Peer Review File

Manuscript Title: Quantum Criticality in Twisted Transition Metal Dichalcogenides

Editorial Notes: *none*

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referee #1 (Remarks to the Author):

The authors characterized the gate-drive metal-insulator transitions and the metallic phase in twisted WSe2 near half-filling of the first moiré subband through transport measurement. In the continuous metal-insulator transition, quantum criticality is further revealed. A new platform for the study of quantum phase transitions is provided. The study is of interest and exhibits a number of elements of novelty. I would like to recommend the acceptance of the manuscript if the following comments are addressed.

1. The authors claimed that T-linear behaviors are observed on either side of insulating state near half-filling, and no such behaviors but T^2 behaviors are observed near trivial insulators at full filling. However, it seems to me that at certain fillings (e.g. v=-2.52, v=-0.94 in fig.3c), either the T^2 or T-linear fitting is fine. Especially, at an even smaller doping of v=-0.84, both T^2 and T-linear fitting are adopted. Why couldn't it be a T-linear fitting with two different slopes? Could the author justify the using of T^2 and T-linear fitting at different fillings?

2. About the discussion of the Planckian dissipation and parameter C, could the author present more details and further explanation? For example, what is the doping dependence of C near the insulating states at half-filling? For the filling with C~1 which satisfies Planckian dissipation, the possible origins should be clarified.

3. Compared with the crossover from B^2 to B-linear dependence near the insulating phase of half-filling, do the authors have B-dependence of the longitudinal and Hall resistance near the insulator of full filling?

4. In the triangular-lattice Hubbard model, the insulating phase at half-filling is predicted to be non-magnetic and is a spin-liquid of some type. Could the author present more evidence in experimental in support of this theory? For example, how do the insulating gaps around half-filling evolve with B-field?

5. With v fixed at -1, when ρ_0 is between 2-3kOhm, the gap fully opens. The authors proposed the singular interplay of impurity scattering and scattering from fluctuations of the order parameter of the insulating state. Do the author have a more detailed scenario to describe such an interplay?

6. In theory [T.Senthil, PRB, 78, 045109 (2008)] that predicts spin liquid behavior in continuous Mott transition, a divergent effective mass at the critical point is also predicted. Do the author have any evidence indicating the divergent effective mass associated with a rapid rise of ρ_0?

Referee #2 (Remarks to the Author):

The manuscript by A. Ghiotto et al. reports continuous metal-insulator transitions (MITs) in twisted WSe2 bilayers near the half-filling of their first moiré band. In particular, the authors observe continuous MITs by tuning either the carrier density or displacement field and explain their results in terms of doping- and bandwidth-controlled MITs, respectively. They also find non-Fermi liquid behaviors near the quantum critical regime in the resistance-temperature and magneto-resistance data.

While continuous MITs have been observed in other systems via doping control, the bandwidth tuning often couples with structural changes that give rise to first-order phase transitions. The current manuscript demonstrates the moiré system as a platform to investigate the quantum critical regime by tuning both doping and bandwidth. Therefore I think it would be of great interest to researchers studying 2D materials and correlated electron systems. However, I have several concerns regarding the experimental scheme and data analysis (listed below). I wish the authors provide additional data/analysis to back up their conclusions before I could recommend its publication in Nature.

Independent control of the density and displacement-field:
1. In most of the experiments (Figs. 2-4), the authors applied a constant top gate voltage while sweeping the other gate voltage. It appears to me that this would change both the doping concentration and the displacement field (purple line in Fig. 1c). However, the authors explained these data mostly in terms of a doping-controlled MIT. While doping might have a more profound effect on the transport than the displacement field, it is not clear whether the effect of varying displacement field can be ignored entirely (particularly for Fig. 3 with a large voltage range with a significant change in D, which would change the electronic structure). I suggest the authors provide additional data at a fixed displacement field to double-check their central thesis: the continuous MIT and quantum critical regime.

2. In Fig. 1c, at low displacement field D, the sample resistance becomes highly asymmetric across nu = -1. The peak resistance seems to shift away from nu = -1 at the lowest D. Why would this happen? I wonder if this could be an artifact related to contact resistance. What are the resistance values in the cut-out white region at the lower right corner?

3. The magnetotransport data in Fig. 4 is taken from a much higher V_TG voltage than Figs. 2 and 3. It seems that this is due to contact resistance preventing magnetotransport measurements at low V_TG. Is that right? What value of D does Fig. 4 correspond to in Fig. 1c?

4. More importantly, because the authors relate the coefficients for temperature- and B-field-dependence of resistance in their analysis, I think such a direct comparison might make more sense if the data are obtained under the same displacement field. I suggest adding the temperature-dependence data under the same condition as in Fig. 4.

Mechanism of field-driven MIT:
5. Another concern I have is regarding the MIT mechanism. The authors propose that the system be considered a one-orbital triangular lattice Hubbard model simulator where displacement controls the bandwidth. However, considering the layer degeneracy, one might also explain the observations in terms of changing the filling ratios on the two layers (i.e., layer polarization under a displacement field, which may require consideration of two bands/orbitals). Could the proposed bandwidth control

picture with one band be an oversimplified one? Another related but more minor point, where is the in-plane Wannier function located in the moiré superlattice? It seems to me that the electrons in the bilayer could form a honeycomb lattice but not a triangular one.

Analysis of the temperature-dependence:
6. Because the resistance value and its temperature dependence are central to the conclusion, I think additional details shall be provided on measuring resistance and quantifying its T-dependence. For example, it would be essential to eliminate any contribution from the contact resistance. What are the typical contact resistance values, and how do they change as a function of temperature and doping? It would be helpful to show a few representative I-V curves too.

7. Besides, the resistance data shows quite complicated temperature dependence, so it would be helpful to explain the data fitting procedure further. For example, near the quantum critical point, the authors use both T-linear and T-quadratic dependence to fit the same data but at different temperature ranges. How are these temperature ranges chosen? If the R-T curve with T-squared dependence begins to saturate, would there always be some region where one can fit with T-linear (d2^R/d^T2 ~0)? For example, one might be able to get a good linear fit between 50K and 75K for nu = -1.66 in Fig. 3c.

8. Similarly, it would be great if the authors can specify how to choose the proper temperature range to extract activation energy for insulating states.

9. Is there a systematic procedure to determine which types of fitting (T vs. T^2) to use? For example, the R-T curve might start to deviate from linear near base temperature (or if the temperature were lowered further) for nv=-0.94 in Fig. 3c.

10. Could the authors explain how error bars are determined in the lower panel of Fig. 3b? A bit surprisingly, the error bars do not change much, especially for the T-linear curve, even when the system approaches the Fermi-liquid region.

11. I wonder if the authors could estimate the level of disorder in the system and comment on the possible disorder effects in explaining the continuous doping-controlled MIT? Do authors observe any spatial inhomogeneity or anisotropy in their measurements?

Other minor points:

12. I am confused by the 2D color plots in Fig. 2: they show the phase diagram down to 0 K, but experiments were only down to 1.5 K. Is that correct? A more accurate way to represent the data would be to leave white spaces below 1.5 K.

13. In Fig. 3, why does not the insulating state appear at nu = -1?

14. Some important details are missing for readers to understand and reproduce the results, which would be very helpful to include. For example, what is the thickness of the top and bottom hBN? This will be important to convert gate voltage into a displacement field. How is the twist angle determined? It would be helpful to show a microscope image of the device for readers to relative positions of gate and source-drain (since a gate may activate the source-drain in certain device structures).

15. Currently, all the experiments are under the positive displacement field, and in principle, the device's behavior shall be the same for negative displacement. Is there any asymmetric gate response since the device structure is not symmetric (because Pt contacts WSe2 from the bottom)? It would

also be great if the authors could define the positive direction of the D.

16. In Fig. 5a, the third R-T curve from the top shows decreasing resistance with increasing T even at high temperature and cross with the fourth curve. Is it real?

Referee #3 (Remarks to the Author):

The authors study the metal-insulator transition in twisted WSe2 bilayers. These systems are attracting great attention for their emergent correlated states arising from the existence of nearly flat bands. The authors show that by controlling the density and dispersion electrically, a continuous correlated-metallic phase transition is found. Importantly, the authors find that at the phase boundary, linear T resistivity behavior emerges, a paradigmatic hallmark of critical behavior in correlated systems. In contrast, deep in the metallic regime, the authors find the usual T^2 behavior of conventional metals. The authors highlight that these findings show that this system is a very well-suited platform to study correlated behavior on the triangular lattice.

