

# Correlated Insulating States in Slow Dirac Fermions on a Honeycomb Moiré Superlattice

Corresponding Author: Professor Ziliang Ye

**This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.**

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The paper presents an exciton-polaron spectroscopy study of strongly interacting states in a WS–tMoSe<sub>2</sub> homobilayer. The doping dependence of the 2S absorption reveals multiple features attributed to various correlated states, including magnetic ones. The conclusions are supported by temperature and magnetic field dependencies and are further corroborated by DFT and Hartree-Fock calculations.

I found the paper highly interesting, with novel results that, in my view, meet the high standards of Nature Communications. I am inclined to recommend publication after the authors address the following points:

1. The manuscript states that 1S excitonic states are less sensitive to the external environment (i.e., electron states in tMoSe<sub>2</sub>) compared to 2S states. Additional clarification would be helpful: do they exhibit similar qualitative behavior with reduced magnitude, or is there a fundamental difference in their doping response? Any quantitative comparison would strengthen this point.
2. The formation of attractive and repulsive peaks is typically interpreted in terms of their spectral weight, energy splitting, and linewidth (e.g., Phys. Rev. X 13, 011029 (2023) and related theoretical works). Including the doping dependence of these parameters (perhaps in the Supplementary Material) would be valuable for theorists working in this area.
3. The manuscript reports oscillatory features in the doping dependence at fractional filling factors. However, the corresponding discussion is minimal and lacks clarity. A more thorough explanation would improve the paper's impact.
4. The authors interpret the quarter-filled state as a generalized Wigner crystal. In contrast, the formation of an electronic Wigner crystal was identified in *Nature* **595**, 53 (2021) by the appearance of an additional small peak, blueshifted from the repulsive exciton-polaron branch—this feature appears absent in the present data. A more detailed discussion in light of these prior observations would be beneficial.
5. Some plots use gate voltage as the horizontal axis, while others use the filling factor. Switching between these representations was not straightforward, and readers may find this confusing.
6. The authors discuss an interaction-induced excitonic insulator state characterized by a gap at the Dirac point. Is exciton-polaron behavior expected to be similar in doped conventional insulators versus excitonic insulators? While the temperature dependence is thoroughly discussed, a complementary discussion of doping dependence in this context would strengthen the interpretation.

Reviewer #2

(Remarks to the Author)

D. Yang et al., reported a series of optical spectroscopic studies on honeycomb moire lattice systems in twisted bilayer MoSe<sub>2</sub> (t-MoSe<sub>2</sub>). The authors observed integer and fractional fillings of correlated insulating states in the hole-doping regime of the t-MoSe<sub>2</sub> employing 2s Rydberg excitons as sensors in a WS<sub>2</sub> layer placed near the t-MoSe<sub>2</sub> layers. Such t-MoSe<sub>2</sub> systems host Dirac type band structures whose Fermi velocity is much larger than the one in graphene.

The experimental results presented in the paper appear sufficiently reliable, and the discussions are well structured, contributing to a storyline that initially comes across as coherent. However, while the paper presents an interesting argument, it seems to place too much emphasis on a single interpretation, without adequately considering alternative perspectives. This may limit the depth of the overall analysis, especially given the likely complexity of the observations.

In the following, I summarize the main concerns I have regarding the paper.

1. Could the authors comment on the case of twisted bilayer graphene? Doesn't twisted bilayer graphene also form slow Dirac fermions if the twist angle is bit misaligned from the magic angle?
2. It was not clear to me what is special about Mott and correlated insulating states of Dirac fermions. Are there some new phenomena expected in such systems?
3. The authors mention "Due to the type-II band alignment between WS<sub>2</sub> and MoSe<sub>2</sub>". Is this type-II band alignment? Previous studies by Mak and Shan showed that it is type-I band alignment without electric field.
4. Could the authors explain the origin of the blueshift of the 2s exciton resonance in the sensor layer at the correlated insulating state of the target layer? It cannot be explained from the exciton screening picture (this origin rather gives redshift).
5. Similarly, in the page 4, the authors mentioned that "we observed a series of blueshifts in the IAP peak" and "These behaviors indicate a reduced screening". Simple screening reduction picture rather results in redshift.
6. In Figure 2b, the assignment of filling zero ( $\nu=0$ ) is clearly wrong. At the  $\nu=0$ , where screening of the 2s excitons in the sensor layer is weakest, the oscillator strength of the 2s excitons should be strongest. However, the assignment of  $\nu=0$  is misaligned from the strongest oscillator strength condition.
7. By taking account of the problem above, I think the authors cannot assign fractional fillings of  $\nu=-1/6$ ,  $-1/4$ ,  $-1/2$ ,  $-2/3$ , and  $-5/6$  precisely due to the lack of linearity in the gate voltage vs fillings  $\nu$ , as it is obvious from the misalignment of  $\nu=0$  above.
8. In the page 6, the authors mentioned that "Although both scenarios open a charge gap at the Dirac point, the Mott insulator phase breaks the chiral symmetry." Why is this the case? Filling single charge at each site of honeycomb lattice does not necessarily open a gap at the Dirac point. Could the clarify what they mean as "chiral symmetry" and why this is broken in this situation?
9. Given that the 1s exciton Bohr radius of MoSe<sub>2</sub> is typically in the order of 1nm, how it can form X1+ trion states where hole and excitons are separated more than 5nm? If such charge transfer trion exists, there should be for example interlayer attractive polaron for the 1s exciton state of WS<sub>2</sub> when t-MoSe<sub>2</sub> is hole doped, since the separation between these layers are in the order of 1nm which is much closer than the moire periodicity of 5nm. Also, if X1+ is this charge transfer trion at  $\nu=1$ , why isn't there usual moire trion at  $\nu=1$ ? Even if there were tiny oscillator strength for such charge transfer trion X1+, I cannot believe that such a state shows the strong binding energy of 17meV. I disagree with this charge transfer trion interpretation, and this suggests that the subsequent storyline using X1+ breaks down.
10. In the page 6, the authors mentioned that "According to our DFT calculations, the excitons and holes in both X1+ and X2+ are located at the XM or MX moire site". Where do the authors provide these DFT calculations?
11. In the page 6 and 7, the authors mentioned that "As the doping level increases further, the holes at a doubly occupied site can no longer couple to any exciton". Why is this the case? Even if there are two holes, wouldn't one of the holes which has the opposite spin to the hole of the exciton form a trion state?
12. In the page 7, the authors mentioned that "We attribute the MCD signal in X2+ to the intervalley exchange interaction between the spin-singlet and spin-triplet trions". Why is this the case? Doesn't the author need to consider valley Zeeman splitting of excitons at  $\pm K$  points and energy shift due to the Fermi polaron formation by considering the spin polarization at the  $\Gamma$  point?

### Reviewer #3

#### (Remarks to the Author)

The authors report the observation of correlated electronic states in twisted MoSe<sub>2</sub> homobilayer heterostructures, which realize a honeycomb moiré superlattice. In these systems, the top two moiré bands originate from the folded valence band at the  $\Gamma$  valley of the monolayer Brillouin zone. Due to strong hybridization and weak spin-orbit coupling in the  $\Gamma$  valley, the moiré potential minima are symmetrically distributed between the MX and XM sites, making it similar to a graphene honeycomb lattice. The various electronic states are optically probed using Rydberg excitons in a WS<sub>2</sub> sensing layer and by moiré trions in the MoSe<sub>2</sub> bilayer. The authors identify the insulating state at  $\nu=-1$  as a sublattice-polarized electronic crystal arising from the nearest neighbor Coulomb repulsion, and the state at  $\nu=-2$  as a Mott insulator arising from the on-site Coulomb repulsion. Additional insulating states are identified at fractional fillings.

Overall, this work provides valuable insights into correlated electron physics and light-matter interactions in two-dimensional materials with moiré superlattices. The manuscript is generally clear, and the key conclusions are well-supported by both experimental data and theoretical modeling. I therefore recommend publication after the authors address the following comments.

1. Why the  $\nu=0$  is not at  $V_{tg}=-1V$  where you observe an abrupt 18-meV blue shift in Figure 2? If the reason is non-linear gate-dependent doping, the assignment for the fractional filling might be wrong. Why the signature of the  $\nu=-1/3$  is weaker than that of  $\nu=-1/6$ ?
2. Does all the gate voltage dependence (in all the figures) follow  $V_{bg}=1.15V_{tg}$ ? If so, I recommend defining it in text (rather than in Figure 2 caption) and using it instead of  $V_{tg}$ .
3. Regarding the X+1 trion in the MoSe<sub>2</sub>, although it might be a charge transfer trion, I don't see why a tightly bound trion cannot form at the same doping condition. Intuitively, I would expect similar oscillator strength for the two species at  $\nu=-1$ .

What is the reason for the abrupt jump from one to the other in Figure 4?

4. It was previously reported on light-induced spin depolarization in MoSe<sub>2</sub>. Did you verify that there is no spin depolarization in the magnetic circular dichroism experiments? [Nature, 623(7987):509–513, 2023 ; Phys. Rev. Lett. 128, 127402 (2022)].

