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Supplementary information

Correlated insulating states at fractional fillings of moiré superlattices

In the format provided by the authors and unedited

nature research

Peer Review File

Manuscript Title: Correlated insulating states at fractional fillings of moiré superlattices

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referee #1 (Remarks to the Author):

The manuscript by Y. Xu et al reports an experimental observation of nearly two-dozen correlated insulating states at fractional fillings of moire bands in WSe2/WS2 heterostructure. The observation is based on an optical sensing technique, where the reflection contrast spectra of a sample layer (monolayer WSe2) is measured. The 2s exciton feature in the spectra from the sample layer is sensitive to dielectric environment, and therefore, becomes prominent when the sample layer WSe2/WS2 develops correlated insulating states driven by enhanced many-body interactions. These correlated insulating states at fractional fillings are interpreted as generalized Wigner crystals or charge density waves, which are calculated using a classical Monte Carlo simulation by neglecting electron hopping in the model. The experimental transition temperatures and those estimated using the Monte Carlo simulation show good consistency at many filling factors below 1.

I find that both the results reported and the new technique developed by this work are novel. The observation of correlated insulating states in moire bands at many fractional filling factors in one system is rare, and brings out new physics. It demonstrates the feasibility of using moire superlattices to realize strongly-correlated effects and to quantum simulate model Hamiltonians. The new optical sensing technique is crucial to reveal these correlated insulating states, which would be hard to detect using other techniques, particularly in TMD systems with large contact resistance. Therefore, I think this work could be potentially published in Nature. However, there are technical questions that need to be clarified, as listed in the following.

(1) In the middle panel of Fig. 1c, the 2s exciton in WSe2 is claimed to have a stepwise jump when the sample layer WS2 is electron doped. However, this is not obvious from this plot. Right above the horizontal dashed white line, the 2s exciton appears to continuously blue shift, instead of a sudden red shift. To address this question, more line cut plots of this figure could be useful.

Suppose that the 2s exciton indeed has a stepwise red shift, what is the reason for the red shift (instead of a blue shift)? With increased screening, the binding energy decreases. I would expect blue shift instead of red shift. Some discussions on this point would be helpful.

(2) In the main experiment that uses WSe2 as the sensor layer and WS2/WSe2 as the sample layer, how to distinguish optical signals from the sensor layer and the sample layer? This is an important question, because the same material WSe2 is used in both the sensor layer and the sample layer. I do not find discussions on this point, and clarification is needed.

Referee #2 (Remarks to the Author):

Xu et al report many previously unobserved correlated insulating states in WSe2/WS2 moiré superlattices. The detection of this states is carried out using an original technique, with a WSe2 monolayer separated from the main heterobilayer by an hBN spacer. The response of the 2s excited exciton state sensitive to its dielectric environment is used for detection of the insulating

states in the heterobilayer. The new observations are very interesting, and in some ways represent the pinnacle of the moiré physics in TMD heterobilayers. Equally, this work opens the ways to further detailed studies of the correlated states as the authors suggest.

While the observations of the 2s state behaviour are presented, further insight in the behaviour in the WSe2/WS2 heterobilayer itself as well as insight in the device operation would be very helpful. With these points clarified I would be happy to recommend this work for publication in Nature.

1. It is not clear why the WSe2 reflection contrast (RC) spectra measured on the device show the RC signal on the 'sensor' WSe2 but not the WSe2 in the heterobilayers.

2. In regards to question 1, it is not entirely clear what is shown in Fig.1f. There are features below 1s exciton. What are they? Can they also tell what's happening in the heterobilayer? They seem to be not explained in detail in the paper. Why?

3. Is there a more direct evidence of the changes occurring in the WSe2/WS2? In the authors' previous paper on the same heterobilayer system, where the insulating states were found for the first time (Tang et al Nature 2020), the optical response from WSe2 in the heterobilayer was measured. Can similar data be presented here? Do fractional states show any signatures when they are measured on the actual heterobilayer? If not, please explain why.

4. Are there any studies of control sample with the 'sensor' WSe2 layer only? It appears that such RC measurements in these fine details and low excitation power have never been done before, and the behaviour of 2s exciton RC as a function of the gate voltage is not really known. It would be very desirable to measure such a control sample too, or to point the readers to a similar study, where in similar conditions the 2s state is insensitive to the doping.

5. Did the authors measure more than one sample? I do appreciate that it is a very complicated structure, which would probably take weeks and several attempts to build. I wonder still if the role of the thickness of the hBN buffer between the 'sensor' and the heterobilayer has been investigated. How have the moiré effects in the sensor WSe2 been excluded from the picture, for example? Is the sensor layer randomly orientated? Is it important which of the layers, WS2 or WSe2 face the sensor?

Referee #3 (Remarks to the Author):

The manuscript of Xu et al. reports abundance of new correlated states at fractional fillings of WSe2/WS2 moiré system. By employing a local dielectric sensor (single-layer WSe2) in proximity to WSe2/WS2 bilayer, Xu et al. manage to resolve many more fractional filling states, compared to a recent work reported by the Berkeley group (Regan, E. C. et al, Nature (2020)). Despite not being the first experimental work addressing fractional filling correlated states in this system, the work of Xu et al. is still significant in following ways.

Scientifically, the cascades of correlated states observed here is fascinating. It makes us understand better the nature of correlated states in moiré system which is intensively studied in recent years.
Technically, detecting dielectric environment (different electronic phases) with exciton states could also be potentially applied to other materials and thus could attract a wide range of interests.

In general, the experimental idea, perspective and results in this manuscript make me a positive impression. I would recommend its publication in Nature if my following concerns can be properly addressed.

1. In Fig. 1c, the 2s states of WSe2 shows similar signatures (shifted to higher energy above white dashed line) like 1s states in WS2. I would suggest the authors intercept these features better.
2. In Fig. 1, all figures are arranged clockwise. Perhaps the authors should rearrange them in normal way to make readers easier to follow.
3. In line 165-166, it is vague by saying "The electron and hole sides behave similarly, suggesting that the same physics is in play". It should be better addressed.
4. I don´t find the assignment of the filling factor in Fig.2 convincing enough. i.e., 1) I cannot see any reliable signatures of 1/7 and 6/7 states. 2) the gate voltage difference between -1/3 and -1/4 states is almost 30 percent larger than that between 1/3 and 1/4 states, etc. More statistics (i.e. more datasets taken from different regions of the device or a second device) is helpful.
5. Last but not latest, I would suggest the authors to rule out the possibilities that some of the correlated states resolved might come from the in-homogeneity of twist angles or charged impurities.