I found their manuscript greatly interesting and an outstanding demonstration of the high-quality correlated physics that can be observed in twisted WSe2 systems. Their experiments are fully consistent with previous findings and provide compelling evidence of strongly correlated phenomena in twisted dichalcogenides beyond reported correlated regimes. Overall, I believe that their results are correct and that they report exciting new phenomena in twisted van der Waals materials. For this reason, I think that their manuscript will be of tremendous interest for the communities of twisted van der Waals materials and correlated physics, and therefore I recommend publication of their work in Nature.

Besides my positive recommendation for publication in Nature, I would also like to bring up a few minor points that the authors may want to consider in the final version of their manuscript:
- The authors claim that this system is “a one-orbital triangular lattice Hubbard model simulator”, yet I would say that this is probably an oversimplification. First, the existence of spin-orbit coupling probably creates spin-dependent hoppings, the electronic structure may involve second neighbor hoppings, and the electronic interactions quite probably involve non-local interactions. Therefore, I would suggest that the authors rewrite that sentence as “a one-orbital correlated triangular model simulator.”
- As the authors noted, linear-T behavior can also appear due to electron-phonon coupling (as suggested in twisted graphene bilayers in Ref. [33]). The authors argue that in their experiment, such electron-phonon mechanism can be discarded, yet from my understanding of their argument, although their argument is suggestive, strictly speaking, it may be partially circumstantial. Therefore, I would suggest that the authors rewrite the statement in a milder way.
- The authors further claim that the correlated state is “most likely a spin liquid”. While theoretical arguments may put that as a possibility, I would say that there is no strong experimental evidence that the system is a spin-liquid. Therefore, this may be an overstatement, and I would suggest that the authors mention the spin liquid just as a hypothetical possibility rather than the most likely scenario.

To summarize, I think that the current experiment provides compelling experimental evidence of further correlated behavior in twisted dichalcogenide bilayers, a topic of critical interest for the community, and therefore I strongly recommend the publication of their manuscript in Nature.

Author Rebuttals to Initial Comments:

REFEREE 1

The authors characterized the gate-drive metal-insulator transitions and the metallic phase in twisted WSe2 near half-filling of the first moiré subband through transport measurement. In the continuous metal-insulator transition, quantum criticality is further revealed. A new platform for the study of quantumphase transitions is provided. The study is of interest and exhibits a number of elements of novelty. I would like to recommend the acceptance of the manuscriptif the following comments are addressed.

We thank the referee for the time taken to review the manuscript and for the constructive comments offered.

1. The authors claimed that T-linear behaviors are observed on either sideof insulating state near half-filling, and no such behaviors but T^2 behaviors are observed near trivial insulators at full filling. However, it seems to me that at certain fillings (e.g. v=-2.52, v=-0.94 in fig.3c), either the T^2 or T-linear fitting is fine. Especially, at an even smaller doping of v=-0.84, both T^2 and T-linear fitting are adopted. Why couldn't it be a T-linear fitting with two different slopes? Could the author justify the using of T^2 and T-linear fitting at different fillings?

The referee raises the question of when the T^2 and $T-$ linear fittings are appropriate. Apart from the insulating regions and the quantum critical points themselves, we find that the lowest temperature resistivity follows a T^2 form, as we describe in more detail below. We therefore first attempt to fit the lowest temperature data to a T^2 form. A fit is con- sidered acceptable if the r^2 value is higher than 0.95. This fit criterion defines a maximum temperature to which the T^2 behavior holds, which is density dependent as shown in figure3b of the main text.

The region of $T-$ linear behavior is found above the region of T^2 behavior, except for atthe quantum critical point itself where the $T-$ linear behavior extends down to the lowest temperatures. A similar criterion ($r^2 > 0.95$) is used to mark the upper boundary of $T-$ linear behavior at each doping.

An example of this procedure is shown in fig. R1 for the doping of $\upsilon = -0.84$ in main

text figure 3c that the referee cites in his question. Following our fitting procedure, we findthat the resistivity is fit well (r^2=0.997) by a T^2 form up to a temperature of 16.5K, and is fit well (r^2=0.993) by $T-$ linear form between 16.5 and 47 K. The referee asks whether a linear fit would be appropriate at low temperature with a different slope. We find this not to be the case - for the data at $v = -0.84$, a linear fit between 1.6 and 6 K yields a r^2 of0.92, clearly worse than the quadratic fit.

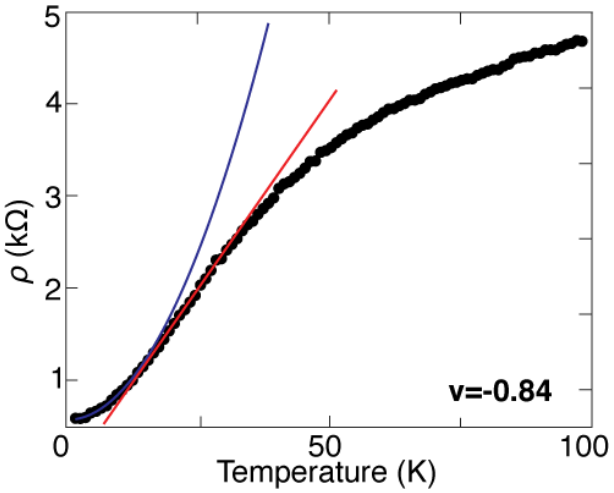

FIG. R. 1: **Appropriateness of T^2 and $T-$ linear fits.** Temperature dependence of the resistivity at $v = -0.84$ (from fig. 3c of the main text), together with fits to quadraticand linear temperature dependence over the appropriate temperature ranges.

To make these points clear to the reader, we have added the discussion above to the Methods section:

"Our fitting procedure first attempts to fit the low temperature resistivity to the form $\rho(T) = a_Q T^2 + \rho_0$ from base temperature to a maximum temperature that yields the highest r^2 value. A fit is considered acceptable if the r^2 value is higher than 0.95. With the exception of the quantum critical point itself and the insulating region, it is always found that a T^2 fit is appropriate at low temperature according to this criterion. The region of T-linear behavioris found above the region of T^2 behavior, except for at the quantum critical point itself where the T-linear behavior extends down to the lowest temperatures. A similar criterion($r^2 > 0.95$) is used to mark the upper boundary of T-linear behavior at each doping."

We also added the discussion about the fits to the supplementary information in new supplementary section 12 and new SI fig. 12.

2. About the discussion of the Planckian dissipation and parameter C, could the author present more details and further explanation? For example, what is the doping dependence of C near the insulating states at half-filling? For the filling with C 1 which satisfies Planckian dissipation, the possible origins shouldbe clarified.

Shown in fig. R2 below is a plot of the parameter C defined by $C = \frac{n e^2 n}{\hbar k_B m} a_L$. In our data, this parameter C is maximum at the quantum critical points. Within a quasiparticle picture, the parameter C is determined by the scattering rate that gives rise to the resistance in the sample. Near the quantum critical point, strong scattering occurs from the fluctuationsof the ordered states on either side of the critical point, with a critical divergence as one approaches the quantum critical point itself. We therefore expect on general grounds that the parameter C will be maximized at the quantum critical point, as we see in our samples. To clarify this point to the reader, we add the following sentences to the manuscript on page 6:

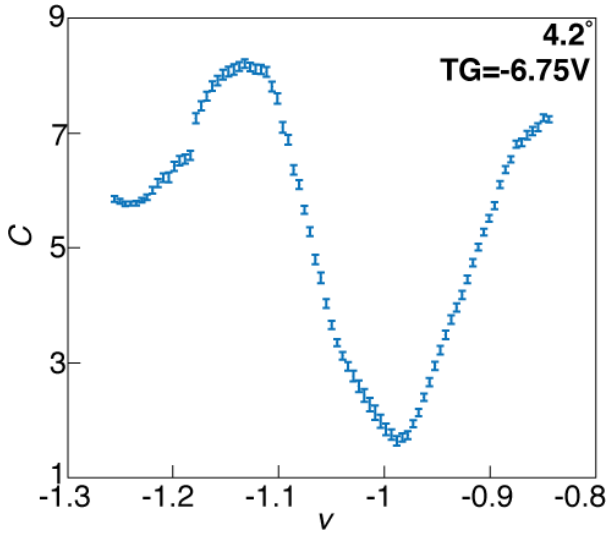

FIG. R. 2: **Planckian parameter C** defined from fits to T-linear resistivity on a devicewith twist angle 4.2° and top gate voltage TG = -6.75V and plotted against nominal density ν as defined in the text. The insulating regime extends from $\nu \approx -1.15$ to $\nu \approx -0.95$.