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

I have carefully read both responses to my comments and the revised manuscript. The authors have clearly addressed all points I raised and have made a substantial effort, including the addition of several new figures. Overall, the experimental data appear very solid, and the interpretations are coherent. While some aspects of the work have been questioned and debated by other referees, I find that these issues are difficult to disentangle at the present stage, given that polaronic effects involving strongly correlated electrons remain a developing field. This study will undoubtedly stimulate further theoretical research. Its impact clearly meets the expectations for Nature Communications and is likely to attract significant attention. Therefore, I recommend publication in its present form.

Reviewer #2

(Remarks to the Author)

I appreciate the authors' detailed reply and their effort to deepen the discussion. Except for the interpretation of X1<sup>+</sup> and X2<sup>+</sup> states, I am convinced by their explanation. I understand that the authors now attribute X1<sup>+</sup> and X2<sup>+</sup> as a kind of shakeup process. However, I think such processes result in blueshift from the original exciton resonances since additional energy to excite those states is required. Even if we assume very small inter-site trion binding energy which perhaps could account for net redshift, X2<sup>+</sup> dominated by on-site e-e Coulomb repulsion energy scale should be on the blue side compared to X1<sup>+</sup> dominated by nearest neighbor site e-e Coulomb repulsion energy scale. I recommend the authors formulate those relations with equations like the authors described for the case of Rydberg state energy blueshift in Response 10. Also, I wonder where the on-site trion (or attractive polaron) state appears which is missing in this new interpretation.

Reviewer #3

(Remarks to the Author)

I have read the response and the revised manuscript. Two important concerns remain:

1. The authors did not address the identification of fractional filling, except for removing the  $\nu=1/3$ . Although the assigned filling is linear with the applied gate voltage, the doping seems non-linear in the low-doping regime.

Regarding the  $\nu=1/3$  state, I still expect that it will show a stronger optical signature than the  $\nu=1/6$  state. What is the reason for its absence?

An additional remark about filling assignments is that in Fig.2a,b, the  $\nu=1$  is at a gate voltage of  $V_g \sim 2.3V$ , while Fig. 4a shows that the  $\nu=1$  is at a gate voltage of  $V_g \sim 2.7V$ . This difference accounts for about 1/3 of the difference between  $\nu=1$  and  $\nu=2$  of Fig. 2.

2. I am not convinced regarding the new interpretation of the X<sup>+(2)</sup> state. With the large  $U_0/t$  ratio and on-site repulsion energy ( $U_0 \sim 25meV$  according to the numerical results, and  $U_0 \sim 100-200meV$  in the last paragraph of the introduction), why do the authors expect excitations in low temperatures?

The authors should address these concerns before I can recommend publication.

Version 2:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

I thank the authors for their detailed response. The explanation that the difference in binding energies leads to X2<sup>+</sup> appearing at a lower energy than X1<sup>+</sup> is convincing. My concern has been satisfactorily addressed, and I recommend publication of the manuscript

Reviewer #3

(Remarks to the Author)

I have carefully considered the authors' reply to my questions. Although I still find this work interesting and important, I am not fully convinced by the interpretation of the  $X^{+2}$  origin.

Version 3:

Reviewer comments:

Reviewer #3

(Remarks to the Author)

I thank the authors for their careful response and for revising the text regarding the origin of  $X^{+2}$ . In my view, the revised, more tentative wording provides an appropriately cautious interpretation of the assignment and adequately addresses my concern.

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## Point-by-Point Response:

Reviewer # 1 (Remarks to the Author):

The paper presents an exciton-polaron spectroscopy study of strongly interacting states in a WS<sub>2</sub>-tMoSe<sub>2</sub> homobilayer. The doping dependence of the 2s absorption reveals multiple features attributed to various correlated states, including magnetic ones. The conclusions are supported by temperature and magnetic field dependencies and are further corroborated by DFT and Hartree-Fock calculations.

I found the paper highly interesting, with novel results that, in my view, meet the high standards of Nature Communications. I am inclined to recommend publication after the authors address the following points:

We thank the reviewer for the positive comments as well as the detailed suggestions that have helped us improve our manuscript. In the following, we respond to the questions in a point-by-point way. The changes in the manuscript have been highlighted accordingly.

1. The manuscript states that 1s excitonic states are less sensitive to the external environment (i.e., electron states in tMoSe<sub>2</sub>) compared to 2s states. Additional clarification would be helpful: do they exhibit similar qualitative behavior with reduced magnitude, or is there a fundamental difference in their doping response? Any quantitative comparison would strengthen this point.

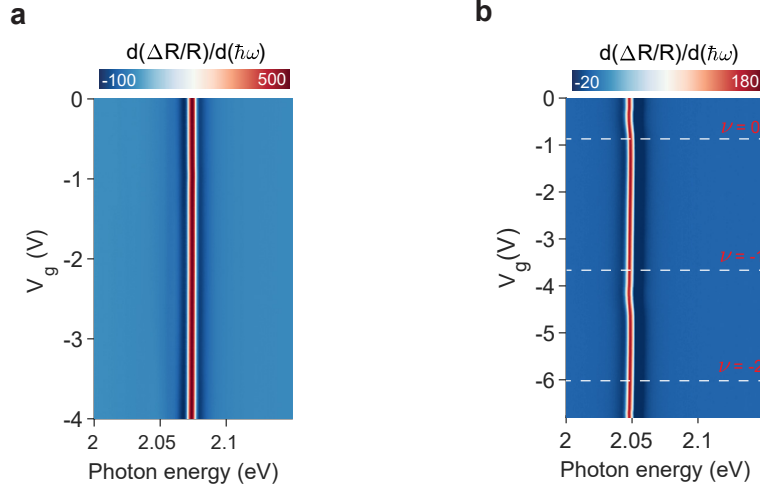
### Response 1

Qualitatively, the 1s state in the sensor layer should behave similarly as the 2s state in response to the changes in the external dielectric environment. Nevertheless, due to its smaller radius ( $\sim 0.5$  nm), 1s exciton's coupling with the dielectric environment is significantly weaker. The screening-induced peak shift in the 1s state is therefore much smaller in magnitude and difficult to resolve in the steady state reflection contrast spectra.

We include the 1s exciton spectra as a function of gate voltage ( $V_g$ ) below for samples D1 and D2. The 1s exciton features in both samples show minimal changes versus doping. In sample D2, two small spectral kinks are observed near the band edge and around  $\nu = -1$  (Figure R1(b)), but overall, the 1s response remains significantly weaker compared to that

of the 2s exciton.

These data have been added to the Supplementary Information Figure S9, and we have revised the relevant discussion in the main text on Page 4, **"In contrast, the 1s exciton remains largely unaffected due to its smaller radius (Figure S8)."**

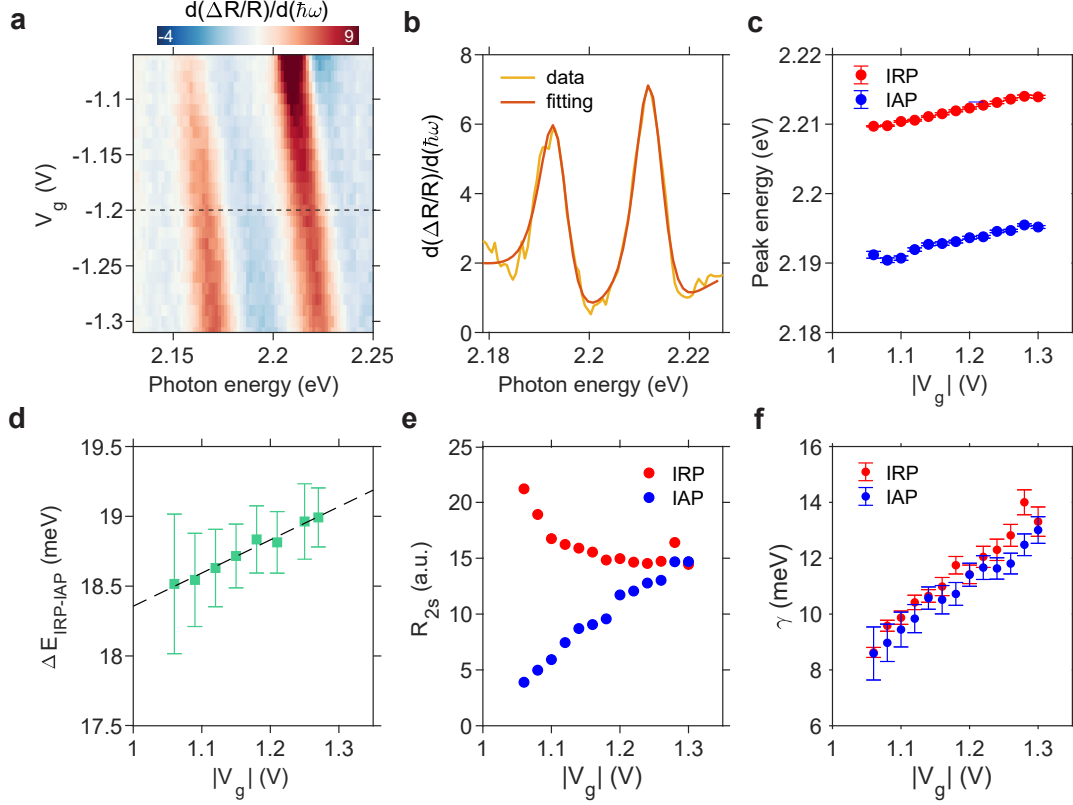


**Figure R1: a,b** The spectra of 1s exciton in the WS<sub>2</sub> sensor layer as a function of doping  $V_g$  in device D1 (**a**) and device D2 (**b**). The doping density is controlled by the effective gate voltage  $V_g = \frac{V_{tg} + (C_b/C_t)V_{bg}}{2}$ , where  $C_b$  and  $C_t$  are the geometrical capacitances of the top and bottom hBN.