Author Rebuttals to Initial Comments:

We would like to thank all of the reviewers for reviewing our manuscript and the thoughtful comments. In the following we address these comments point-by-point. We have also performed new experiments to further support our conclusion. These include measurements on different regions of the device to illustrate the role of inhomogeneities and on a new device to verify data reproducibility. For clarity, the reviewers' comments are in blue, our responses are in black.

Referee #1:

The manuscript by Y. Xu et al reports an experimental observation of nearly two-dozen correlated insulating states at fractional fillings of moiré bands in WSe_2/WS_2 heterostructure. The observation is based on an optical sensing technique, where the reflection contrast spectra of a sample layer (monolayer WSe_2) is measured. The 2s exciton feature in the spectra from the sample layer is sensitive to dielectric environment, and therefore, becomes prominent when the sample layer WSe_2/WS_2 develops correlated insulating states driven by enhanced many-body interactions. These correlated insulating states at fractional fillings are interpreted as generalized Wigner crystals or charge density waves, which are calculated using a classical Monte Carlo simulation by neglecting electron hopping in the model. The experimental transition temperatures and those estimated using the Monte Carlo simulation show good consistency at many filling factors below 1.

I find that both the results reported and the new technique developed by this work are novel. The observation of correlated insulating states in moiré bands at many fractional filling factors in one system is rare, and brings out new physics. It demonstrates the feasibility of using moiré superlattices to realize strongly-correlated effects and to quantum simulate model Hamiltonians. The new optical sensing technique is crucial to reveal these correlated insulating states, which would be hard to detect using other techniques, particularly in TMD systems with large contact resistance. Therefore, I think this work could be potentially published in Nature. However, there are technical questions that need to be clarified, as listed in the following.

Response 1:

We thank the referee for the positive assessment of our work.

(1) In the middle panel of Fig. 1c, the 2s exciton in WSe_2 is claimed to have a stepwise jump when the sample layer WS_2 is electron doped. However, this is not obvious from this plot. Right above the horizontal dashed white line, the 2s exciton appears to continuously blue shift, instead of a sudden red shift. To address this question, more line cut plots of this figure could be useful.

Suppose that the 2s exciton indeed has a stepwise red shift, what is the reason for the red shift (instead of a blue shift)? With increased screening, the binding energy decreases. I would expect blue shift instead of red shift. Some discussions on this point would be helpful.

Response 2:

We apologize for the over simplified discussion on the result in original Fig. 1c (now 1d). Figure R1a (same as the middle panel of Fig. 1d in the main text) is the gating dependence of the 2s exciton in the WSe_2 sensor. Above the dashed white line (onset of electron doping in the WS_2 sample), two branches come out of the original neutral 2s state. One branch blue shifts and loses oscillator strength continuously with doping. At the same time, another branch emerges at ~ 25 meV below the original 2s resonance and gains oscillator strength with doping. (The feature around 1.88 eV is the 3s resonance in the sensor, which also goes away with doping in the sample.) At higher doping densities in the sample, the blue-shifted branch disappears and only the red-shifted branch remains. The gate dependence of the spectral weight of the two branches is shown in Fig. R1c. It resembles a broadened step-like dependence. We note that the doping dependence of the 2s resonance in the sensor mirrors that of the fundamental exciton resonance in the WS_2 sample (right panel of Fig. 1d).

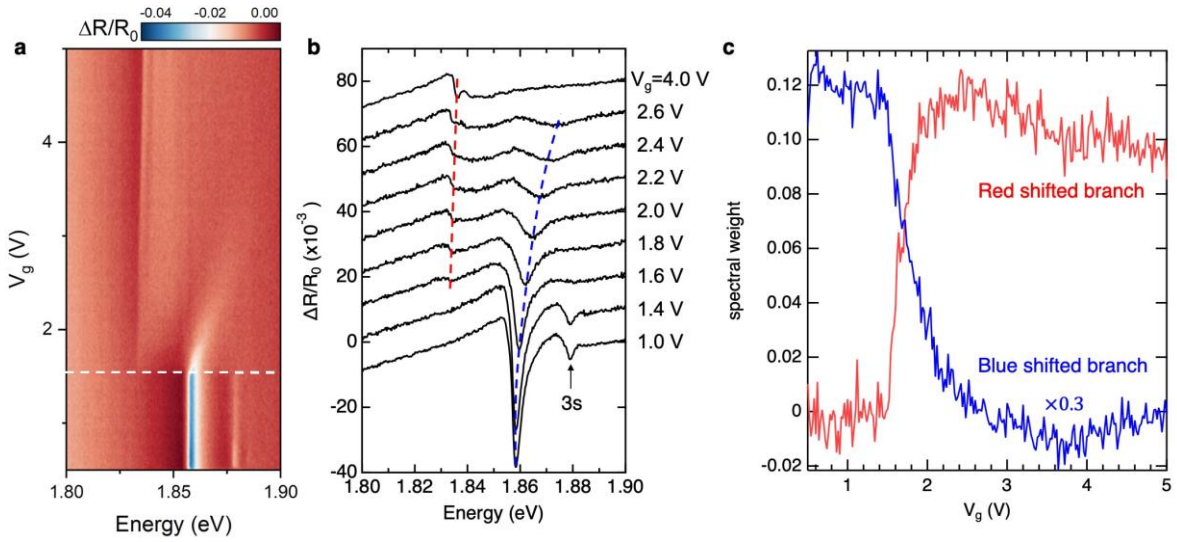

Figure R1. (a) Gate-dependent reflection contrast spectrum ($\Delta R/R_0$) near the WSe_2 2s transition energy for the control device (same data as the middle panel of Fig. 1d). The WS_2 sample is electron doped above V_g = 1.5 V (white dashed line). (b) Representative linecuts of (a), vertically displaced for clarity. The red and blue dashed curves highlight the red-shifted and blue-shifted branches, respectively. (c) Gate dependence of the integrated spectral weight of the two branches. The

featureless response at 1 V and 4 V was used as background for the red-shifted and blue-shifted branches, respectively. The spectral weight of the blue-shifted branch was scaled by a factor of 0.3.