"... In our case, C ranges from 1 to 10, and a plot of C as a function of density is shown

in SI figure 3. C is maximized at the quantum critical point itself, as expected from thecritical divergence of the scattering rate as one approaches the quantum critical point"

We have added this discussion and figure to the supplementary information as a newsection 3.

3. Compared with the crossover from B^2 to B-linear dependence near the insulating phase of half-filling, do the authors have B-dependence of the longi- tudinal and Hall resistance near the insulator of full filling?

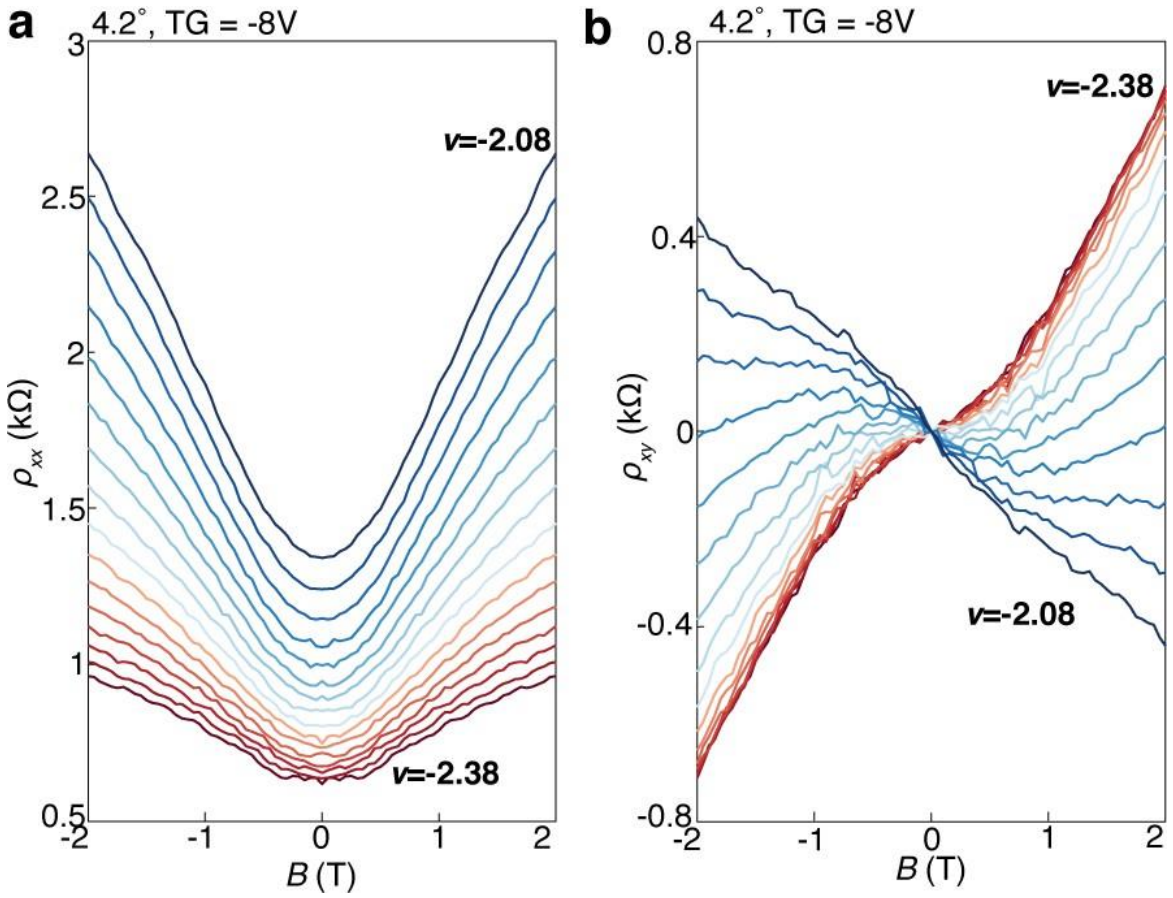

FIG. R. 3: **B-quadratic behaviour of resistivity near full-filling for the 4.2° atTG=-8V.**

We have measured the B-dependence of the longitudinal and Hall resistivity near full- filling for the 4.2° sample. These data are shown above in fig. R3. As is observed from the plots, the low field behavior remains B^2 near full filling ($\nu = -2.38$ to -2.08). The Hall

effect shows a sign change in this region, where the bottom of the first moiré subband gives way to the second moiré subband.

We have added the plots above to the supplementary information section 5 and refer toit in the main text on page 7 as follows:

"The extracted values of β as a function of ν is shown in fig. 4c, clearly showing a large increase on approaching the insulating phase, corresponding to a crossover from B^2 to B-linear behaviour. Such crossover does not occur near full-filling, where it remains withB^2 behaviour as it nears full-filling (see SI fig. 5)."

4. In the triangular-lattice Hubbard model, the insulating phase at half-fillingis predicted to be non-magnetic and is a spin-liquid of some type. Could the author present more evidence in experimental in support of this theory? For example, how do the insulating gaps around half-filling evolve with B-field?

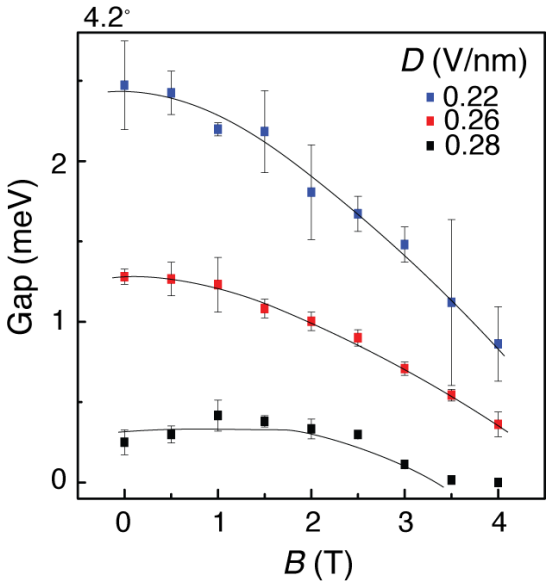

FIG. R. 4: **Gap versus perpendicular magnetic field for the 4.2° sample**

We have no direct evidence of spin liquid behavior from our experiments so far. We introduce the spin liquid as an interesting theoretically predicted phase in our systems. We have the following indirect evidence. When a perpendicular magnetic field is applied, we would expect the insulating gap to increase if the insulator is a ferromagnet, while we would expect it to decrease if it is an antiferromagnetic state or a spin liquid. Indeed in our

experiments, we see the insulating gap disappearing as we apply a magnetic field as shownin fig. R4.

We have added a plot of the measured gap versus magnetic field for different displace- ment fields for the 4.2° sample in the supplementary information section 8. We added the following sentence to the main text on pages 7-8:

" Hartree-Fock calculations show (see SI fig. 7) that for band dispersions believed rele- vant to $tWSe_2$, and for interaction strengths in a physically reasonable intermediate couplingrange, a metal-insulator-metal sequence of transitions occurs as the displacement field is varied. Theory suggests [30, 31] that at zero displacement field the insulating phase is non- magnetic, does not break a spatial symmetry, and is most probably a spin liquid of sometype. Experimentally, we observe that the insulating gaps decrease at high field, consistent with a non-magnetic state (see SI fig. 8)."

5. With υ fixed at -1, when ρ_0 is between 2-3kΩ, the gap fully opens. The authors proposed the singular interplay of impurity scattering and scattering from fluctuations of the order parameter of the insulating state. Do the author have a more detailed scenario to describe such an interplay?

In regions of the phase diagram where the sample is metallic at low temperature (re-sistance as $T \longrightarrow 0$ is not large, and resistance decreases as T is decreased), the residual resistivity is straightforwardly defined as an extrapolation of the lowest temperature re- sistance to zero temperature. The precise nature of the extrapolation (whether linear or quadratic) does not make much difference to the value of the residual resistivity obtained.In regions of the phase diagram where the sample is insulating (resistance becomes large as$T \longrightarrow 0$ and resistance increases as T is decreased at low T), we cannot use the low tem-perature resistance directly since it is in the insulating state. Instead, we extrapolate the temperature dependence of the conducting state in the higher temperature regime where theresistance is metallic-like (resistance decreases or remains constant as T is decreased) from above the metal-insulator transition down to zero temperature. We use a linear extrapola- tion, since we observe phenomenologically that the resistance as a function of temperature is linear above the metal-insulator transition. We observe that the extrapolation to T=0 of the resistance defined in this way is much higher than the residual resistance defined from the low-T metallic regions of the phase diagram. Our expectation is that disorder scattering

does not change significantly across the metal-insulator transition, since the sample itself is not changing. The implication is that the jump in the "residual resistivity" that we see is due to fluctuations of the insulating order parameter itself.