2. The formation of attractive and repulsive peaks is typically interpreted in terms of their spectral weight, energy splitting, and linewidth (e.g., Phys. Rev. X 13, 011029 (2023) and related theoretical works). Including the doping dependence of these parameters (perhaps in the Supplementary Material) would be valuable for theorists working in this area.

## Response 2

We thank the reviewer for the suggestion. Since the attractive and repulsive interlayer Fermi polarons (*IAP* and *IRP*) are mostly observed in the dilute doping region ( $-1.31 \text{ V} < V_g < -1.05 \text{ V}$ ), we focus on extracting the parameters from this part of the spectra.



**Figure R2:** **a** Enlarged contour plot of 2s interlayer Fermi polarons (*IAP* and *IRP*) in the dilute doping region. **b.** A linecut of fitting results at  $V_g = -1.2$  V. **c.** Extracted peak energies of *IAP* (blue) and *IRP* (red). **d.** Energy difference between *IRP* and *IAP* as a function of gate voltage. **e.** The oscillator strength ( $R_{2s}$ ) transfers from *IRP* to *IAP* as the doping increases. **f.** Extracted linewidth ( $\gamma$ ) increases with doping for both *IRP* and *IAP*.

Here we employ a multi-Lorentzian model to fit the reflection contrast spectra near the *IAP* and *IRP* resonances as shown in Figure R2(a). Specifically, we fit the data using the derivative form:

$$\frac{d}{d\hbar\omega} \frac{\Delta R}{R} = \frac{d}{d\hbar\omega} \left( \frac{A \cos(\alpha) \gamma^2 / 2}{(\hbar\omega - \hbar\omega_0)^2 + \gamma^2 / 4} + \frac{A \sin(\alpha) \gamma (\hbar\omega - \hbar\omega_0)}{(\hbar\omega - \hbar\omega_0)^2 + \gamma^2 / 4} \right) + const$$

$A$  is the oscillator strength,  $\hbar\omega_0$  is the resonance energy, and  $\gamma$  is the full linewidth at half-maximum.  $\alpha$  is a phase factor that depends on the photon energy and the gate voltage. A representative linecut and its corresponding fit are shown in Figure R2(b). The extracted parameters, such as peak energies, oscillator strengths, and linewidths, are summarized in Figures R2(c-f).

As shown in Figure R2(c), both *IAP* and *IRP* resonances exhibit a blueshift with increas-

ing doping. The energy separation between *IRP* and *IAP* increases linearly with gate voltage [Figure R2(d)], from which we estimate the intrinsic *IAP* binding energy to be approximately 18.4(8) meV. The oscillator strength gradually transfers from *IRP* to the lower-energy branch *IAP* as doping increases, consistent with the expected behavior of Fermi polarons. Furthermore, within this doping window, the linewidths of both *IAP* and *IRP* increase with carrier density, as shown in Figure R2(f).

The above observations agree with the general expectations for Fermi polarons. Following the reviewer's suggestion, we have included the discussion on *IAP* and *IRP* in the Supplementary Information and revised the manuscript on page 4, Paragraph 1, **"As the carrier density increases, the *IRP* gradually transfers its oscillator strength to the *IAP* (Supplementary Information S6 and Figure S9)"**

3. The manuscript reports oscillatory features in the doping dependence at fractional filling factors. However, the corresponding discussion is minimal and lacks clarity. A more thorough explanation would improve the paper's impact.

### Response 3

We thank the reviewer for raising this important point. The peaks in the oscillator strength of the 2s exciton arise from the reduced screening associated with the insulating states at different fillings. The peaks are blue-shifted because the quasiparticle bandgap increases more than the binding energy. (See the detailed explanation in Response 10.) When additional holes are introduced into these charge-ordered states, the system undergoes a quantum melting process, during which the charge gap diminishes. As a result, the mobile charges screen the 2s exciton and reduce its oscillator strength. Such a screening effect continues as the system transits into the next insulating state. The oscillatory behavior in the 2s oscillator strength originates from the system transforming between electronic crystal and fluid states as the doping increases.

We have included the above discussion in the main text, on page 4, Section 3, **"The peaks in  $R_{2s}$  correspond to charge incompressible states. When additional holes are introduced into these charge-order states, the system undergoes a quantum melting process..."**

4. The authors interpret the quarter-filled state as a generalized Wigner crystal. In contrast, the formation of an electronic Wigner crystal was identified in "Nature 595, 53 (2021)" by the appearance of an additional small peak, blueshifted from the repulsive exciton-polaron branch—this feature appears absent in the present data. A more detailed discussion in light of these prior observations would be beneficial.

#### Response 4

We thank the reviewer for the suggestion. We have carefully examined our data near the repulsive Fermi polaron feature in the t-MoSe<sub>2</sub> bilayer, but do not observe any clear evidence related to the Umklapp scattering. This suggests that the signal, if present, is below our current sensitivity.

A potential explanation lies in the doping dependence of the umklapp resonance. According to the aforementioned study, the feature is expected to appear at very low doping levels (less than  $3.0 \times 10^{11} \text{ cm}^{-2}$ ) and becomes significantly broadened and eventually unresolvable as doping increases. In our device D1, the quarter-filled state corresponds to a carrier density of approximately  $1.15 \times 10^{12} \text{ cm}^{-2}$ , substantially higher than the literature. Any potential Umklapp resonance signal is therefore likely to be broadened into the background and becomes unresolvable in the reflectance contrast spectra.

We have included the discussion on the potential umklapp resonance in the main text, page 5, paragraph 1, **"We do not observe any Umklapp excitation resonance states near the 1s resonance of t-MoSe<sub>2</sub>, likely due to their relatively high doping density ( $\sim 10^{12} \text{ cm}^{-2}$ )."**

5. Some plots use gate voltage as the horizontal axis, while others use the filling factor. Switching between these representations was not straightforward, and readers may find this confusing.

#### Response 5

We have updated all the primary horizontal axes to the effective gate voltage ( $V_g = \frac{V_{tg} + (C_b/C_t)V_{bg}}{2}$ ). To aid the readability of Figure 2c, we use a dual horizontal axis - both the filling factor and gate voltage are labeled.

6. The authors discuss an interaction-induced excitonic insulator state characterized by a gap at the Dirac point. Is exciton-polaron behavior expected to be similar in doped conventional insulators versus excitonic insulators? While the temperature dependence is thoroughly discussed, a complementary discussion of doping dependence in this context would strengthen the interpretation.

### Response 6

We thank the reviewer for the question. We first need to clarify that we observe a Mott insulator induced by electronic interactions, rather than an excitonic insulator. Second, after carefully analyzing the data and considering the comments of reviewer 1 and 2, we have developed a new explanation for the two types of exciton polarons ( $X_1^+$  and  $X_2^+$ ) observed in the t-MoSe2 bilayer. In short, we think they are indeed different from the normal Fermi polarons in doped monolayers. We tentatively attribute  $X_1^+$  to an exciton coupled to charge excitations across the charge-transfer gap, whereas  $X_2^+$  corresponds to an exciton dressed with charge excitations across the Mott gap. We have discussed more details of such a picture in **Response 15**.

### Reviewer # 2 (Remarks to the Author):

D. Yang et al., reported a series of optical spectroscopic studies on honeycomb moire lattice systems in twisted bilayer MoSe2 (t-MoSe2). The authors observed integer and fractional fillings of correlated insulating states in the hole-doping regime of the t-MoSe2 employing 2s Rydberg excitons as sensors in a WS2 layer placed near the t-MoSe2 layers. Such t-MoSe2 systems host Dirac type band structures whose Fermi velocity is much larger than the one in graphene. The experimental results presented in the paper appear sufficiently reliable, and the discussions are well structured, contributing to a storyline that initially comes across as coherent. However, while the paper presents an interesting argument, it seems to place too much emphasis on a single interpretation, without adequately considering alternative perspectives. This may limit the depth of the overall analysis, especially given the likely complexity of the observations.

We thank the reviewer for the insightful comments and detailed suggestions that have helped us improve our manuscript. We have extensively revised our conclusion and included an

alternative picture to explain the results. In the following, we respond to the following concerns in a point-by-point way.

In the following, I summarize the main concerns I have regarding the paper.

1. Could the authors comment on the case of twisted bilayer graphene? Doesn't twisted bilayer graphene also form slow Dirac fermions if the twist angle is bit misaligned from the magic angle?