The observation can be explained by the formation of polarons. The concept of polarons in the context of exciton-electron interactions has been invoked to understand the doping dependence of the fundamental excitons in monolayer TMDs, such as the data in the right panel of Fig. 1d [Phys. Rev. B 95, 035417 (2017) and Nature Physics 13, 255–261 (2017)]. In short, an exciton in a degenerate electron gas can polarize the electron gas and form an attractive polaron bound state (often also referred to as a charged exciton) and a repulsive polaron state. They correspond to the red-shifted and blue-shifted branch, respectively. The attractive polaron shows a sudden red shift and an increase in oscillator strength with doping, whereas the repulsive polaron exhibits a continuous blue shift and rapidly diminishing oscillator strength. At higher doping levels, where screening of exciton-electron interactions becomes significant, the repulsive polaron disappears and the attractive polaron evolves continuously into the band-to-band transitions. Similarly, excitons in the sensor can interact with electrons in the sample to form (interlayer) polarons, which could account for the observed doping dependences of the 2s exciton in Fig. R1. Interlayer polaron effect is not observed for the 1s exciton in the sensor since it's tightly bound with a Bohr radius of ~ 1 nm.

The referee also raised the question on why the 2s exciton resonance generally red shifts with enhanced screening (upon moderate doping when only the red-shifted branch remains). The reason is that screening renormalizes both the exciton binding energy and the quasiparticle band gap. Enhanced screening or weakened Coulomb interaction decreases both energies. The 2s resonance red shifts because the decrease in the 2s exciton binding energy is less than the decrease in the quasiparticle band gap. It has also been shown that the two effects cancel out for the 1s exciton [Nano Lett. 16, 5568–5573 (2016)]. As a result, the 1s resonance shows a negligible change in energy.

In the revised manuscript, we have included a more detailed discussion on the doping dependence of the fundamental exciton in the sample (WS_2) and pointed out the similarity between the behavior of the 2s exciton in the sensor and the fundamental exciton in the sample (last two paragraphs on page 2). We have also added a new paragraph in the Methods section (Mechanism of sensitivity of 2D excitons to dielectric environment) to discuss the polaron effect in the low doping regime. Figure R1 has been included as new Extended Data figure 1.

(2) In the main experiment that uses WSe_2 as the sensor layer and WS_2/WSe_2 as the sample layer, how to distinguish optical signals from the sensor layer and the sample layer? This is an important question, because the same material WSe_2 is used in both the sensor layer and the sample layer. I do not find discussions on this point, and clarification is needed.

Response 3:
We thank the reviewer for this important question. The optical reflection contrast contains information of WSe_2 in both the sensor and the sample (Fig. 1f). But their response is well separated in energy. The fundamental exciton resonance energy of WSe_2 in the sample is about 50 meV smaller due to the moiré potential in the heterobilayer.

We illustrate this point in Fig. R2. We show the doping-dependent reflection contrast of the device from regions without and with the sensor, respectively in panel a and b. Figure R2a clearly shows the fundamental moiré excitons around 1.68 eV and their characteristic doping dependences in the WSe_2 layer of the moiré superlattice. The data is in full agreement with a recent study [Nature 579, 353–358 (2020)]. Figure R2b shows the 1s (~ 1.73 eV) and 2s resonances (~ 1.83 eV) of the sensor in addition to the fundamental moiré excitons in the sample. The nearly identical behavior of the moiré excitons in the two panels verifies that the sensor has a negligible effect on the sample. In addition, the spectral window around the 2s exciton in the sensor is free of any strong exciton resonances from the sample, which allows a high detection sensitivity.

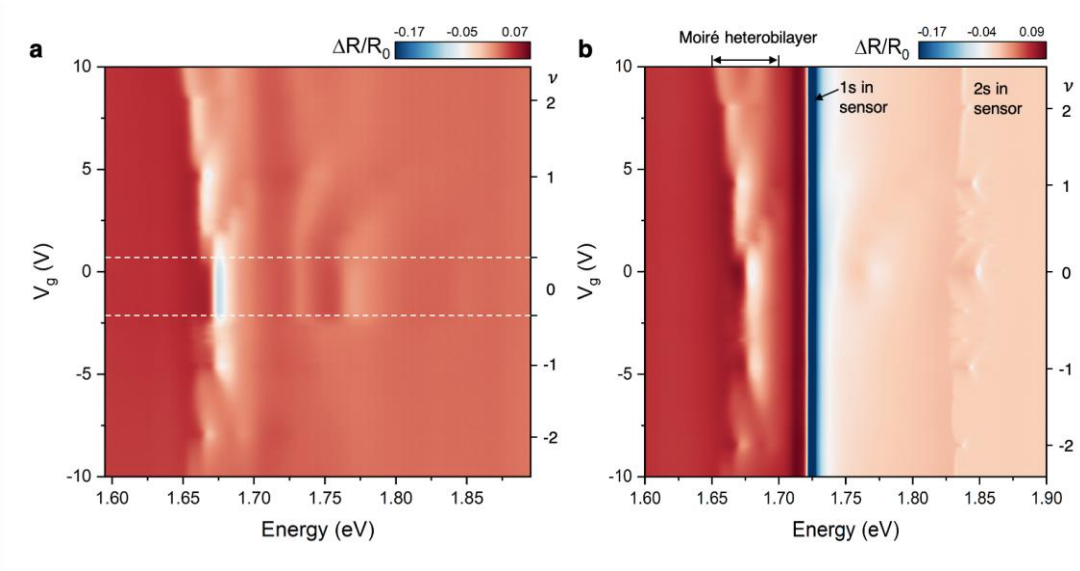

Figure R2. Gate-dependent reflection contrast spectrum in regions of the main device without (a) and with (b) the WSe_2 sensor at 1.6 K. Between the two horizontal dashed lines in (a) is the charge neutral region of the WSe_2/WS_2 heterobilayer.