To make these statements clear to the reader, we add:

"It is important to note that in the regimes of the phase diagram where the ground stateis insulating, the residual resistivity is defined from an extrapolation of the high T resistivity defined for temperatures greater than the insulating gap (see fig. 5d) and therefore reflects the physics of fluctuations of the order parameter of the insulating state. Understanding these fluctuations is an important open theoretical problem."

6. In theory [T.Senthil, PRB, 78, 045109 (2008)] that predicts spin liquid behavior in continuous Mott transition, a divergent effective mass at the criticalpoint is also predicted. Do the author have any evidence indicating the divergenteffective mass associated with a rapid rise of ρ_0?

Unfortunately, the experimental probes available for this system do not permit us to determine the effective mass directly. We do have strong indirect evidence in the formof the collapse of the region over which T^2 behavior is observed in the resistivity, as it approaches the quantum critical point, indicating the collapse of the Fermi temperature. Along with this collapse in the temperature region over which T^2 behavior is observed, the T^2 coefficient shows a dramatic increase. In the original manuscript, we showed this behavior as a function of density, as shown in figure 3b of the main text. We also find the same behavior as a function of displacement field as we approach the metal-insulator transition from the metallic side. Shown in figure R5 below are the results of quadratic fits to resistivity at half-filling as a function of displacement field as we approach $D_{critical}$ where the metal-insulator transition occurs. We see clearly the critical enhancement in the T^2 coefficient in these data. To make this point clear to the reader, we have added the following sentence to the main text on page 7:

"At even higher values of displacement field, T^2 behaviour is recovered in the low tem-perature resistivity (also in the inset to fig. 5a) and a_Q rises by an order in magnitude as itapproaches $D_{critical}$ (see SI fig. 6), also indicating the collapse of the Fermi temperature inthe D-driven transition."

We also added the plot shown above to the supplementary information in new section 6.

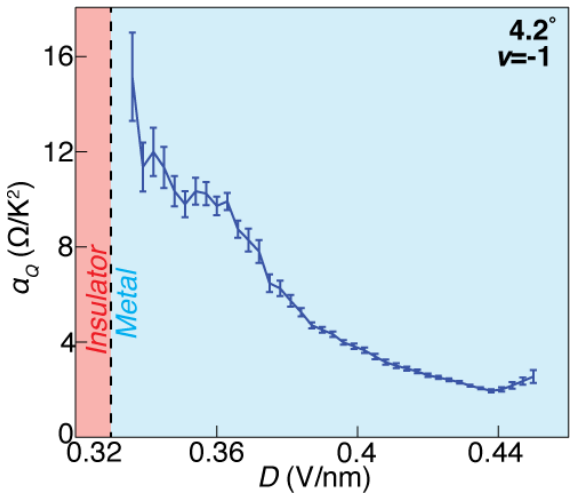

FIG. R. 5: T^2 **coefficient as a function of D at half-filling shows a strongenhancement on approaching the metal-insulator boundary.**

REFEREE 2

The manuscript by A. Ghiotto et al. reports continuous metal-insulator tran- sitions (MITs) in twisted WSe2 bilayers near the half-filling of their first moiré band. In particular, the authors observe continuous MITs by tuning either the carrier density or displacement field and explain their results in terms of doping- and bandwidth-controlled MITs, respectively. They also find non-Fermi liquid behaviors near the quantum critical regime in the resistance-temperature and magneto-resistance data.

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We thank the referee for the time taken to review our manuscript and the detailed com-

ments provided, which we address below.

Independent control of the density and displacement-field:

1. In most of the experiments (Figs. 2-4), the authors applied a constant top gate voltage while sweeping the other gate voltage. It appears to me that this would change both the doping concentration and the displacement field (purpleline in Fig. 1c). However, the authors explained these data mostly in terms of a doping-controlled MIT. While doping might have a more profound effect on the transport than the displacement field, it is not clear whether the effect of varying displacement field can be ignored entirely (particularly for Fig. 3 witha large voltage range with a significant change in D, which would change the electronic structure). I suggest the authors provide additional data at a fixed displacement field to double-check their central thesis: the continuous MIT andquantum critical regime.

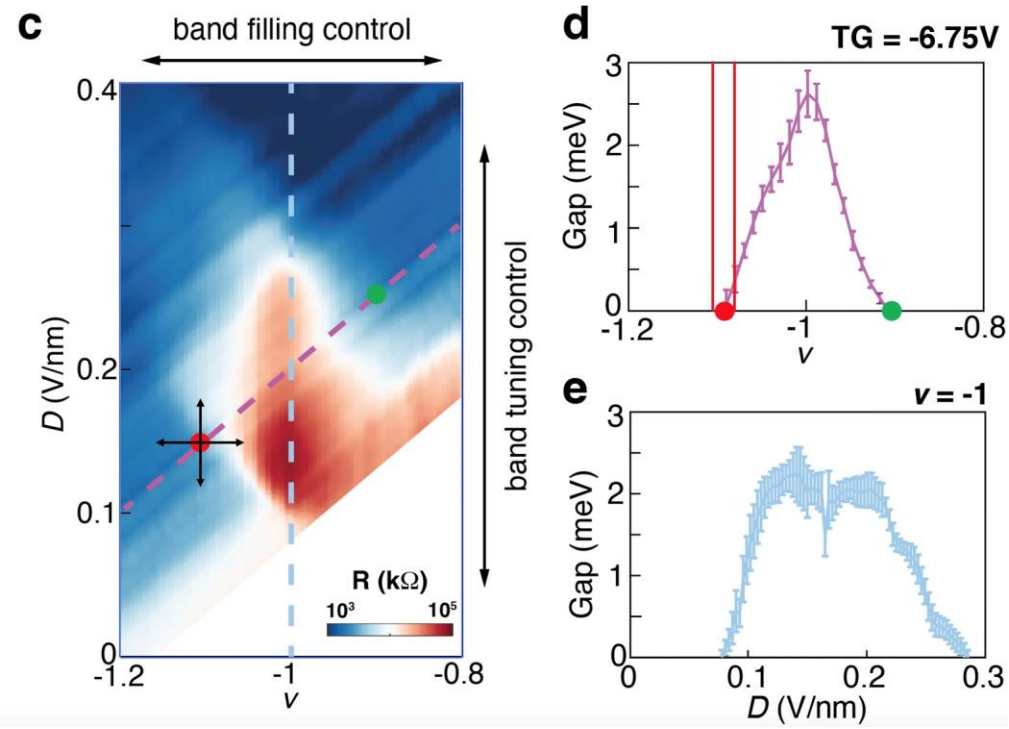

FIG. R. 6: **Annotated main text fig. 1**

In our density dependent measurements, we keep the top gate at a constant value andsweep the back gate, which avoids changes in contact resistance during the temperature sweep that is used to extract gaps and allows us to more accurately characterize the metallic behavior. The referee is indeed correct that we are simultaneously changing the doping and displacement field. However, for the trace shown in figure R6d (reproduced from figure 1 of the main text), the resistance changes across each of the quantum critical points (red and green dots in figure R6c and d) is dominated by changes in density rather than displacement field. Consider the quantum critical point shown by the red dot in fig. R6c. We can estimate the sensitivity of the resistance to the doping and displacement field in the quantum critical region near the red dot in figure R6. Moving along the density axis as shown by the horizontal black arrow in figure R6c, the resistance changes by 750 kΩ/υ. On the other hand, moving along the displacement field axis (vertical black arrow in figure R6c) gives a resistance change of 110 kΩ/D. For the insulator-metal transition, we can define a widthof the transition by two points where the temperature dependence is definitely metallic and definitely insulating respectively. This width of the transition region in figure R6dis indicated by the two red vertical lines. This transition region corresponds to a density change of 0.02υ and displacement field change of 0.01V/nm. Based on the low temperature resistance, we estimate that 94% of the change in resistance comes from density changes.

While we could perform measurements as a function of density alone by simultaneously varying both gates as the referee suggests, this procedure results in noisier data since the contact resistance is also simultaneously changing during such a procedure. We have added the following note in the main text to address this point:

”Although keeping a top gate fixed varies both displacement field and doping, at this top gate, the metal-insulator transition is dominated by changes in doping.”