### Response 7

We thank the reviewer for raising this important point. We agree that twisted bilayer graphene (TBG) can indeed host slow Dirac fermions because of the interference of Dirac cones from the two layers, particularly when the twist angle is slightly detuned from the magic angle. The Fermi velocity is also renormalized to be a very small value.

On the other hand, there is an important distinction in lattice symmetry: TBG forms a triangular moiré superlattice, where doped electrons or holes are most concentrated in the AA region. The twisted MoSe<sub>2</sub> hosts a honeycomb superlattice. The doped holes are mainly located in the MX (AB) and XM (BA) regions. It is therefore more complicated in TBG since it hosts both slow Dirac fermions and heavy fermions. Interestingly, the Gross-Neveu phase transition has recently also been proposed in the twisted bilayer graphene system. (*Phys. Rev. B* 112, L041109 (2025), *Nature Communications* 16, 7176 (2025)).

Following the reviewer's suggestion, we have added comments on the slow Dirac Fermions in twisted bilayer graphene on Page 2, Paragraph 1, **"A prominent strategy to enhance correlations is to reduce the Fermi velocity, as demonstrated in twisted bilayer graphene, where the interference of two Dirac cones leads to significant flattening of the bands."**

2. It was not clear to me what is special about Mott and correlated insulating states of Dirac fermions. Are there some new phenomena expected in such systems?

### Response 8

We thank the reviewer for letting us further elaborate the implications in our findings. As

explained in the introduction, compared to the previously studied semiconductor moiré systems where most correlated phenomena happen in the  $K$  valleys, the folding around the band maximum in our system happens around the  $\Gamma$  point where spin-orbital coupling is zero, thus providing an ideal platform for studying slow Dirac Fermions with degenerate spins. Although we haven't found experimental evidences, a range of intriguing new phenomena have been predicted theoretically in such a system, including exotic magnetic orders ("Physical Review B 107, 205142 (2023)"; "Communications Physics 5, 1 (2022)"), single- and double-stripe phases ("Physical Review B 103, 155142 (2021)"), and spin-polarized superconductivity (arXiv:2503.05863). We hope our experimental demonstration will open new opportunities to future discoveries in this direction.

On the other hand, the semi-metal to Mott insulator transition of Dirac Fermions is also of fundamental interest. By tuning the twist angle, the transition belongs to the Gross-Neveu universality class, a critical behavior of high-energy physics that is rare in condensed matter systems, unlike Ising or Landau universality classes. The  $\Gamma$ -valley moiré materials thus provide a promising solid-state platform for simulating such a unique critical behavior. (The Cornell group recently published an experimental work that is related and parallel to ours on this regard (L. Ma, et al, Nature Materials, 1-7 (2025)) Finally, tuning the  $U/t$  ratio in such artificial graphene-like systems opens new opportunities to revisit some interesting quantum electrodynamic phenomena on a different energy scale, such as the Klein tunneling effect (Nature Physics 2, 620–625 (2006)).

To fully address the reviewer's concern, we have emphasized the novelty and importance of studying correlated phenomena in the  $\Gamma$  valley moiré system in the revised manuscript, on page 3 Paragraph 1, **"In the large  $U_0/t$  limit, correlated states with novel magnetic orders, single- and double-stripe phases, as well as unconventional superconductivity have been theoretically predicted. Tuning the  $U_0/t$  ratio in such an artificial graphene-like systems also opens new opportunities to revisit interesting quantum electrodynamic phenomena such as Klein tunneling on a different energy scale."**

3. The authors mention "Due to the type-II band alignment between WS2 and MoSe2,". Is this type-II band alignment? Previous studies by Mak and Shan showed that it is type-I

band alignment without electric field.

## Response 9

We thank the reviewer for pointing out this important issue. We are aware that the Cornell group has reported a type-I band alignment between monolayer WS<sub>2</sub> and monolayer MoSe<sub>2</sub>. However, the situation in our system appears to be more complex due to the interlayer hybridization between the MoSe<sub>2</sub> layers. Experimentally, we do observe both cases where electrons can be doped in either WS<sub>2</sub> or twisted MoSe<sub>2</sub>, depending on the twist angle, while holes are always doped in the twisted MoSe<sub>2</sub> layer.

A detailed investigation of the conduction band alignment in this system is beyond the scope of this study. To avoid potential confusion or misinterpretation, we have removed the mention of "type-II band alignment" from the manuscript on Page 3, Paragraph 2, **"Since the valence band of WS<sub>2</sub> is much lower than that of MoSe<sub>2</sub>..."** . **The band structure in Figure 1 has also been modified.**

4. Could the authors explain the blueshift of the 2s exciton resonance in the sensor layer at the correlated insulating state of the target layer? It cannot be explained from the exciton screening picture (this origin rather gives redshift).

## Response 10

We thank the reviewer for this thoughtful question. We understand the reviewer's concern that a reduction in dielectric screening leads to a decrease in the effective dielectric constant ( $\epsilon$ ), which in turn increases the exciton binding energy. From this perspective, one might expect the exciton resonance to redshift when a correlated insulating state forms.

However, the exciton resonance energy is determined by the difference between the quasi-particle bandgap and the exciton binding energy:

$$E_{ex} = E_g - E_{binding}$$

When the effective dielectric constant decreases, both the exciton binding energy and the quasiparticle bandgap are renormalized due to long-range Coulomb interactions. Specifically, the reduction in screening increases both the quasiparticle bandgap ( $E_g$ ) and the exciton

binding energy ( $E_{\text{binding}}$ ). In the case of the 2s exciton, the increase in  $E_g$  ( $\Delta E_g$ ) exceeds the increase in exciton binding energy ( $\Delta E_{\text{binding}}$ ). As a result, the overall 2s exciton resonance exhibits a **blueshift**, rather than a redshift, when the sample enters a correlated insulating state.

This behavior is consistent with previous observations reported by the Cornell group as well as the results in monolayer TMDs (*Nature* 587.7833 (2020), *Nature Communications*, 8, 15251 (2017) ). Since this question overlaps with the concern raised in **Question 5**, we have addressed both questions together in this response. To fully address the reviewer’s concern, we have revised the discussion on Page 4, Paragraph 2, **"These behaviors are a result of reduced screening together with renormalized quasi-particle band gap, indicating the emergence of insulating states at these doping levels."**

5. Similarly, in the page 4, the authors mentioned that “we observed a series of blueshifts in the IAP peak” and “These behaviors indicate a reduced screening”. Simple screening reduction picture rather results in redshift.

#### Response 11

The discussion has been included in Response 10.

6. In Figure 2b, the assignment of filling zero ( $\nu=0$ ) is clearly wrong. At the  $\nu=0$ , where screening of the 2s excitons in the sensor layer is weakest, the oscillator strength of the 2s excitons should be strongest. However, the assignment of  $\nu=0$  is misaligned from the strongest oscillator strength condition.

#### Response 12

We appreciate the reviewer’s comment - the assignment of zero filling is always more difficult than it seems. First of all, we agree that the gate voltage  $V_g = -1.0$  V, where the 2s oscillator strength is strongest, should correspond to the charge-neutral condition. However, at this specific gate voltage, the Fermi level still lies in the quasiparticle gap of t-MoSe<sub>2</sub>. Due to the existence of in-gap quantum capacitance, a certain amount of extra gate voltage is needed to have the Fermi level touch the valence band edge.

Although the exact voltage corresponding to the band edge is hard to determine directly, we

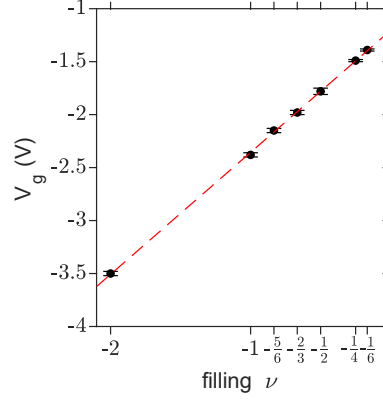
experimentally deduce it by assuming a linear gate dependence with an equal filling spacing between  $\nu = -2, -1, 0$  states, which leads us to label the  $\nu = 0$  a bit off from  $-1.0$  V in the original submission. The practice of assigning zero filling to the voltage corresponding to the strongest 2s oscillator strength and to a voltage range near it is both recorded in the literature (eg. *Nature*, 587, 214–218 (2020), *Nature Physics*, 18, 1214–1220 (2022) and *Nature Photonics*, 19, 1219–1224 (2025)). To avoid confusion, we have labeled  $\nu = 0$  as a voltage range because the exact position is not critical for our results. **We have revised Figure 2 in the new manuscript to address the reviewer’s concern.**

7. By taking account of the problem above, I think the authors cannot assign fractional fillings of  $\nu=-1/6, -1/4, -1/2, -2/3$ , and  $-5/6$  precisely due to the lack of linearity in the gate voltage vs fillings  $\nu$ , as it is obvious from the misalignment of  $\nu=0$  above.