In the revised manuscript, we have explicitly discussed how to distinguish the optical response of the sensor and the sample at the end of the first full paragraph on Page 3. We have also included Fig. R2 as Extended Data Fig. 7.

Referee #2:

Xu et al report many previously unobserved correlated insulating states in WSe_2/WS_2 moiré superlattices. The detection of this states is carried out using an original technique, with a WSe_2 monolayer separated from the main heterobilayer by an hBN spacer. The response of the 2s excited exciton state sensitive to its dielectric environment is used for detection of the insulating states in the heterobilayer. The new observations are very interesting, and in some ways represent the pinnacle of the moiré physics in TMD heterobilayers. Equally, this work opens the ways to further detailed studies of the correlated states as the authors suggest.

While the observations of the 2s state behaviour are presented, further insight in the behaviour in the WSe_2/WS_2 heterobilayer itself as well as insight in the device operation would be very helpful. With these points clarified I would be happy to recommend this work for publication in Nature.

Response 4:

We thank the referee for the positive comments and the suggestions for improvement.

(1) It is not clear why the WSe2 reflection contrast (RC) spectra measured on the device show the RC signal on the 'sensor' WSe_2 but not the WSe_2 in the heterobilayers.

Response 5:

We apologize for not explaining this point clearly in our original manuscript. The reflection contrast of the device shows the response from WSe_2 in both the sensor and sample layer, but they belong to different spectral windows. Referee #1 asked the same question. Please refer to **Response 3**.

(2) In regards to question 1, it is not entirely clear what is shown in Fig.1f. There are features below 1s exciton. What are they? Can they also tell what's happening in the heterobilayer? They seem to be not explained in detail in the paper. Why?

Response 6:

Again, we apologize for the confusion. As shown in Fig. R2, the features below the 1s exciton of the sensor are the fundamental moiré excitons in the WSe_2 layer of the moiré superlattice. Their observation has been the subject of a recent study in Nature 567, 76–80 (2019). These features exhibit clear modulations at each integer filling of the moiré superlattice. The enhancement of the exciton oscillator strength at integer fillings is related to the formation of electronic insulating states and reduced screening of the electron-hole interactions [Nature 579, 353–358 (2020)]. See **Response 7** for a discussion on the behavior of moiré excitons at fractional fillings. We would like to note that the exciton response in a doped semiconductor particularly with moiré flat bands is a complicated many-body problem. There is so far no quantitative understanding of the moiré excitons interacting with a system of strongly correlated electrons.

(3) Is there a more direct evidence of the changes occurring in the WSe_2/WS_2? In the authors' previous paper on the same heterobilayer system, where the insulating states were found for the first time (Tang et al Nature 2020), the optical response from WSe_2 in the heterobilayer was measured. Can similar data be presented here? Do fractional states show any signatures when they are measured on the actual heterobilayer? If not, please explain why.

Response 7:

Some fractional filling states can potentially be detected directly by the moiré excitons in the WSe_2/WS_2 heterobilayer. Figure R3 shows the doping-dependent reflection contrast of the main device. The features below the 1s resonance of the sensor are the fundamental moiré excitons of the WSe_2/WS_2 heterobilayer. The result is consistent with that in Nature 579, 353–358 (2020) but of higher quality. We also show a line cut (along the black dashed line) near the moiré exciton resonance at 1.66 eV (right panel). Signatures of the fractional filling states (marked by red lines) can be identified as an enhancement of the optical reflection contrast.

Not only fewer fractional filling states can be identified from the doping dependence of the moiré excitons in the WSe_2/WS_2 heterobilayer, but also the interpretation is complicated because the optical response of a doped moiré system is not well understood. In contrast, the exciton sensing technique employed in this work has substantially higher sensitivity. It directly probes the dielectric function of the correlated electron system. In the low-energy and long-wavelength limit, it is a probe of the electronic compressibility (see Methods). The result is therefore more straightforward to interpret.

In the revised manuscript, we have adopted Fig. R3 for Fig. 1f and discussed briefly the possibility of detecting fractional filling states by the moiré excitons in the second full paragraph on page 3.

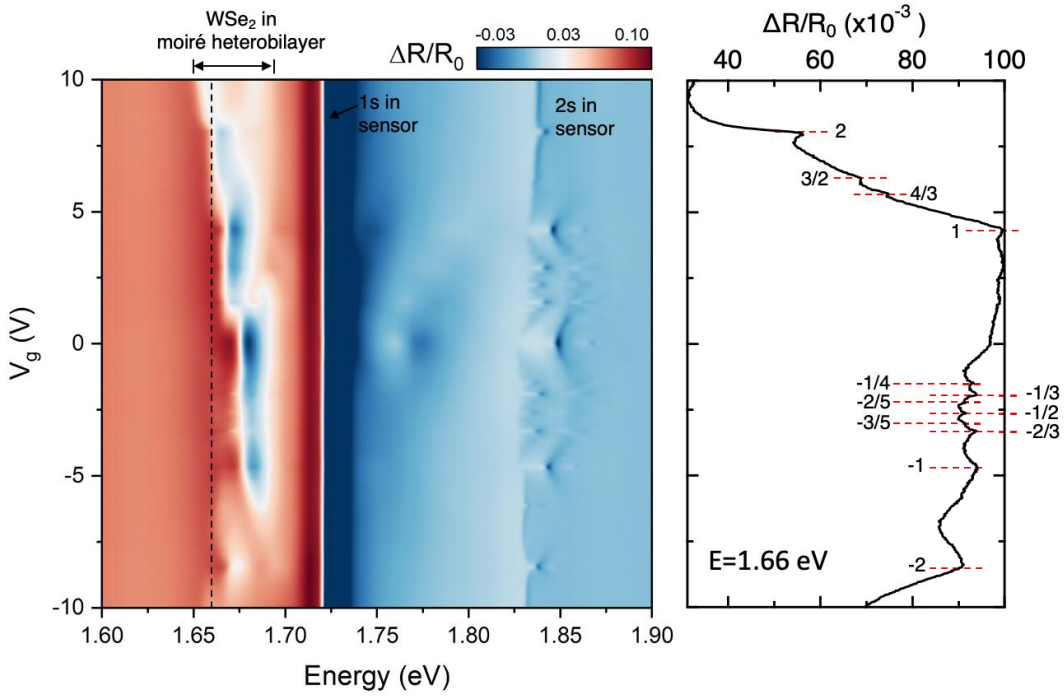

Figure R3. Gate-dependent reflection contrast spectrum at 1.6 K (left) with a line cut at 1.66 eV (right). Identifiable fractional filling states are marked by red dashed lines and filling factors.