2. In Fig. 1c, at low displacement field D, the sample resistance becomes highly asymmetric across nu = -1. The peak resistance seems to shift away from nu = -1 at the lowest D. Why would this happen? I wonder if this could bean artifact related to contact resistance. What are the resistance values in the cut-out white region at the lower right corner?

The referee is correct that this is an artifact related to contact resistance. Fig R7 isa schematic of the device geometry. For our measurements, contact resistance is mostly controlled by the top gate, since the contacts screen the effects of the back gate. The

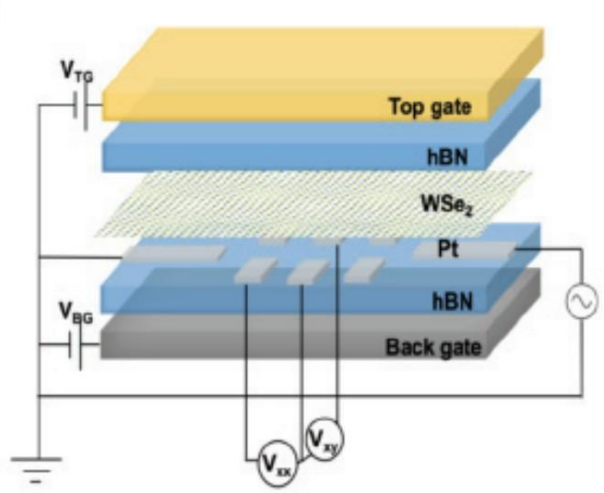

FIG. R. 7: **Device schematics** Top gate solely controls the contact doping. Channeldoping and displacement field are controlled by both top and back gates.

contact resistance is between 2 to 10kΩ and is found to be temperature independent. We perform a four-probe measurement to minimize the influence of contact resistance. We also monitor the lock-in phase of our low frequency (~17.7Hz) AC measurements to ensure thatit is close to zero, a sign of low contact resistance and metallic behavior in the tWSe_2.

At lower displacement fields, the top gate values are generally small, approaching the limit where the contacts are turned off. The asymmetry in fig 1c that the referee points out, also highlighted by the box bounded by heavy black dashed lines in fig. R8 is due to contacts nearing failure (contact resistances of order 100 kΩ), and are not indicative of a true gap in the sample itself. In the white region below the black box, contact resistance is greater than 1MΩ and the data are completely unreliable and are therefore not shown.

3. The magnetotransport data in Fig. 4 is taken from a much higher V_{TG} voltage than Figs. 2 and 3. It seems that this is due to contact resistance preventing magnetotransport measurements at low V_{TG}. Is that right? What value of D does Fig. 4 correspond to in Fig. 1c?

The referee is correct in pointing out that contact resistance limits the regions where we can obtain high quality magnetoresistance data since we need several working contacts to cleanly separate the Hall and longitudinal components. The data shown in Figure 4 is froma sample at an angle of 4.5°, different from the sample in figure 1. This data set is chosensince it has the cleanest data. Magnetoresistance data for the 4.2° sample at TG=-8V

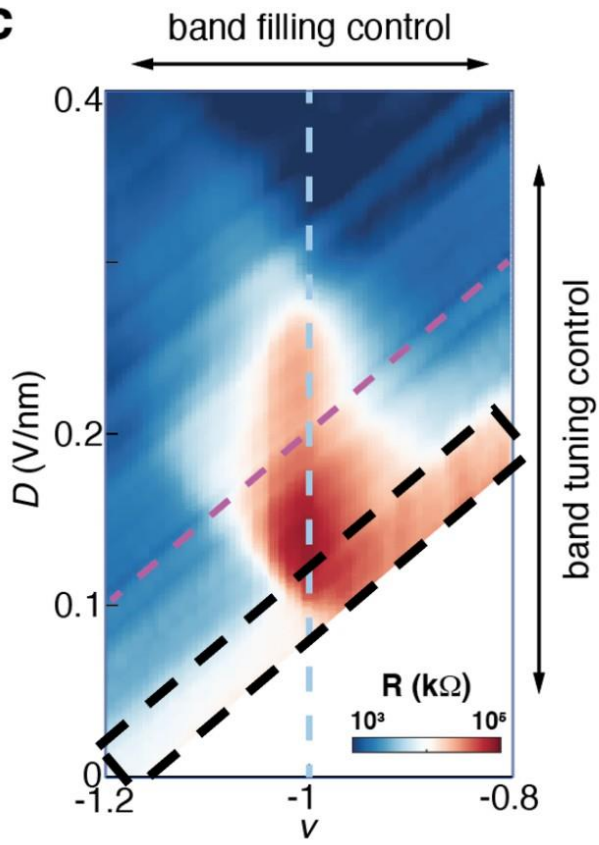

FIG. R. 8: **Contacts nearing failure** in the box bounded by heavy black dashed lines.

Reproduced from figure 1c of the main text.

(same as figs. 1 and 3) shows a similar crossover from B^2 to B– linear, and is shown in SIfig 4. To avoid confusion on the part of the reader, we have added the following sentence:

"Reliable magnetoresistance data requires multiple good contacts to the sample to sep- arate the longitudinal and Hall resistance cleanly, which is a challenge for tWSe_2 samples, especially at low doping. However, we are able to obtain reliable data at fillings above half filling at selected displacement fields. Shown in fig. 4a and b is an exemplary set of curvesof the longitudinal and Hall magnetoresistance for a sample with a twist angle of 4.5° at atop gate voltage of -19 V, for which the maximum insulating gap at half filling is ~ 0.7 meV. Similar behaviour is observed in other samples - as an example, the field data corresponding to the temperature data shown in fig. 3 is shown in SI fig. 4"

4. More importantly, because the authors relate the coefficients for temperature-

and B-field-dependence of resistance in their analysis, I think such a direct comparison might make more sense if the data are obtained under the same displacement field. I suggest adding the temperature-dependence data under the same condition as in Fig. 4.

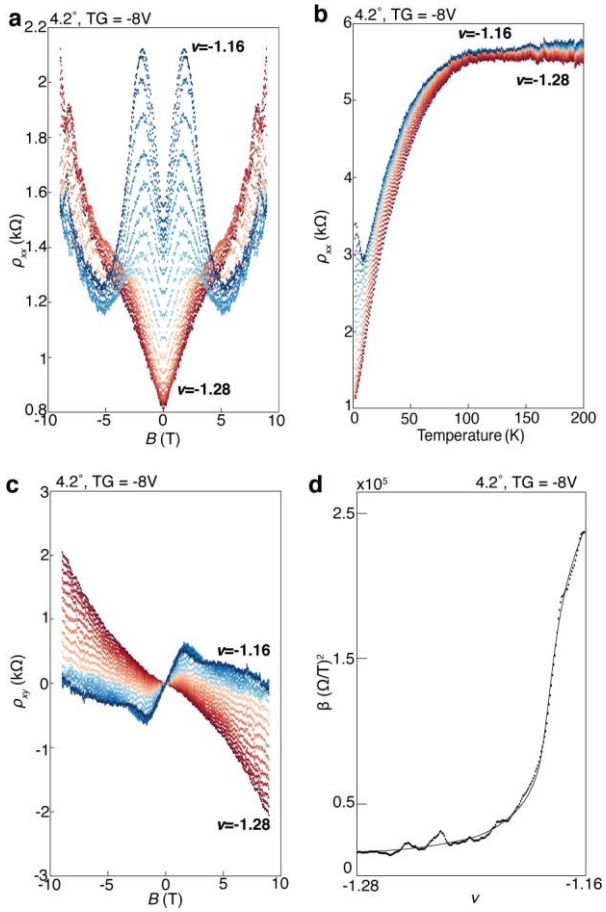

FIG. R. 9: **Comparison of magnetotransport with temperature dependencetransport. β is defined from the fit $\rho(B) \propto \sqrt{\gamma + \beta B^2}$**

We thank the referee for the suggestion. We have updated SI figure 4 to show both the temperature and field dependence in the same figure (also reproduced here in fig. R9).

Mechanism of field-driven MIT:

5. Another concern I have is regarding the MIT mechanism. The authorspropose that the system be considered a one-orbital triangular lattice Hubbard

model simulator where displacement controls the bandwidth. However, consid-ering the layer degeneracy, one might also explain the observations in termsof changing the filling ratios on the two layers (i.e., layer polarization undera displacement field, which may require consideration of two bands/orbitals). Could the proposed bandwidth control picture with one band be an oversimpli- fied one? Another related but more minor point, where is the in-plane Wannierfunction located in the moiré superlattice? It seems to me that the electrons in the bilayer could form a honeycomb lattice but not a triangular one.