### Response 13

We thank the reviewer for the comments. As explained above, the assignment of these fractional fillings does not rely on the exact position of  $\nu = 0$ . Due to the complexity of assigning  $\nu = 0$ , we use the voltage difference between  $\nu = -1$  and  $\nu = -2$  to calibrate those fractional fillings. In Figure R3, we plot the gate voltage  $V_g$  of the corresponding insulating states versus the assigned filling number  $\nu$ . The uncertainty is derived from the FWHM of the insulating states versus gate voltage. Within the uncertainty, the gate voltages of all insulating states lie on a straight line. Although such a linear model may not strictly hold at extremely low doping and may cause some misassignments for peaks such as  $\nu = -1/6$ , based on the good correlation presented in Figure R3, we are confident that most fractional peaks should be assigned correctly. To be on the cautious side, we have removed the assignments for the  $\nu = -1/3$  peak.

We have added Figure R3 in the Extended Figure 8 to support our assignment of the fractional filling numbers. We also add a comment on the uncertainty of the insulating state when the doping is low on page 5, Paragraph 2, **"This nonlinear gating effect may cause uncertainty on the filling number assigned at low dopings."**



**Figure.R3:** The plot shows the gate voltage  $V_g$  as a function of the filling factor  $\nu$ . The black dots represent experimental data, extracted from the peak oscillator strength of the 2s exciton, with error bars indicating the measurement uncertainty. The red dashed line is a linear fit to all observed states.

8. In the page 6, the authors mentioned that “Although both scenarios open a charge gap at the Dirac point, the Mott insulator phase breaks the chiral symmetry.” Why is this the case? Filling single charge at each site of honeycomb lattice does not necessarily open a gap at the Dirac point. Could the clarify what they mean as “chiral symmetry” and why this is broken in this situation?

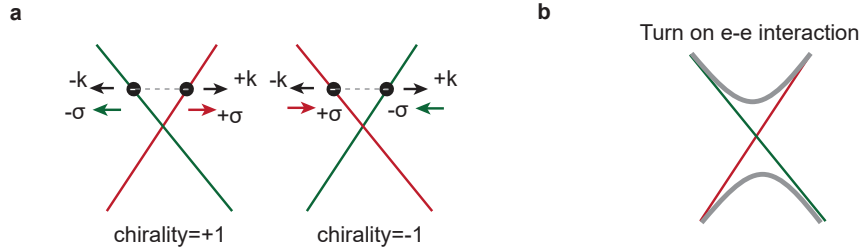
### Response 14

We appreciate the reviewer’s comments. The single-particle band structure of the  $\Gamma$  valley bands in twisted  $\text{MoSe}_2$  closely resembles that of monolayer graphene. In monolayer graphene, hopping between two equivalent sublattices (A and B) gives rise to two cosine-like energy bands, which intersect at the corners of the Brillouin zone, forming Dirac cones. The two branches of the linear energy dispersion near the  $K$  point originate from the symmetric ( $\frac{1}{\sqrt{2}}(|A\rangle + |B\rangle)$ ) and antisymmetric ( $\frac{1}{\sqrt{2}}(|A\rangle - |B\rangle)$ ) combinations of wavefunctions at the A and B sublattices. As a result, one could define an in-plane pseudospin vector, as shown in Figure R4(a). Under such a convention, a state with a certain momentum always has a pseudospin either parallel or antiparallel to the momentum (depending on the valley), which allows one to define it as a chirality index - the projection of the sublattice pseudospin onto the momentum direction.

The chiral symmetry arises from the Dirac Hamiltonian - in other words, the states with

well-defined chirality are eigenstates of the Hamiltonian. It has been theoretically proposed that such a symmetry can be broken by the many-body interaction between the charges on different sublattices ("Physica Scripta, 2012, T146"). In the limit of a Mott insulator, the strong Coulomb interaction between the doped electrons can cause hybridization between the two cosine-like bands, thus opening a charge gap at the Dirac point. On the other hand, the Dirac point can also be gapped by directly breaking the AB sublattice symmetry. For example, if one sublattice has a different chemical potential compared to the other sublattice, as in boron nitride or transition metal dichalcogenides, a gap can also appear. The CDW phase is another example, where two holes occupy the same sublattice, leaving the other empty. We think the charge gap at the Dirac point in our system is generated through the Mott mechanism.

To address the reviewer's question, we have expanded the discussion on the chiral symmetry breaking of the Mott insulator formed at the Dirac point on Page 6, Paragraph 1 **"In the Mott phase, the Coulomb interactions between the holes doped at the two sublattices break the chiral symmetry in the Dirac Hamiltonian and generate a many-body charge gap at the Dirac point..."**



**Figure.R4:** **a.** Dirac cones at  $\pm K$  valley with opposite chiralities. **b.** Interaction-induced direct coupling between different sublattices(pseudospin) opens a mass gap at the Dirac point.

9. Given that the 1s exciton Bohr radius of MoSe2 is typically in the order of 1nm, how it can form X1+ trion states where hole and excitons are separated more than 5nm? If such charge transfer trion exists, there should be for example interlayer attractive polaron for the 1s exciton state of WS2 when t-MoSe2 is hole doped, since the separation between these layers are in the order of 1nm which is much closer than the moire periodicity of 5nm. Also,

if  $X1+$  is this charge transfer trion at  $\nu=1$ , why isn't there usual moire trion at  $\nu=1$ ? Even if there were tiny oscillator strength for such charge transfer trion  $X1+$ , I cannot believe that such a state shows the strong binding energy of 17 meV. I disagree with this charge transfer trion interpretation, and this suggests that the subsequent storyline using  $X1+$  breaks down.

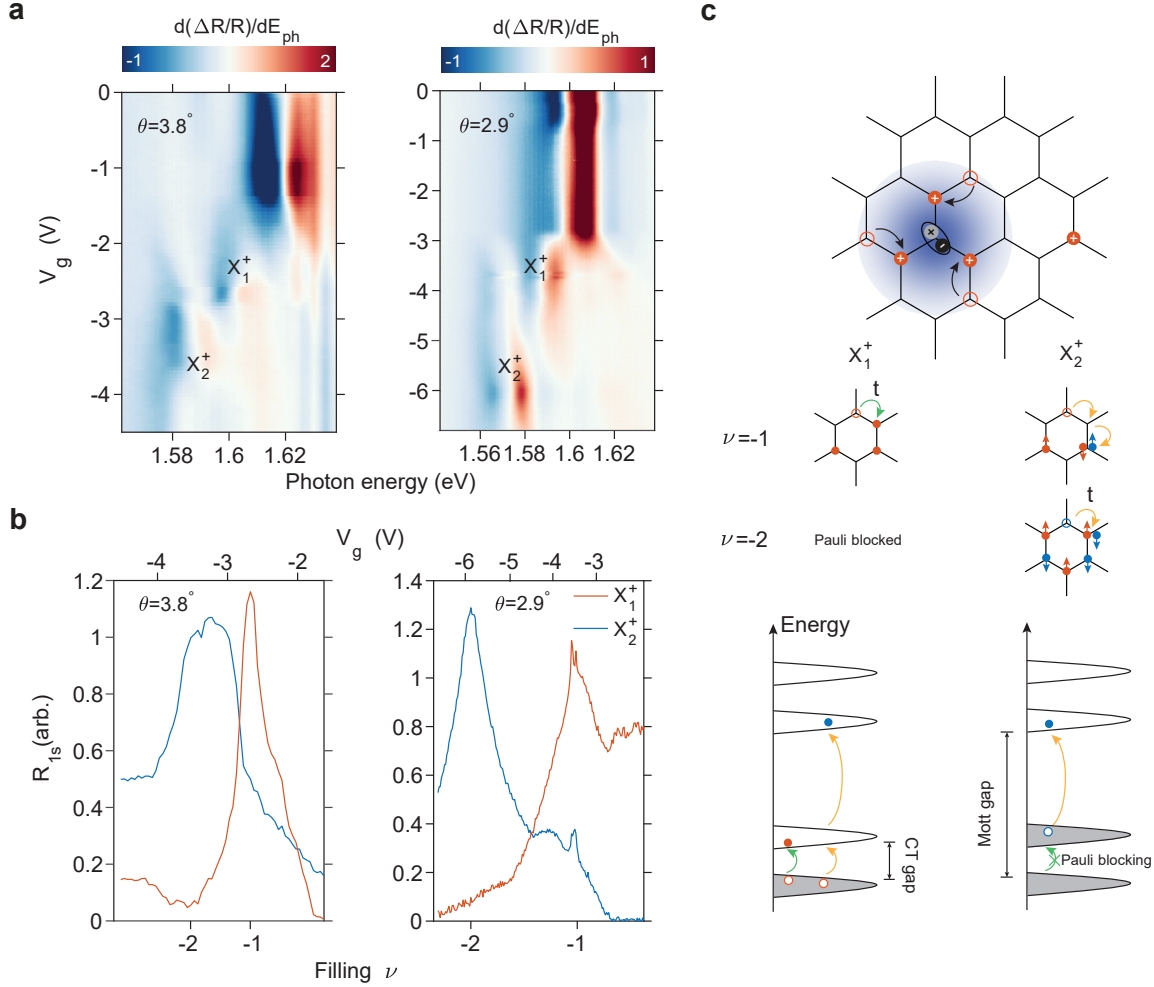
## Response 15

We thank the reviewer for the critical comments, which help us better understand the results regarding the interpretation of  $X_1^+$  and  $X_2^+$  features. Following the UT Austin group's work (arXiv:2407.17025), we assigned the two emerging peaks as the intra- and inter-cell trions. However, upon closer inspection of our doping dependent data and in light of the reviewers' feedback, we find that such a picture cannot fully explain our observations, especially regarding their doping dependence. In the revised manuscript, we propose an alternative exciton-polaron-based picture, where  $X_1^+$  is an exciton dressed by a polarization associated with the charge excitation across the charge-transfer (CT) gap, whereas  $X_2^+$  arises from an exciton coupled to the charge excitation across the Mott gap.