(4) Are there any studies of control sample with the 'sensor' WSe_2 layer only? It appears that such RC measurements in these fine details and low excitation power have never been done before, and the behaviour of 2s exciton RC as a function of the gate voltage is not really known. It would be very

desirable to measure such a control sample too, or to point the readers to a similar study, where in similar conditions the 2s state is insensitive to the doping.

Response 8:

Figure R4a shows the doping-dependent reflection contrast spectrum of a WSe_2 monolayer measured with white light of a power < 1 nW. The WSe_2 monolayer is encapsulated between hBN layers. Figure R4b shows the same result in a smaller range of the reflection contrast to bring out the weaker features (The strong features are saturated). We can clearly identify the 1s, 2s and 3s exciton resonances when the sample is charge neutral. When the sample is electron or hole doped, attractive and repulsive polarons can be identified for both the 1s and 2s resonances. The result is consistent with that reported in the literature [e.g. Nat. Nanotech. 12, 144 (2017), PRX 10, 021024 (2020)].

The behavior of the WSe_2 sensor (Fig. 1f) is very different from that in Fig. R4. In that device it remains charge neutral for the entire range of applied gate voltages. This is verified by the gate-independent 1s exciton resonance of the sensor (Fig. 1f). Figure R4c illustrates the band alignment of the device. The WSe_2/WS_2 heterobilayer has a type-II band alignment. The moiré potential further reduces the WSe_2 band gap. As a result, the valence band edge of the WSe_2 sensor is above the first moiré valence band edge in WSe_2. Therefore, the negative gate voltages hole dope the WSe_2 layer in the heterobilayer, and the positive gate voltages electron dope the WS_2 layer in the heterobilayer.

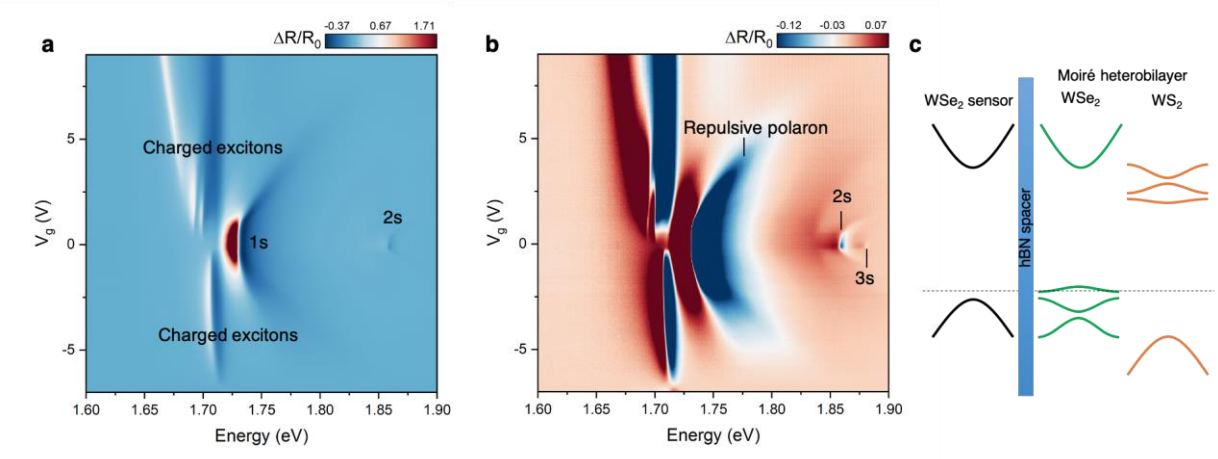

Figure R4. (a) Gate-dependent reflection contrast spectrum of a monolayer WSe_2 sample (encapsulated in hBN dielectrics and having a few-layer graphite as gate contact). (b) Same plot as (a) in a smaller reflection contrast range to visualize the 2s and 3s exciton states better. (c) Band alignment of the WSe_2 sensor and the WSe_2/WS_2 moiré heterobilayer. They are separated by a thin hBN spacer.

(5) Did the authors measure more than one sample? I do appreciate that it is a very complicated structure, which would probably take weeks and several attempts to build. I wonder still if the role of the thickness of the hBN buffer between the 'sensor' and the heterobilayer has been investigated. How have the moiré effects in the sensor WSe_2 been excluded from the picture, for example? Is the sensor layer randomly orientated? Is it important which of the layers, WS_2 or WSe_2 face the sensor?

Response 9:

We have fabricated a new device that differs from the main device in several ways. It has 1) an hBN spacer of ~ 0.65 nm, compared to ~ 1.3 nm in the main device; 2) an angle-misaligned (~ 25 degree) sensor to the moiré superlattice, compared to an angle-aligned sensor to the moiré superlattice in the main device; and 3) the sensor on the WS_2 side rather than the WSe_2 side of the heterobilayer as in the main device. Because this device has poor electrical contacts, we had to apply a constant back gate voltage of 8 V to heavily electron dope the contact region to achieve reasonably good contact. The electron density in the heterobilayer is tuned by sweeping the top gate. The top gate does not cover the contact region, which remains largely unaffected by the top gate voltage. Unfortunately we were not able to achieve good electrical contact to the sample on the hole side and could investigate this device only for the electron side.