The referee raises important points concerning the accepted analysis of twisted WSe_2, which relies on mapping the system to a "single band" triangular lattice Hubbard model. There are three questions. The first is a restriction to a one-band (two spin/valley) model. Available DFT calculations strongly support the idea that for experimentally accessible dis- placement fields the one-band approximation remains valid—the band shifts are not large enough to bring lower bands into the relevant energy range. The second is the effect of inter-layer charge disproportionation within the one-band two-valley model. These effects exist, and are in part responsible for the changes in band structure occurring as the displacement field is varied. However they have a negligible effect on the interaction parameters because the interlayer distance is small compared to the size of the Wannier function, so that the distribution of charge over planes does not significantly affect the interactions. The third issue is the Wannier centers. These have previously been investigated in DFT calculations (Phys. Rev. Research 2, 033087) and STM experiments (Nat. Phys. 16, 1093–1096 (2020). Both measurements unambiguously place the Wannier function centers at the triangular points.

Analysis of the temperature-dependence:

6. Because the resistance value and its temperature dependence are central to the conclusion, I think additional details shall be provided on measuring resistance and quantifying its T-dependence. For example, it would be essentialto eliminate any contribution from the contact resistance. What are the typical contact resistance values, and how do they change as a function of temperature and doping? It would be helpful to show a few representative I-V curves too.

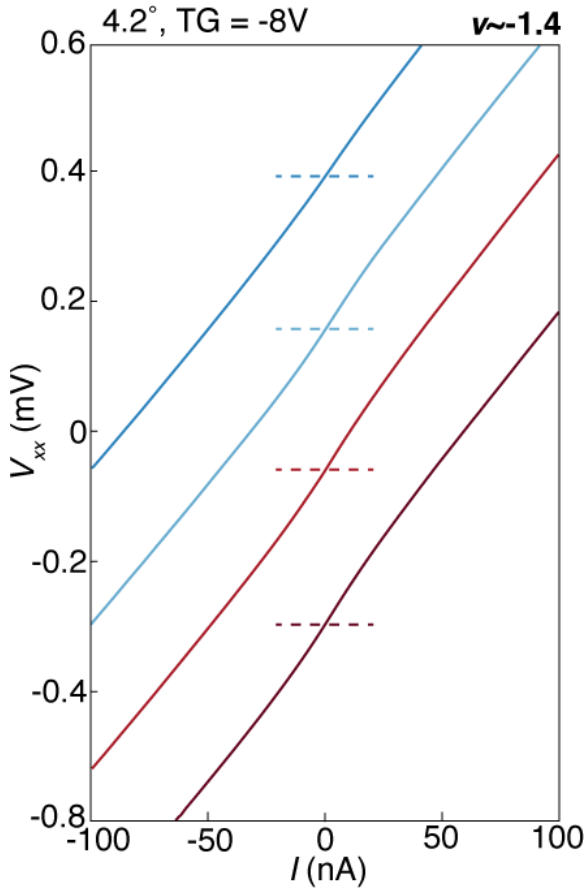

FIG. R. 10: **Representative I-V curves displaying Ohmic contacts at 200mK.Curves are offset for clarity. Dashed lines indicate V_{xx} = 0.**

Indeed, ensuring good contact quality in our samples is crucial to our work. Previous work on atomically thin WSe_2 has shown that platinum surface contacts produce Ohmic contacts at a wide range of temperatures [H. C. P. Movva et al. ACS Nano 2015, 9, 10, 10402–10410]. We find our contact resistances to be between 2 to 10kΩ for all contactsfor the data shown in figures 2-5 and that value is fairly temperature independent. As wehave pointed out above, we perform a four-probe measurement to minimize the effects of contact resistance. We also monitor the lock-in phase of our low frequency (~17.7Hz) AC measurements to ensure that it is close to zero at all times.

Regarding I-V curves - our usual procedure is to measure the resistance at a few different excitation voltages to confirm that we are in the linear response regime. In a few cases, we didmeasure I-V curves, some of which are reproduced in fig. R10. These curves are taken for a

4.2° sample at 200mK. These data are also added to the supplementary information fig. 12.

7. Besides, the resistance data shows quite complicated temperature depen- dence, so it would be helpful to explain the data fitting procedure further. For example, near the quantum critical point, the authors use both T-linear and T- quadratic dependence to fit the same data but at different temperature ranges. How are these temperature ranges chosen? If the R-T curve with T-squared dependence begins to saturate, would there always be some region where one can fit with T-linear (d^2R/dT^2 0)? For example, one might be able to get a good linear fit between 50K and 75K for nu = -1.66 in Fig. 3c.

This point raised by the referee is very similar to the first question raised by referee 1,and we would ask the referee to consider that response. The referee is correct that in going from T^2 to a saturated resistance, there will always be some region of $T-$ linear behavior. In general, we consider a minimum of 10 K with $r^2 > 0.95$ above the T^2 region in order todefine a region of $T-$ linear behavior.

8. Similarly, it would be great if the authors can specify how to choose the proper temperature range to extract activation energy for insulating states.

As we go through the metal-insulator transition as a function of temperature, the resis- tance goes through a minimum and starts to increase at lower temperature. We find that in the temperature range when the sample just enters into the insulating phase, the resistance displays activated behavior over 1-2 decades in resistance (see figure R11 below for a typical curve, also in SI section 2). This region in temperature is used to define the activation gap. The temperature range is always chosen so that the the Arrhenius fit has $r^2 > 0.9$. At even lower temperatures, the temperature dependence falls off from the exponential form, likely due to disorder.

We have added the following sentence to the SI section 2 to describe our fitting procedure:"The linear fit is defined over a region chosen such that r^2 is maximized (always greater than 0.9)."

9. Is there a systematic procedure to determine which types of fitting (T vs. T^2) to use? For example, the R-T curve might start to deviate from linear near

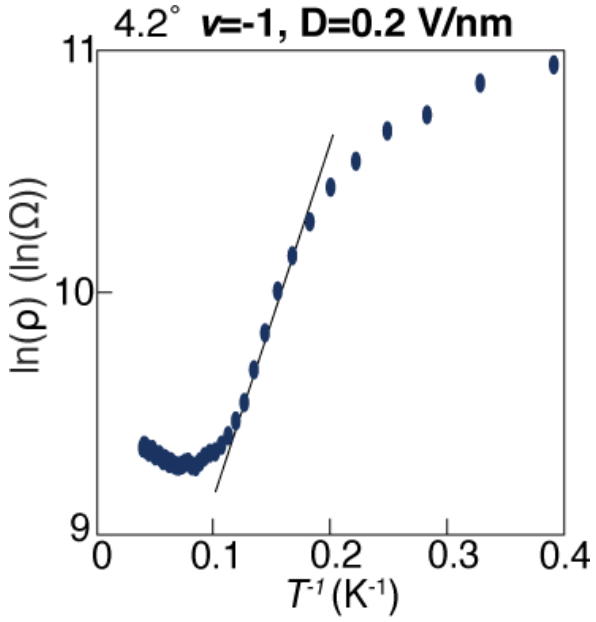

FIG. R. 11: **Example of Arrhenius fit to gaps**

base temperature (or if the temperature were lowered further) for nv=-0.94 in Fig. 3c.

The fitting procedure we chose, described in the answer to the first question of referee1, always attempts to perform a T^2 fit at low temperature. The upper cutoff in temperature is determined by the goodness of fit, as described above. The $T-$ linear behavior is found above this T^2 cutoff, and the region of $T-$ linear behavior is again determined by the goodness of fit. Experimentally, we always find that the resistance has a T^2 form at sufficiently low temperature, apart from the insulating region and the quantum critical points themselves. It is certainly possible that the temperature dependence of the resistance could deviate from linear at the quantum critical points at temperature below the lowest temperatures of our measurement. Even in this case, that would not change the basic conclusion that there is a quantum critical "fan" where the $T-$ linear behavior gives way to T^2 behavior as one goes further into the metallic phase, and that the coefficient of T^2 shows a critical enhancement as one approaches the quantum critical point.

10. Could the authors explain how error bars are determined in the lowerpanel of Fig. 3b? A bit surprisingly, the error bars do not change much, espe-

cially for the T-linear curve, even when the system approaches the Fermi-liquid region.