In a doped monolayer  $\text{MoSe}_2$ , excitons couple with a Fermi sea to form attractive and repulsive exciton-Fermi polarons. In correlated insulating states, there is no well-defined Fermi sea, but a similar polaron can be formed by dressing excitons with charge excitations associated with inter-site hopping (Figure R5c). In the low-doping regime near  $\nu = -1$ , the dominant charge excitation involves nearest-neighbor hopping across the charge-transfer (CT) gap between MX and XM sites, as indicated by the green arrow in Figure R5c. The ensemble-averaged amplitude of this first-order hopping process is maximized precisely at  $\nu = -1$ , when every MX or XM site hosts one hole; adding more holes suppresses the amplitude via Pauli blocking. The doping dependence of  $X_1^+$  matches such an expectation, which is why we attribute it to excitons hybridized with the above CT-gap excitations.

A second class of excitations involves hopping across the Mott gap to create a holon-doublon pair. In this case, a doped hole must hop twice to form a doubly occupied state at an MX (or XM) site (yellow arrow in Figure R5c), a second-order process that is small but finite at low doping—consistent with the weak oscillator strength of  $X_2^+$  at  $\nu = -1$ . The holon-doublon creation probability increases toward  $\nu = -2$ , at which doped holes only need to hop once

between the nearest MX or XM sites, and subsequently decreases when some sites become doubly occupied, consistent with the maximum oscillator strength of  $X_2^+$  at half-filling.



**Figure.R5: a.** The reflection contrast spectrum as a function of gate voltage near the A exciton resonance of t-MoSe<sub>2</sub> in device D1 (left) and device D2 (right).  $X_2^+$  and  $X_1^+$  denote the low-energy and high-energy exciton-polaron species, respectively. **b.** Oscillator strength of  $X_1^+$  (red) and  $X_2^+$  (blue) as a function of doping for devices D1 (left) and D2 (right). **c.** Schematics of excitons coupled with charge excitations in correlated insulating states.  $X_1^+$  arises from an exciton dressed up by a polarization associated with the charge excitations across the charge-transfer (CT) gap (green arrow), while  $X_2^+$  couples with charge excitations across the Mott gap (yellow arrow). At  $\nu = -2$ , the charge excitation across the charge-transfer gap is Pauli-blocked.

As a result, we attribute  $X_2^+$  as excitons dressed up by charge excitations across the Mott gap. Based on this picture, the binding energy difference between the two exciton polarons is likely rooted in the difference in the polarizability associated with the excitations above

different gaps. While a full many-body calculation is beyond our expertise, this exciton-polaron picture is at least consistent with the observed doping dependence.

In the revised manuscript, we have updated Figure 4 and rewritten the section to be **"In this section, we propose an alternative picture, in which the two features arise from the hybridization between excitons and the charge excitations above the Mott or charge-transfer gap (Figure 4b)..."** on Page 6, section "Excitons coupling to correlated insulating states".

10. In the page 6, the authors mentioned that "According to our DFT calculations, the excitons and holes in both X1+ and X2+ are located at the XM or MX moire site". Where do the authors provide these DFT calculations?

#### Response 16

We thank the reviewer for the question. Our DFT calculation is included in Supplementary Information S5 and Figure S5. Our DFT calculation confirms that the Wannier orbitals of excitons are located at MX and XM sites. The doped holes are also at the MX and XM sites, which have been theoretically verified by previous DFT calculations(*Proceedings of the National Academy of Sciences 118.10 (2021)*). In the revised manuscript, we have highlighted this on Page 7, Paragraph 2: **"Our DFT calculation confirms that the Wannier orbitals of excitons are located at MX and XM sites (Supplementary Information S5)."**

11. In the page 6 and 7, the authors mentioned that "As the doping level increases further, the holes at a doubly occupied site can no longer couple to any exciton". Why is this the case? Even if there are two holes, wouldn't one of the holes which has the opposite spin to the hole of the exciton form a trion state?

#### Response 17

We thank the reviewer for this question. As discussed in response 15, we conclude in the revised manuscript that  $X_2^+$  arises from excitons coupled with charge excitations across the Mott gap, as illustrated in the **Response 15**. When the moiré site has already been filled with two holes( $\nu < -2$ ), the direct hopping between the nearest MX and XM site is turned off

due to Pauli blocking; the charge excitation across the Mott gap is thus suppressed, leading to a decrease of  $X_2^+$  oscillator strength. We have updated such a mechanism on Page 7, Paragraph 4, **"The holon-doublon creation probability increases toward  $\nu = -2$ , at which the doped holes only need to hop once between the nearest MX and XM sites, and subsequently decreases when some sites become doubly occupied, consistent with the maximum oscillator strength of  $X_2^+$  at half-filling."**

12. In the page 7, the authors mentioned that "We attribute the MCD signal in  $X_2^+$  to the intervalley exchange interaction between the spin-singlet and spin-triplet trions". Why is this the case? Doesn't the author need to consider valley Zeeman splitting of excitons at  $\pm K$  points and energy shift due to the Fermi polaron formation by considering the spin polarization at the  $\Gamma$  point?

## Response 18

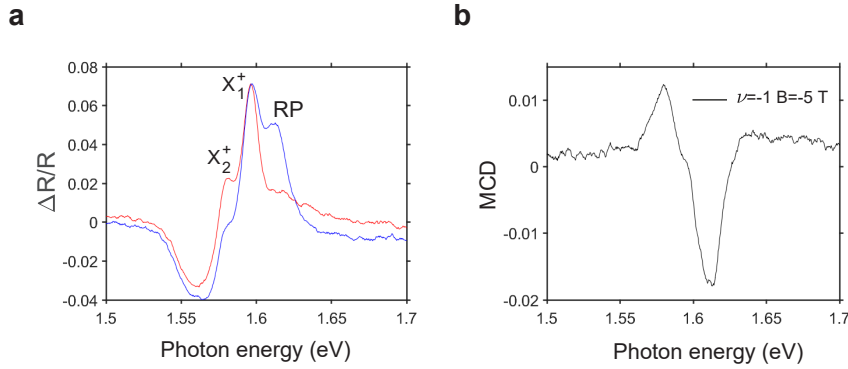
We thank the reviewer for raising this question. We first need to clarify that our measurement probes the absorption of exciton polarons. In this case, the valley Zeeman effect primarily leads to a peak splitting between  $\sigma^+$  and  $\sigma^-$  helicity channels. Unlike photoluminescence, the absorption process does not involve any relaxation of excited states. As a result, the valley Zeeman effect is not expected to lead to a significant difference in the oscillator strength, i.e. magnetic circular dichroism (MCD), as observed in our experiment. Instead, we attribute the MCD signals arising from the exchange interaction between the exciton and doped holes.

Since a bright exciton in the  $\pm K$  valley consists of a hole of a definite spin (up or down), it should preferentially couple with holes of opposite spin drawn from the  $\Gamma$  valley ( $\Gamma$ -valley hole) due to the exchange effect. On the other hand, when a magnetic field is applied, the spin Zeeman effect would split the hole band in the  $\Gamma$  valley, producing a net spin polarization among the doped holes. Such a spin polarization leads to more exciton-polaron formation from the preferred  $K$  valley, and therefore a larger absorption peak in the corresponding helicity channel, following the valley selection rule.

Experimentally, we find the found MCD is strongest at the  $X_2^+$  peak, which we now assign to be related to excitations across the Mott gap, and a branch of repulsive polaron, where

the MCD signal is integrated (Figure R6). Little MCD is found in the  $X_1^+$  exciton polaron associated with the above charge-transfer gap excitation. These subtle contrasts require more studies in the future.

In the revised manuscript, we deleted the schematic of three-particle binding in Figure 5, consistent with the new exciton-polaron picture. We have further illustrated this point on Page 8, Paragraph 2, **"We attribute the MCD signal in  $X_2^+$  to the exchange interaction between the spin-polarized holes doped in the  $\Gamma$  valley and the excitons in the  $\pm K$  valleys. A  $\Gamma$ -valley hole preferentially couples to the exciton with a hole of opposite spin, leading to more exciton-polarons formed in a particular K valley."**



**Figure.R6:** **a.** Reflection contrast with  $\sigma^+$  (red) and  $\sigma^-$  (blue) optical excitation of a t-MoSe<sub>2</sub> at  $\nu = -1$ . The magnetic field is -5 T. **b.** MCD as a function of photon energy.