Figure R5 shows the gate-dependent reflection contrast spectrum of the new device. Both moiré exciton resonances of the WSe_2/WS_2 heterobilayer and the 1s and 2s exciton resonances of the WSe_2 sensor can be identified in Fig. R5a. We can also identify most of the correlated insulating states from the 2s exciton states with an energy blue shift and oscillator strength enhancement (Fig. R5b). The overall reflection contrast of this device is lower than the previous one. It is due to the optical interference effect (the two devices have different thicknesses measured from the top to the back gate). Given the similarity of the results in Fig. R5 and Fig. 1f and 2 in the main text, we conclude that the main results are reproduced. We do not observe any obvious dependence on sensor-sample angle alignment and order of the layer arrangement.

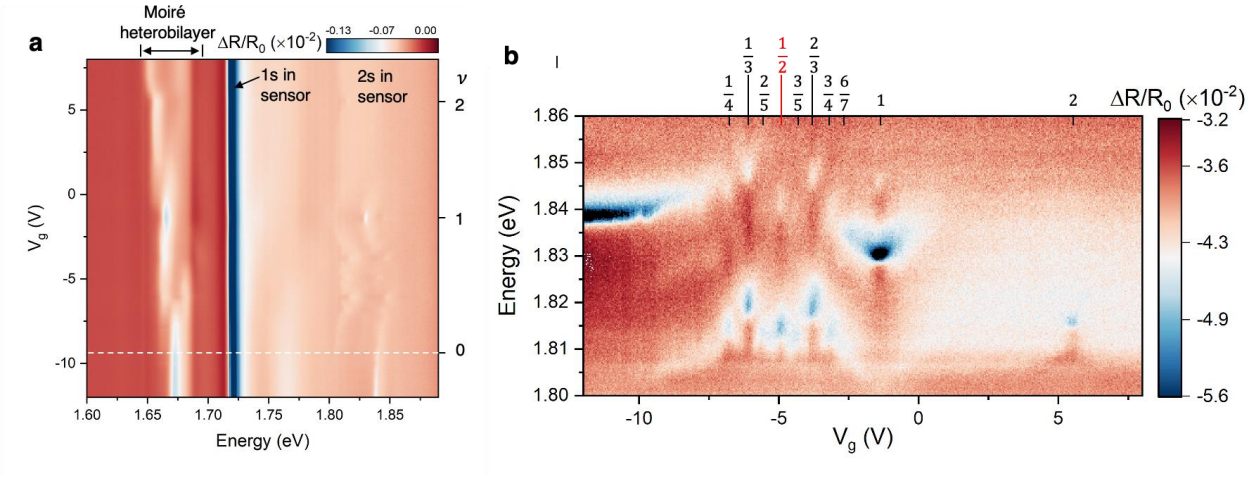

Figure R5. (a) Gate-dependent reflection contrast spectrum of a new device at 1.6 K. (b) Same data as (a) focusing on the 2s transition of the sensor. A fix back gate voltage of 8 V is applied to make good contact to the heterobilayer.

We discuss the role of the hBN spacer thickness. The 2s resonance energy shift is expected to decrease with increasing spacer thickness since screening of the electron-hole interaction in the sensor becomes less effective. This is verified in our experiment. For instance, the 2s resonance energy shift between the $\upsilon = 1$ insulating state and the compressible state immediately above $\upsilon = 1$ is ~ 12 meV in the main device. In this new device with a thinner spacer, the shift is over 20 meV. In the new device, the sensor is on the side of electron-doped WS_2, which further reduces the effective sensor-sample distance. Given the 2s exciton Bohr radius of ~ 5 nm in monolayer WSe_2, we

expect a significant drop in the sensor sensitivity when the spacer thickness becomes comparable to 5 nm. A systematic spacer thickness study is an interesting problem and deserves further investigation, but is beyond the scope of the current study.

The usual moiré effects on the sensor are ruled out by the independence of the results on the alignment angle between the sensor and sample, and on which side of the heterostructure the sensor faces. The result is not surprising because the usual moiré potential is originated from spatially dependent interlayer carrier hopping between two layers. The interlayer carrier hopping is expected to decay exponentially with layer separation on the atomic length scale.

We have included Fig. R5 as Extended Data Fig. 4 in our revised manuscript. We have also briefly discussed the effect of the spacer thickness and layer alignment in Methods (Effect of the sensor orientation and location).

Referee #3:

The manuscript of Xu et al. reports abundance of new correlated states at fractional fillings of WSe_2/WS_2 moiré system. By employing a local dielectric sensor (single-layer WSe2) in proximity to WSe2/WS2 bilayer, Xu et al. manage to resolve many more fractional filling states, compared to a recent work reported by the Berkeley group (Regan, E. C. et al, Nature (2020)). Despite not being the first experimental work addressing fractional filling correlated states in this system, the work of Xu et al. is still significant in following ways.

Scientifically, the cascades of correlated states observed here is fascinating. It makes us understand better the nature of correlated states in moiré system which is intensively studied in recent years.

Technically, detecting dielectric environment (different electronic phases) with exciton states could also be potentially applied to other materials and thus could attract a wide range of interests.

In general, the experimental idea, perspective and results in this manuscript make me a positive impression. I would recommend its publication in Nature if my following concerns can be properly addressed.

Response 10:

We thank the referee for the positive assessment of our work and detailed suggestions for improvement.

(1) In Fig. 1c, the 2s states of WSe_2 shows similar signatures (shifted to higher energy above white dashed line) like 1s states in WS_2. I would suggest the authors intercept these features better.

Response 11:

We thank the referee for the suggestion. Referee #1 and #2 raised similar questions. Please refer to **Response 2**.

(2) In Fig. 1, all figures are arranged clockwise. Perhaps the authors should rearrange them in normal way to make readers easier to follow.

Response 12:

We thank the referee for the suggestion and have modified Fig. 1 accordingly.

(3) In line 165-166, it is vague by saying “The electron and hole sides behave similarly, suggesting that the same physics is in play”. It should be better addressed.

Response 13:

We have changed the sentence to make the meaning clearer. It now reads as the following:
“The electron and hole sides display almost identical correlated insulating states with similar energy scales, suggesting that the same correlation effect is in play”.