Our error bars are determined as the standard error (root mean square) of the fits. Since we impose a minimum of 95% confidence interval for our linear fits (see above), the error bars in our linear fits are indeed small at all doping levels.

11. I wonder if the authors could estimate the level of disorder in the sys- tem and comment on the possible disorder effects in explaining the continu- ous doping-controlled MIT? Do authors observe any spatial inhomogeneity or anisotropy in their measurements?

Most of our samples display very homogeneous behavior across contacts, as also measured by piezoresponse force microscopy measurements prior to full device fabrication [McGilly, L. J. et al., Nature Nanotechnology 15, 580–584(2020)]. Occasionally, we do find that differentpairs of contacts do see some twist angle inhomogeneity in a single device. For example, shown below in figure R12 is a sample where two different pairs of contacts display slightly different doping dependent behaviour, corresponding to slightly different twist angles for thetwo pairs of contacts (4.1° and 4.2°). Such behavior is rare in our samples.

We have added this discussion to new supplementary section 10.

Other minor points:

12. I am confused by the 2D color plots in Fig. 2: they show the phase diagram down to 0 K, but experiments were only down to 1.5 K. Is that correct? A more accurate way to represent the data would be to leave white spaces below 1.5 K.

The referee is correct. We have replaced the label to be more accurate in our plots.

13. In Fig. 3, why does not the insulating state appear at υ = -1?

At that displacement field value for that top gate (0.31V/nm at υ=-1), the insulating state is very weak and, although we do observe an upturn in resistivity as well as the effectsof quantum criticality at the boundaries, we are unable to extract a clear gap from transport. As the referee points out, the insulating state does appear away from υ = 1. This could

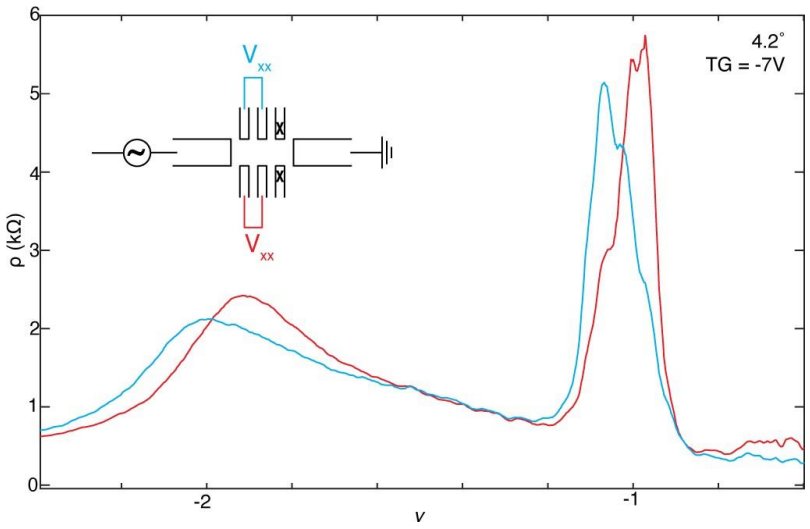

FIG. R. 12: Sample homogeneity. Resistivity curves at 1.6K from two different pairs ofleads. Crossed out leads indicated high contact resistance. Hall bar channel is 3μm wideand 7μm long.

possibly be because we are near the metal-insulator boundary as a function of displacement field, and the insulating phase may start to ”track” the van Hove singularity which for thisdisplacement field occurs at $\nu = -1.23$. We do not have definitive evidence of this currently.

We have substituted the sentence: “At this top gate voltage, the sample displays a smaller gap than shown in figure 2 but is otherwise similar.” with “At this top gate voltage, although we see an upturn in resistivity near half-filling, we cannot extract a gap from Arrhenius fits. We also note that the correlated insulator shifted away from half-filling and speculate that at such weak interactions, it would be directly influenced by the position ofthe vHS.”

14. Some important details are missing for readers to understand and repro-duce the results, which would be very helpful to include. For example, what is the thickness of the top and bottom hBN? This will be important to convert gate voltage into a displacement field. How is the twist angle determined? It would be helpful to show a microscope image of the device for readers to relativepositions of gate and source-drain (since a gate may activate the source-drain incertain device structures).

We thank the referee for pointing out the missing information. We have expanded the method section to include more fabrication details and doping, displacement field and twist angle analysis (see below). We have also included a microscope image of the 4.2° sample in the supplementary information section 9 (shown below in fig. R13).

"**Sample fabrication**

Sample preparation is discussed in detail in Wang et al. [13]. Twisted WSe_2 samples were prepared using the dry stamp and 'tear-and-stack' techniques [38,39]. tWSe_2 is first picked up using the top hBN flake and then placed on top of pre-patterned Cr/Pt (2nm/20nm) contacts [40] on the bottom hBN. A combination of metal and/or graphite is used as topand bottom gates as shown in fig. 1a (see SI fig. 9 for device image). From atomic force microscopy measurements, we determine the thicknesses of the hBN: 4.2° sample (top 38nm,bottom 32nm) and 4.5° sample (top 52.1nm, bottom 31.3nm).

Doping, displacement field and twist angle determination

As described in Wang et al. [13], the geometric capacitance of the top and bottom gates can be extracted via Landau levels from $n = \nu \frac{eB}{h}$. We measure two Landau fans in the samples listed, one projecting to the band edge, from which we determine the intrinsic doping n_0 and the other projecting to full-filling of the moiré unit cell, from which we determine the full filling density n_s. By assuming a linear gate response of the system, doping and displacement field can be obtained via $n = \frac{C_{BG}BG + C_{TG}TG}{e} + n_0$ and $D = \frac{C_{BG}BG - C_{TG}TG}{2\epsilon_0}$.

Since the contact doping is solely controlled by the top gate voltage, only displacement fieldin the +z direction is accessible.

DFT calculations predict a two-fold degeneracy of the top moiré valence band, for which we find experimental evidence in the Landau fans from the band edge that are also two-fold degenerate at low magnetic fields [13]. The twist angle is determined from $n_s = \frac{-2}{A}$ and $A = \frac{a^2\sqrt{3}}{4(1-cos\theta)}$, where a = 0.328nm [25] is the WSe_2 lattice constant."

15. Currently, all the experiments are under the positive displacement field, and in principle, the device's behavior shall be the same for negative displace- ment. Is there any asymmetric gate response since the device structure is not symmetric (because Pt contacts WSe_2 from the bottom)? It would also be great if the authors could define the positive direction of the D.

As the referee points out, in principle it is expected that the phenomenology should be

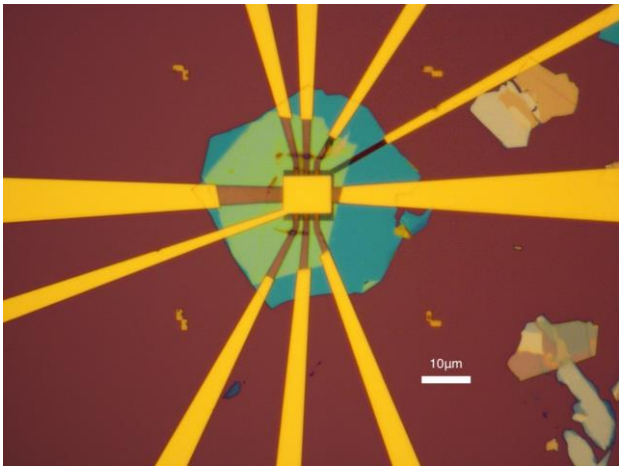

FIG. R. 13: **Optical microscope image of the 4.2˚ sample.**

the same whether D is positive or negative since we have a homobilayer system. However, this is not something we can verify experimentally due to contact resistance limitations. Given that the top gate almost solely controls the contacts, we are limited in it's rangeto negative values where the contact resistance is small. Therefore, for our measurements, displacement field is pointing upwards in the sample. We have added the following sentence to the methods section to clarify this point: "Since the contact doping is solely controlledby the top gate voltage, only displacement field in the +z direction is accessible."

16. In Fig. 5a, the third R-T curve from the top shows decreasing resistance with increasing T even at high temperature and cross with the fourth curve. Is it real?

It is not real. The guide to the eye in fig, 5a is misleading and we have updated it, thanks to the referee's observation. The last five data points have resistivity values of: 6,177Ω (14K), 6,247Ω (15K), 6,250Ω (16K), 6,338Ω (17K), 6,360Ω (18K).