Reviewer # 3 (Remarks to the Author):

The authors report the observation of correlated electronic states in twisted MoSe<sub>2</sub> homobilayer heterostructures, which realize a honeycomb moiré superlattice. In these systems, the top two moiré bands originate from the folded valence band at the  $\Gamma$  valley of the monolayer Brillouin zone. Due to strong hybridization and weak spin-orbit coupling in the  $\Gamma$  valley, the moiré potential minima are symmetrically distributed between the MX and XM sites, making it similar to a graphene honeycomb lattice. The various electronic states are optically probed using Rydberg excitons in a WS<sub>2</sub> sensing layer and by moiré trions in the MoSe<sub>2</sub> bilayer. The authors identify the insulating state at  $\nu = -1$  as a sublattice-polarized

electronic crystal arising from the nearest-neighbor Coulomb repulsion, and the state at  $\nu=-2$  as a Mott insulator arising from the on-site Coulomb repulsion. Additional insulating states are identified at fractional fillings.

Overall, this work provides valuable insights into correlated electron physics and light-matter interactions in two-dimensional materials with moiré superlattices. The manuscript is generally clear, and the key conclusions are well-supported by both experimental data and theoretical modeling. I therefore recommend publication after the authors address the following comments.

We thank the reviewer for the positive assessment of our work. The questions have helped us improve the quality of the manuscript. In the following, we will address the reviewer's concerns point by point.

1. Why the  $\nu=0$  is not at  $V_{tg}=-1$  V where you observe an abrupt 18-meV blue shift in Figure 2? If the reason is non-linear gate-dependent doping, the assignment for the fractional filling might be wrong. Why the signature of the  $\nu=-1/3$  is weaker than that of  $\nu=-1/6$ ?

#### **Response 19**

We thank the reviewer for this question. We have addressed most aspects of this concern in **Response 13**. We are confident that most fractional filling assignments are accurate, as evidenced by the clear linear relationship between the gate voltage and the filling factor in Figure R3. Nevertheless, we agree that the filling assignments at low doping have larger uncertainties due to the nonlinear gating effect. After re-examining the experimental data, we have removed the assignment for  $-1/3$  state to avoid any potential misinterpretation.

2. Does all the gate voltage dependence (in all the figures) follow  $V_{bg}=1.15V_{tg}$ ? If so, I recommend defining it in text (rather than in Figure 2 caption) and using it instead of  $V_{tg}$ .

#### **Response 20**

We thank the reviewer for the question. We apologize for the confusion caused by the gate voltage label. In all the main figures, the gate voltage dependence follows  $V_{bg} = 1.15V_{tg}$ . In the revised manuscript, we have defined an effective gate voltage ( $V_g = \frac{V_{tg} + (C_b/C_t)V_{bg}}{2}$ ) in the

main text and all the figures, instead of using  $V_{tg}$ .

3. Regarding the  $X+1$  trion in the MoSe<sub>2</sub>, although it might be a charge transfer trion, I don't see why a tightly bound trion cannot form at the same doping condition. Intuitively, I would expect similar oscillator strength for the two species at  $\nu=-1$ . What is the reason for the abrupt jump from one to the other in Figure 4?

### Response 21

We thank the reviewer for this important question. We apologize if the original plot caused any confusion regarding the doping dependence of  $X_2^+$ . We want to clarify that the oscillator strength of  $X_2^+$  does not exhibit any abrupt jump. In the revised Figure R5a, we replot the first derivative of the reflectance spectra as a function of gate voltage ( $V_g$ ). The data clearly show that the oscillator strength of  $X_2^+$  gradually increases from  $\nu = 0$  to  $\nu = -2$ , and then decreases when  $\nu < -2$ .

We agree with the reviewer that a purely real-space picture cannot fully capture the doping dependence of the oscillator strength, which is also pointed out by reviewer 2. As discussed in **Response 15**, we now interpret  $X_1^+$  as an exciton dressed up by a polarization associated with charge excitations across the charge-transfer (CT) gap, while  $X_2^+$  arises from excitons coupled to charge excitations across the Mott gap. At low doping levels (e.g.,  $\nu = -1$ ), hopping across the Mott gap requires a second-order process involving an intermediate site, making it weaker than the first-order nearest-neighbor hopping across the CT gap. Consequently, the oscillator strength of  $X_2^+$  is smaller than that of  $X_1^+$  at  $\nu = -1$ .

To address the reviewer's concern, we have updated Figure 4a and explained why  $X_2^+$  has a smaller oscillator strength at  $\nu = -1$ , on Page 7, paragraph 4, **"A second class of excitations involves hopping across the Mott gap to create a holon-doublon pair. In this case, a doped hole must hop twice to form a doubly occupied state at an MX (or XM) site (yellow arrow in Figure 4c), a second-order process that is small but finite at low doping, consistent with the weak oscillator strength of  $X_2^+$  at  $\nu = -1$ ..."**

4. It was previously reported on light-induced spin depolarization in MoSe<sub>2</sub>. Did you verify that there is no spin depolarization in the magnetic circular dichroism experiments? [Nature,

623(7987):509–513, 2023 ; Phys. Rev. Lett. 128, 127402 (2022)].

## Response 22

We thank the reviewer for the suggestion. These two works previously reported that the degree of circular polarization of the attractive Fermi-polaron at the electron-doping side decreases with increasing light intensity. In this work, we instead focus on the hole-doped regime, where doped holes and excitons are separated in momentum space. Experimentally, we did not observe any prominent spin depolarization effect. We attribute this observation to two factors: 1. The excitation level is kept low in our experiments to avoid any nonlinear effect. Although we use a halogen-lamp based white-light source, a long-pass filter (700 nm cutoff) is used to filter the short-wavelength component. The total power at the sample is only about 10 nW, with the amount of power absorbed being even less. 2. The spin depolarization in MoSe<sub>2</sub> is only prominent in the dilution fridge temperature regime (tens of mK), while the base temperature in our experiments is 1.6 K.

In the revised manuscript, we have included the discussion on the spin depolarization in the Methods section, **"No significant spin depolarization is observed."**

## Round 2

### Point-by-Point Response:

Reviewer # 1 (Remarks to the Author):

I have carefully read both responses to my comments and the revised manuscript. The authors have clearly addressed all points I raised and have made a substantial effort, including the addition of several new figures. Overall, the experimental data appear very solid, and the interpretations are coherent. While some aspects of the work have been questioned and debated by other referees, I find that these issues are difficult to disentangle at the present stage, given that polaronic effects involving strongly correlated electrons remain a developing field. This study will undoubtedly stimulate further theoretical research. Its impact clearly meets the expectations for Nature Communications and is likely to attract significant attention. Therefore, I recommend publication in its present form.

### Response 23

We appreciate the reviewer's encouragement. The interaction between excitons and correlated electrons in semiconducting moiré materials is indeed a very interesting and complex topic that certainly deserves further study from both theoretical and experimental perspectives.

Reviewer # 2 (Remarks to the Author):

I appreciate the authors' detailed reply and their effort to deepen the discussion. Except for the interpretation of  $X1+$  and  $X2+$  states, I am convinced by their explanation. I understand that the authors now attribute  $X1+$  and  $X2+$  as a kind of shakeup process. However, I think such processes result in blueshift from the original exciton resonances since additional energy to excite those states is required. Even if we assume very small inter-site trion binding energy which perhaps could account for net redshift,  $X2+$  dominated by on-site e-e Coulomb repulsion energy scale should be on the blue side compared to  $X1+$  dominated by nearest neighbor site e-e Coulomb repulsion energy scale. I recommend the authors formulate those relations with equations like the authors described for the case of Rydberg state energy blueshift in Response 10. Also, I wonder where the on-site trion (or attractive polaron) state appears which is missing in this new interpretation.

## Response 24

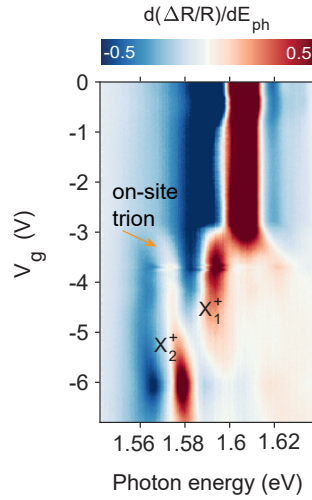
We thank the reviewer for raising this important question. We agree that if the  $X_1^+$  and  $X_2^+$  branches were repulsive exciton-polarons, i.e., no binding exists between excitons and charges, the branch associated with the larger many-body charge gap should exhibit a higher resonance energy. In that case,  $X_2^+$  would have appeared on the blue side of  $X_1^+$ . However, the experimentally observed resonances correspond to attractive exciton-polarons, whose energies are determined by the competition between the binding energy between constituents and the charge gap energy. In the simplest picture, the resonance energies can be expressed as

$$\begin{aligned} E_{X_1^+} &= E_X - E_{b1} + \Delta_{CT}, & (\nu = -1), \\ E_{X_2^+} &= E_X - E_{b2} + U_0, & (\nu = -2). \end{aligned}$$

Here  $E_X$  is the exciton resonance energy,  $E_{bi}$  denotes the effective binding energy between exciton and charges, while  $\Delta_{CT}$  and  $U_0$  represent the energy required to create the corresponding charge excitations. Since  $E_{b1}$  and  $E_{b2}$  are associated with very different dressing processes, their values can differ significantly and therefore determine the relative ordering of the peaks.