(4) I don´t find the assignment of the filling factor in Fig.2 convincing enough. i.e., 1) I cannot see any reliable signatures of 1/7 and 6/7 states. 2) the gate voltage difference between -1/3 and -1/4 states

is almost 30 percent larger than that between 1/3 and 1/4 states, etc. More statistics (i.e. more datasets taken from different regions of the device or a second device) is helpful.

Response 14:
We thank the reviewer for the suggestion. We have performed measurements on different regions of the device and also on a different device. Please see **Response 9** for results on the second device. Figure R6 shows the gate-dependent reflection contrast near the 2s resonance of the sensor at six different locations spread over the entire device during two different cool-downs (The device size is ~12 μm by 12 μm, and the spatial resolution of our optical measurements is ~ 1 μm). Here P0 is the same dataset as presented in Fig. 2 of the main text. The data quality varies from location to location, but most fractional states are still discernable except in P5, where the weaker states are smeared out.

The ν = 1/7 and 6/7 states are the weakest with the lowest T_c ~ 10-13 K among the group of states reported in this work. They also sit on a large background from the nearby strong insulating states (ν = 0 and 1), which makes their identification by the 2s exciton feature more difficult. However, a consistent increase of the oscillator strength at these fillings in P0-P4 can be identified, particularly, at a higher photon energy (e.g. ~ 1.855 eV in P0). It adds to the oscillator strength of the 2s exciton from the ν = 0 (or 1) state and makes the feature asymmetric about ν = 0 (or 1). At higher temperatures after melting of the 1/7 and 6/7 states (Fig. 3), the 2s exciton at ν = 0 and 1 becomes symmetric again. Future experiments on samples of higher quality and at lower temperature should help to better resolve the 1/7 and 6/7 states.

We briefly comment on the effect of sample inhomogeneities. More discussions are provided in **Response 15**. The insulating states have a finite width in gate voltage or doping density. The width can be used to characterize charge inhomogeneity in the heterobilayer within the probe beam area. Figure R6 shows that the level of charge inhomogeneities varies significantly throughout the device. In addition, we also observe small variations in the 2s exciton energy for the same state at different locations. This presumably arises from spatial strain variation in the senor layer since the exciton energy is sensitive to strain.

The reviewer made a good catch on the difference in gate voltage spacing between the ν = 1/4 and 1/3 states on the two doping sides. We believe that we are seeing a nonlinear gating effect in the limit of low doping density, particularly, on the hole doping side. In general, it's harder to achieve good electrical contact to the sample for hole doping than electron doping with our current choice of the contact metal. On the hole side at low density, the gating efficiency is smaller than the average value of 0.25 filling per volt. This trend is observed throughout the datasets in Fig. R6.

The nonlinear gating effect certainly complicates the assignment of low-filling states, particularly on the hole doping side. However, the particle-hole symmetry about $\nu = 1/2$ observed in our experiment and supported by our modeling helps to determine the low-filling states. (For instance, the 1/3 and 2/3 states have similar T_c's and display similar amount of 2s spectral shift. The same is true for the 1/4 and 3/4 states and the other particle-hole symmetric states.) Since the states above $\nu = 1/2$ do not suffer from the nonlinear gating effect, we first assign ν for these states, and then

assign 1- ν for the corresponding states with the same amount of 2s spectral shift. In future studies, different contact metals will be explored to achieve better contacts.

In our revised Methods (Assignment of the filling factor for the insulating states), we have addressed the issue of nonlinear gating and discussed the use of particle-hole symmetry in assigning small filling factors. Figure R6 (except dataset P0 that has been presented in Fig. 1 and 2) is now included as Extended Data Fig. 3 in our revised manuscript.

(5) Last but not latest, I would suggest the authors to rule out the possibilities that some of the correlated states resolved might come from the in-homogeneity of twist angles or charged impurities.

Response 15:
We agree that it is extremely important to rule out any possible artifacts in assigning the correlated states. Observing almost all of the correlated states simultaneously at different regions of the device suggests that the observed correlated states are likely intrinsic rather than induced by sample inhomogeneities. Since the optical measurements probe the average properties of a region ~ 1 μm, inhomogeneities tend to smear out the correlated states as illustrated in Fig. R6, where the correlated states at lower quality locations (e.g. P5) display a wider distribution in filling factor. One can use the width in filling as a measure of inhomogeneity. We estimate a typical full width of about 0.05 filling from the dataset in Fig. 2a. It is smaller than the spacing between the correlated states, allowing us to resolve them (see Fig. 2b, where the width of each state is taken to be 0.05).

In addition, the moiré period or density in angle-aligned WSe_2/WS_2 heterobilayers that are studied here are not sensitive to twist angle because there is a large (~ 4%) lattice mismatch between the two materials. This is in sharp contrast to the twisted homobilayers (Fig. R7). We typically have less than 0.5-degree twist angle variation in our heterobilayers, which corresponds to a ~ 3% variation in moiré period (or ~ 6% in moiré density). This is on par with the observed width of the correlated states.

Moreover, the observed correlated states have different energy scales or T_c's, which are correlated with the 2s spectral shift in the sensor (Fig. 2b and 2c). These states exhibit ordering in energy or T_c that is nearly symmetric about $\nu = 1/2$. Such a characteristic ordering in energy is also in good agreement with our Monte Carlo simulations. All of these pieces together show that the observed correlated states are distinct states. If they were the same/similar states induced by sample inhomogeneities, we wouldn't expect such systematic scaling.

We believe that there is sufficient evidence to rule out the possibilities that some of the correlated states come from the sample inhomogeneity. In the revised manuscript, we have added a new section in Methods (Effects of sample inhomogeneity) to address the effect of sample inhomogeneities on the observed correlated states.