REFEREE 3

The authors study the metal-insulator transition in twisted WSe2 bilayers. These systems are attracting great attention for their emergent correlated statesarising from the existence of nearly flat bands. The authors show that by con-

trolling the density and dispersion electrically, a continuous correlated-metallicphase transition is found. Importantly, the authors find that at the phase bound- ary, linear T resistivity behavior emerges, a paradigmatic hallmark of critical behavior in correlated systems. In contrast, deep in the metallic regime, the au-thors find the usual T^2 behavior of conventional metals. The authors highlightthat these findings show that this system is a very well-suited platform to study correlated behavior on the triangular lattice.

I found their manuscript greatly interesting and an outstanding demonstra- tion of the high-quality correlated physics that can be observed in twisted WSe2systems. Their experiments are fully consistent with previous findings and pro-vide compelling evidence of strongly correlated phenomena in twisted dichalco-genides beyond reported correlated regimes. Overall, I believe that their re- sults are correct and that they report exciting new phenomena in twisted van der Waals materials. For this reason, I think that their manuscript will be of tremendous interest for the communities of twisted van der Waals materials and correlated physics, and therefore I recommend publication of their work inNature.

We thank the referee for the time to read our manuscript and the kind words about the results.

Besides my positive recommendation for publication in Nature, I would also like to bring up a few minor points that the authors may want to consider inthe final version of their manuscript: - The authors claim that this system is "a one-orbital triangular lattice Hubbard model simulator", yet I would say that this is probably an oversimplification. First, the existence of spin-orbit couplingprobably creates spin-dependent hoppings, the electronic structure may involvesecond neighbor hoppings, and the electronic interactions quite probably involve non-local interactions. Therefore, I would suggest that the authors rewrite that sentence as "a one-orbital correlated triangular model simulator."

As suggested by the referee, we have replaced "a one-orbital triangular lattice Hubbard model simulator" with "a one-orbital correlated triangular model simulator."

- As the authors noted, linear-T behavior can also appear due to electron- phonon coupling (as suggested in twisted graphene bilayers in Ref. [33]). The authors argue that in their experiment, such electron-phonon mechanism can be discarded, yet from my understanding of their argument, although their argument is suggestive, strictly speaking, it may be partially circumstantial. Therefore, I would suggest that the authors rewrite the statement in a milder way.

We have substituted "This strong correlation between the T-linear behaviour and the magnitude of the correlated gap, as well as the presence of T^2 resistivity far from the quan-tum critical points, indicate that in this temperature regime the resistance is dominated by strong electronic correlations rather than by electron-phonon scattering." with "This strong correlation between the T-linear behaviour and the magnitude of the correlated gap, as well as the presence of T^2 resistivity far from the quantum critical points, indicate that in this temperature regime the resistance is well described by contributions from strong electronic correlations. Electron-phonon scattering may play a role in the observed temperature de-

pendence, but a crossover from $T-$ linear to T^2 behaviour would not be expected solely from electron-phonon scattering."

- The authors further claim that the correlated state is "most likely a spin liquid". While theoretical arguments may put that as a possibility, I would say that there is no strong experimental evidence that the system is a spin-liquid. Therefore, this may be an overstatement, and I would suggest that the authors mention the spin liquid just as a hypothetical possibility rather than the most likely scenario.

As suggested by the referee, we have substituted "most likely a spin liquid" with "poten- tially a spin liquid" in the manuscript.

To summarize, I think that the current experiment provides compelling ex- perimental evidence of further correlated behavior in twisted dichalcogenide bilayers, a topic of critical interest for the community, and therefore I strongly recommend the publication of their manuscript in Nature.

FINAL NOTE: The following sentence was added to the manuscript: "Furthermore, theoretical predictions on the doping-tuned metal-insulator transition indicate a wealth of interaction-driven metallic phases in this particular system [37]."

Reviewer Reports on the First Revision:

Referee #1:
Remarks to the Author:
The authors have addressed my questions and comments.
But I have two more questions based on the author's response 2 to the referee2. In Fig.1c, that the peak resistance shifting away from nu=-1 at lowest D is attributed to an artifact related to the contact resistance. Does the artifact still exist at higher temperature? The authors also present data from the other one sample (~4.5º). Could the author display the plot of low temperature resistance versus D and band filling for the sample with ~4.5º?

Referee #2:
Remarks to the Author:
The authors provided additional analysis and data, which addressed my comments well. I am happy to recommend the paper's publication in Nature.

I only have two minor optional suggestions the authors might consider:

1) It could be useful to explain the limit of contact resistance and the white cutout region in the caption of Fig. 1c, particularly for readers outside of the field and non-experimentalists.

2) It would also be worthwhile to cite the two references included in the rebuttal letter when discussing the triangular lattice (Phys. Rev. Research 2, 033087 and Nat. Phys. 16, 1093–1096 (2020)), since it is quite important for the spin liquid hypothesis. Many existing pieces of the literature suggest a honeycomb lattice in TMD homobilayers.

Referee #3:
Remarks to the Author:
The authors have addressed the different comments of my first report satisfactorily. As stated in my previous report, I believe that their results are novel and of high interest for the correlated van der Waals community, and therefore I strongly recommend publication in Nature.

Author Rebuttals to First Revision:

REFEREE 1

The authors have addressed my questions and comments. But I have two

more questions based on the author's response 2 to the referee 2. In Fig.1c, that the peak resistance shifting away from $\nu = -1$ at lowest D is attributed to an artifact related to the contact resistance. Does the artifact still exist at higher temperature? The authors also present data from the other one sample(4.5º). Could the author display the plot of low temperature resistance versus D and band filling for the sample with 4.5º?

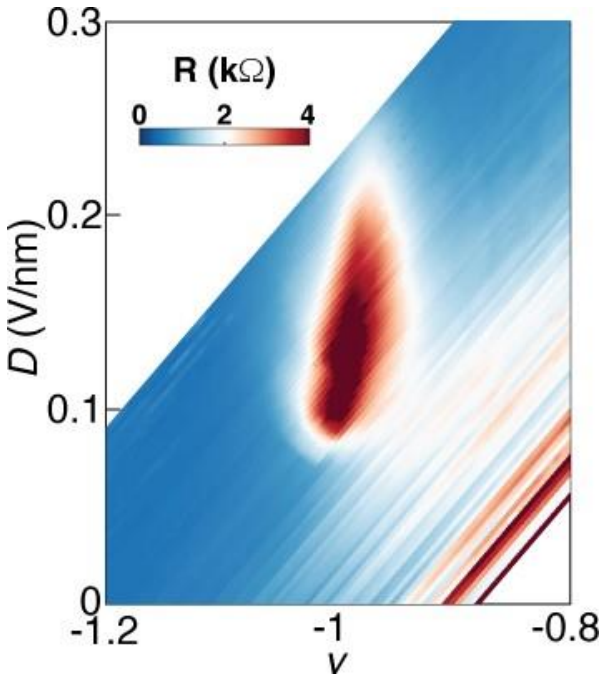

FIG. R. 1: Displacement field versus doping map at 200mK of the 4.5˚ sample.

As stated previously, we find our contact resistances to be temperature independent. Such asymmetry in doping when the contacts are nearing failure therefore persists at higher temperatures. The overall resistivity in that region increases with increasing temperature, signaling metallic behavior.

Shown in fig. R1 is the resistivity map in displacement field and doping for the 4.5˚

sample. For that particular sample, the contacts are better behaved at lower displacement fields and the asymmetry in resistivity is not so prominent.

REFEREE 2

The authors provided additional analysis and data, which addressed my com-ments well. I am happy to recommend the paper's publication in Nature.

I only have two minor optional suggestions the authors might consider:

1) It could be useful to explain the limit of contact resistance and the white cutout region in the caption of Fig. 1c, particularly for readers outside of the field and non-experimentalists.

2) It would also be worthwhile to cite the two references included in the rebuttal letter when discussing the triangular lattice (Phys. Rev. Research 2, 033087 and Nat. Phys. 16, 1093–1096 (2020)), since it is quite important for the spin liquid hypothesis. Many existing pieces of the literature suggest a honeycomb lattice in TMD homobilayers.

We thank the referee for the constructive review and the positive recommendation of our manuscript.

1) Following the advice, we have added the following sentence to figure 1 caption: "Sincethe range of top gate values is limited in our experiments (see Methods), a higher overall resistance is observed at lower displacement fields because of larger contact resistances."

2) We have added the suggested references to page 3 in the following sentence: "Due to large spin-orbit coupling and layer hybridization, tWSe_2 is effectively a one-orbital correlatedtriangular model simulator (fig. 1b), where a correlated insulating phase has been previouslyobserved at half-filling [13, 24, 25]"