^* \sim 10$ K is identified, at which the resistance R exhibits a maximum; see the right panel of Fig. 4(a). Our calculated ρ_{xx} in Fig. 4(a) agrees well with the experimental data. Notably, ρ_{xx} is smallest at $\nu = 1.1$ and ρ_{xx} at $\nu = 0.9$ and $\nu = 1$ become comparable in the low-temperature regime ($T < 10$ K). Additionally, ρ_{xx} exhibits a maximum at $\nu = 1$ as in the experiment, although the corresponding T^* differs by about a factor of 3.

The T^* is associated with the destruction of quasiparticles as shown in Fig. 4(b), which is characteristic of a correlated metal in proximity to a Mott insulator at elevated temperatures [76–79]. At very low temperatures, a (resilient) quasiparticle emerges and thus a sharp peak in the spectral function at $\omega = 0$ is identified along with Hubbard bands at higher energies. As the temperature increases, the resilient quasiparticles become progressively shorter lived and are completely destroyed in the temperature range between 23.2 and 38.7 K, revealing a correlation-induced minimum in the density of states resulting from the combination of proximity to a Mott state and the absence of coherent quasiparticles. Thus the state at $T > T^*$ may be considered to have a pseudogap. We therefore

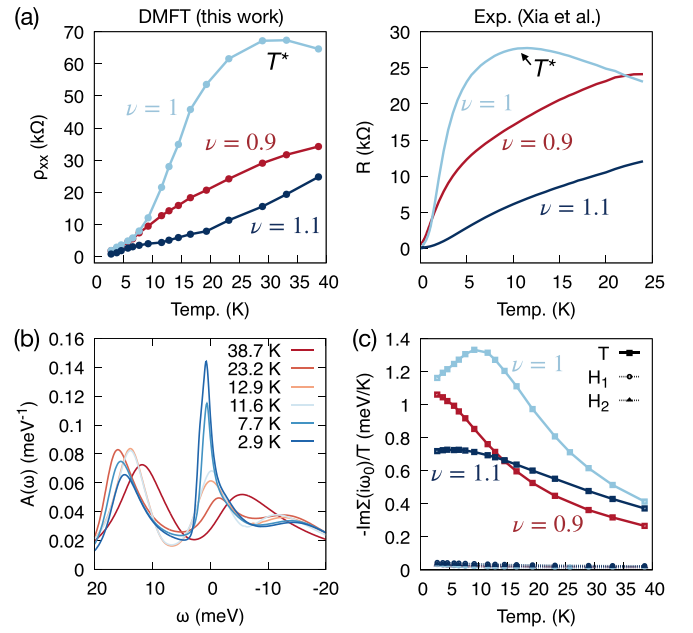


FIG. 4. (a) Left: T dependence of the resistivity ρ_{xx} at $\varepsilon = 9.3$ and $E_z = 1$ meV. Right: the experimental resistance R reported by Xia *et al.* at the displacement field of $E \simeq 8$ mV/nm [32]. (b) Momentum-integrated spectral function $A(\omega)$ for $\nu = 1$ at various temperatures. (c) T dependence of $-\text{Im}\Sigma(i\omega_0)/T$ for the data underlying panel (a).

suggest that the resistivity maximum observed in experiment is interpreted as a signature of an incoherent nearly Mott state.

On the doped sides, the pronounced asymmetry between $\nu = 0.9$ and $\nu = 1.1$ due to the vHS persists up to higher temperatures in agreement with the experiment [32]; see Fig. 4(a). An interesting experimental observation is that the FL coherence temperature T_{FL} is significantly larger on the hole-doped side and rapidly decreases as ν approaches $\nu = 1$ from above ($\nu > 1$). Extracting the T scaling of ρ_{xx} at low temperatures, however, is challenging in our calculations as it requires highly precise analytic continuation of three independent self-energy data (one for each orbital) at every T . Instead, we investigate the imaginary part of the self-energy at the lowest Matsubara frequency $\text{Im}\Sigma(i\omega_0)$, as shown in Fig. 4(c). In a FL regime at sufficiently low temperatures, $-\text{Im}\Sigma(i\omega_0)/T = (Z^{-1} - 1)\pi + \mathcal{O}(T^2)$ [14,80,81]. Thus T_{FL} can be identified as the temperature below which $-\text{Im}\Sigma(i\omega_0)/T$ forms a plateau. Indeed, we find the asymmetry in T_{FL} as in the experiment. Namely, $-\text{Im}\Sigma(i\omega_0)/T$ saturates below ~ 7 K at $\nu = 1.1$, which is comparable to the experimental T_{FL} of 3.5 K [32]. In contrast, it continues to increase at $\nu = 0.9$ and decrease at $\nu = 1$ down to the lowest temperature we examined (2.8 K), indicating that T_{FL} is lower in those cases. Thus the vHS here not only increases the magnitude of resistivity, but also suppresses the FL coherence by enhancing electron correlations.

To conclude, we have demonstrated that the charge-transfer physics and proximity to the vHS of the correlated metal sandwiched between two distinct site-polarized insulators provide a coherent explanation for several key features observed in the

experiment on 3.65° tWSe₂ [32], including metal-insulator transitions, their tunability with displacement field, doping and temperature dependence of the electronic transport properties, and a coherence-incoherence crossover. A central open question regards ordering phenomena, particularly possible spin- or valley-polarized electronic orders emerging from site-polarized Mott insulators and the mechanism by which superconductivity arises in the correlated metallic phase at small $|E_z|$. Understanding how electronic phases evolve with decreasing correlation strength as $\theta \rightarrow 5^\circ$ [33] and developing a unified framework for superconductivity in tWSe₂ remain outstanding challenges. The present study, focused on the strongly correlated regime at 3.65° , provides a natural starting point and theoretical foundation for this effort.

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Data availability. The data that support the findings of this Letter are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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