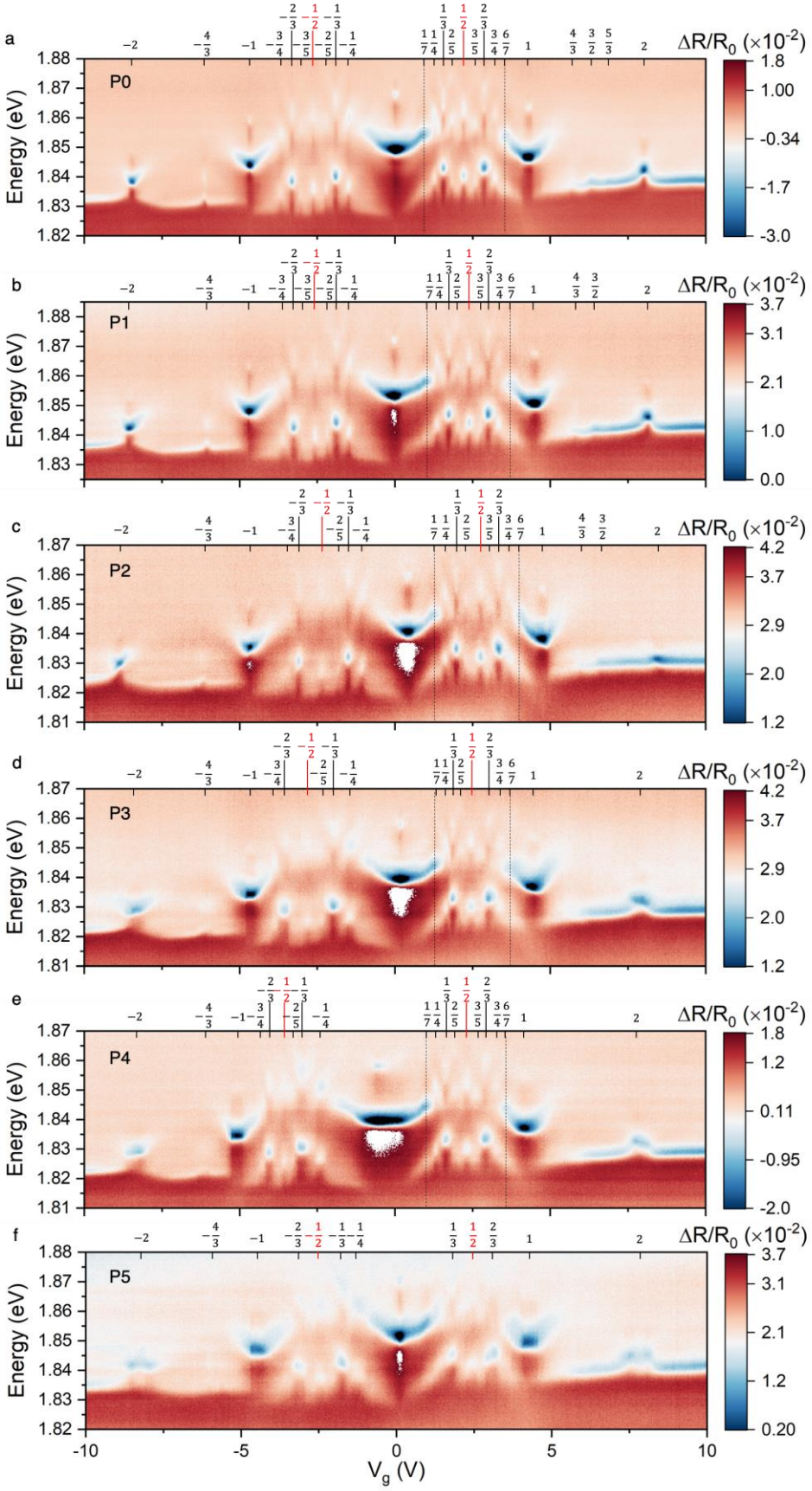
a
P0
b
P1
c
P2
d
P3
e
P4
f
P5
Energy (eV)
ΔR/R0 (×10-2)
Vg (V)

Figure R6. (a-h) Gate-dependent reflection contrast spectrum measured at different regions P0-P5 at T=1.6 K. The figures share the same x-axis on the bottom. The filling factors for the identifiable states are labeled on the top axis.

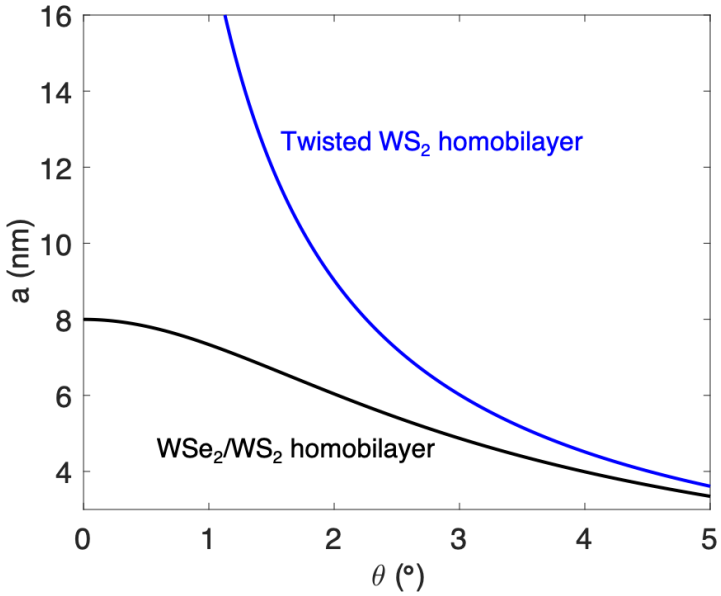

Figure R7. Comparison of the moiré superlattice constant a as a function of twist angle θ in WSe_2/WS_2 heterobilayers and twisted WS_2 homobilayers.

Reviewer Reports on the First Revision:

Referee #1 (Remarks to the Author):

I have read through the response letter and the revise manuscript. In my opinion, the authors have thoroughly addressed referees' comments and improved the manuscript accordingly. Therefore, I think that the manuscript is suitable for publication in its current form.

Referee #2 (Remarks to the Author):

The authors provided comprehensive answers to all questions of the referees. Additional detailed data sets have been provided, that give further insight in both the device operation and the physics of the insulating states. This work is of a very high quality and significance. The paper has been improved following the revision, and made much more accessible to readers. I recommend this paper in its current form for publication in Nature.

Referee #3 (Remarks to the Author):

The revised manuscript is of very high quality. All my previous comments have been properly addressed. I support its publication now.