In the low doping limit, the weak feature highlighted in Figure R8 is consistent with a conventional trion resonance, with a peak energy  $\sim 30\text{meV}$  redshifted from the neutral exciton. In contrast to the usual trion (attractive polaron) behavior, where increasing carrier density leads to further redshift, we observe a blueshift as doping increases. This unusual behavior indicates that many-body effects become important as the doping density increases and that the simple few-body trion evolves into the exciton-polaron  $X_2^+$  discussed in the manuscript. Based on this picture, the short-range exciton-charge attraction dominates the  $X_2^+$  branch while the  $X_1^+$  branch is associated with a more spatially extended exciton-charge interaction. As a result, the binding energy of  $X_2^+$  at  $\nu = -2$  is larger than the binding energy of  $X_1^+$  at  $\nu = -1$  ( $E_{b2} > E_{b1}$ ). Since the binding energy difference exceeds the gap energy difference, the net result is a lower resonance energy for  $X_2^+$  compared to  $X_1^+$ .

On the other hand, we also acknowledge that the relative energy positions of different exciton-polaron peaks associated with various inter-band transitions can be further influenced by other doping-dependent factors. As the doping level increases, the Coulomb interaction will affect the attraction strength between the exciton and charge as well as the exciton energy (through reduced quasiparticle gap and reduced electron-hole binding), while the relative exciton-charge distance is reduced on average. Potentially, these many-body effects jointly determine the experimentally observed spectra, which warrant more rigorous theoretical investigations, as pointed out by the Reviewer 1.



**Figure.R8:** The reflection contrast spectrum as a function of gate voltage near the A exciton resonance of t-MoSe<sub>2</sub> in device D2 ( 2.9°).

In the revised manuscript, we have included the discussion on the resonance energies of  $X_1^+$  and  $X_2^+$  on page 7, paragraph 3, "**The resonance energy of  $X_2^+$  becomes lower than that of  $X_1^+$ , indicating that the binding energy difference between the two attractive branches exceeds their gap energy difference.**" The onsite trion at low doping is labeled by black arrows in Figure 4a.

Reviewer # 3 (Remarks to the Author):

I have read the response and the revised manuscript. Two important concerns remain:

1. The authors did not address the identification of fractional filling, except for removing the  $\nu=1/3$ . Although the assigned filling is linear with the applied gate voltage, the doping

seems non-linear in the low-doping regime. Regarding the  $\nu=1/3$  state, I still expect that it will show a stronger optical signature than the  $\nu=1/6$  state. What is the reason for its absence? An additional remark about filling assignments is that in Fig.2a,b, the  $\nu=1$  is at a gate voltage of  $V_g \sim 2.3$  V, while Fig. 4a shows that the  $\nu=1$  is at a gate voltage of  $V_g \sim 2.7$  V. This difference accounts for about 1/3 of the difference between  $\nu=1$  and  $\nu=2$  of Fig. 2.

## Response 25

We fully understand the reviewer’s concern over the nonlinear doping effect, which is precisely why we chose to focus on the results taken at 7 K when we assigned the filling number. (Fig. 2) Experimentally, we find that the non-linear doping effect becomes weaker as the temperature increases, likely due to the improved quality of the electrical contacts. At 7 K and above, the voltage spacings between adjacent integer fillings, i.e. between  $\nu = -2$  and  $-1$  and , between  $\nu = -1$  and 0, becomes equal within  $\pm 0.2$  V,. Additionally many fractional states such as  $\nu = -1/2, -2/3, -5/6$  fall on the linear voltage-filling relationship extrapolated from the integer states, indicating a negligible contribution from the non-linear doping effect at moderate doping densities. This same method is consistent with that adopted in prior pioneering studies (*Nature*, 587, 214–218 (2020), *Nature Materials*, 23, 219–223 (2024)) These corroborations let us conclude that our peak assignments are accurate for filling assignments larger than 1/2. In the low-doping regime, there could exist a small non-linear gating effect, giving rise to an uncertainty of  $\pm 1/6$  for fillings smaller than 1/2.

On the other hand, the electrical contacts become less conductive as the temperature decreases, as evidenced by an increasing voltage required to reach the  $\nu = -1$  state (Extended Figure 3). The data down in Fig. 4 are measured at 1.6 K, which is the best condition to observe exciton-polarons and their spin susceptibility, but the integer states are no longer equally spaced at such a low temperature due to non-linear gating effects. For this reason, we deliberately based our filling assignments on the higher-temperature data, where linearity is better established.

Regarding the optical signature strength of  $\nu = -1/6$  versus  $\nu = -1/3$ , we agree with the reviewer’s concern regarding the hierarchy of Wigner crystal states. Since generalized Wigner crystals are stabilized by long-range Coulomb interactions, the  $\nu = -1/3$  state should ex-

hibit a larger gap and thus a stronger optical than  $\nu = -1/6$ . Since this behavior is not observed experimentally, we cannot unambiguously attribute the weak feature near  $\nu = -1/6$  to a genuine Wigner crystal state. Possible explanations include defect-induced insulating behavior or residual nonlinear gating effects that can affect the filling calibration at low densities. To be on the conservative side, we have therefore removed all explicit the filling assignments for the features above  $\nu = -1/2$  state in the revised manuscript.

2. I am not convinced regarding the new interpretation of the  $X_2^+$  state. With the large  $U_0/t$  ratio and on-site repulsion energy ( $U_0 \sim 25$  meV according to the numerical results, and  $U_0 \sim 100$ -200 meV in the last paragraph of the introduction), why do the authors expect excitations in low temperatures?

The authors should address these concerns before I can recommend publication.

## Response 26

We thank the reviewer for raising this critical issue, which allows us to clarify the nature of the charge excitations involved in the formation of the  $X_2^+$  state. We agree with the reviewer that, given the large Mott gap ( $U_0$ ) compared to the thermal energy at low temperatures ( $k_B T \ll U_0$ ), the thermally populated charge excitations (doublon-holon) is negligible. In other words, the background charge distribution is frozen in the Mott insulating state.

However, the formation of the attractive polaron  $X_2^+$  does not rely on pre-existing thermal excitations. Instead, when the exciton is introduced into the moiré lattice, the strong local dipole-charge attraction couples the ground state to excited states, even at zero temperature. As described in the exciton-polaron theory (*Physical Review B* 103.7 (2021): 075417), the wavefunction of an exciton-polaron is a superposition state between the bare exciton and charge excitations (Holon-doublons excitations from the Mott insulator in our case). The absorption peak of the attractive polaron  $X_2^+$  corresponds to the simultaneous creation of an exciton and charge excitations from the lower Hubbard band to the upper Hubbard band. In other words, the excitations arise from exciton dressing induced by the strong exciton-charge interaction. Following the simple formalism proposed in response 24, such excitation is possible as long as the binding energy exceeds the gap energy, which is indeed the case for any attractive branch. Thus despite the large value of  $U_0$ , low-temperature optical excitations involving  $X_2^+$  remain fully consistent with the Mott-insulating ground

state.

In the revised manuscript, we have included the relevant discussion **"The absorption peak of the attractive polaron  $X_2^+$  corresponds to the simultaneous creation of an exciton and charge excitations from the lower Hubbard band to the upper Hubbard band. The holon-doublon excitations arise from exciton dressing induced by the strong exciton-charge interaction."** on page 7, paragraph 3.

### Round 3

#### Point-by-Point Response:

Reviewer # 2 (Remarks to the Author):

I thank the authors for their detailed response. The explanation that the difference in binding energies leads to  $X_2^+$  appearing at a lower energy than  $X_1^+$  is convincing. My concern has been satisfactorily addressed, and I recommend publication of the manuscript

#### Response 23

We appreciate the reviewer’s support for our work.

Reviewer # 3 (Remarks to the Author):

I have carefully considered the authors’ reply to my questions. Although I still find this work interesting and important, I am not fully convinced by the interpretation of the  $X_2^+$  origin.

We appreciate the reviewer’s positive comments, as well as their reservations regarding the interpretation of  $X_2^+$ ’s microscopic origin. We fully acknowledge that this aspect of the work involves subtle many-body physics, and that achieving a definitive microscopic interpretation is currently very challenging. As Reviewer 1 previously wrote, “these issues are difficult to disentangle at the present stage,” reflecting the broader complexity of polaronic effects in strongly correlated systems.

We agree with the reviewer that our experimental results cannot definitively resolve the open question regarding  $X_2^+$ , and that care should be taken to avoid over-interpretation in the absence of additional theoretical or experimental input. Therefore, we have revised the manuscript to clarify such limitations, with the new insertion quoted below. Since the interpretation is not central to our main conclusions, we believe this approach appropriately reflects the level of interest and uncertainty surrounding this topic in the field.

“Based on the doping dependence, we **tentatively** interpret  $X_1^+$  as an exciton dressed by a polarization associated with the charge excitation across the charge-transfer (CT) gap... **On the other hand, exciton-polaron (or trion) excitation in a strongly correlated Fermi sea is a complicated phenomenon and more research is required to reveal**

their microscopic origin.”