**Bosonic Mott insulator in WSe\(_2\)/WS\(_2\) moiré superlattice**

Richen Xiong\(^1\), Jacob H. Nie\(^1\), Samuel L. Brantly\(^1\), Patrick Hays\(^2\), Renee Sailus\(^2\), Kenji Watanabe\(^3\), Takashi Taniguchi\(^4\), Sefaattin Tongay\(^2\), Chenhao Jin\(^1\)*

\(^1\)Department of Physics, University of California at Santa Barbara; Santa Barbara, CA 93106, USA

\(^2\)School for Engineering of Matter, Transport, and Energy, Arizona State University; Tempe 85287 AZ, USA.

\(^3\)Research Center for Functional Materials, National Institute for Materials Science; 1-1 Namiki, Tsukuba 305-0044, Japan.

\(^4\)International Center for Materials Nanoarchitectonics, National Institute for Materials Science; 1-1 Namiki, Tsukuba 305-0044, Japan

* Corresponding author. Email: jinchenhao@ucsb.edu

**Abstract:** A panoply of unconventional electronic states is recently observed in moiré superlattices. On the other hand, similar opportunities to engineer bosonic phases remain largely unexplored. Here we report the observation of a bosonic Mott insulator in WSe\(_2\)/WS\(_2\) moiré superlattices composed of excitons, i.e., tightly bound electron-hole pairs. Using a novel pump probe spectroscopy, we find an exciton incompressible state at exciton filling \(\nu_{\text{ex}} = 1\) and charge neutrality, which we assign to a bosonic Mott insulator. When further varying charge density, the bosonic Mott insulator continuously transitions into an electron Mott insulator at charge filling \(\nu_e = 1\), suggesting a mixed Mott insulating state in between. Our observations are well captured by a mixed Hubbard model involving both fermionic and bosonic components, from which we extract the on-site Coulomb repulsion to be 15meV and 35meV for exciton-exciton and electron-exciton interactions, respectively. Our studies establish semiconducting moiré superlattices as intriguing platforms for engineering novel bosonic phases.
Main Text:

Strongly correlated phases emerge when many body interaction dominates over kinetic energy in flat-band systems(1, 2). Semiconducting transition metal dichalcogenide (TMDC) moiré superlattices offer a unique platform that hosts both fermionic and bosonic quasi-particles -- charges(3–12) and excitons(13–19) -- with flat-bands. Intriguing electronic phases are reported from the fermionic sector, such as Mott and Wigner crystal states(3–8), stripe phase(9), continuous Mott transition(10, 11), and quantum anomalous Hall insulator(12). Recently, increasing efforts are put into bringing the bosonic sector into play. Examples include exciton-mediated ferromagnetism(20) as well as excitonic insulator(21, 22) -- an insulator for charge but “metal” for exciton, where bosonic excitons coexist with and affect correlated electrons. In all studies so far, however, the bosonic sector is in a compressible fluid state, and strongly correlated phases of bosons remain elusive.

Here we create and identify a bosonic Mott insulator consisting of interlayer excitons in a 60-degree-aligned WSe₂/WS₂ moiré superlattice. Each interlayer exciton contains an electron in the WS₂ layer and a hole in WSe₂ with a large binding energy of hundreds of meV(23), which is the ground state exciton configuration of a type II heterostructure (Fig. 1a). We find an exciton incompressible state at ν_{ex} = 1, i.e., one exciton per moiré site, a hallmark of a bosonic correlated insulator. We further study the phase diagram spanned by ν_e and ν_{ex} and observe a mixed Mott insulator along the path of ν_{tot} = ν_{ex} + ν_e = 1. Owing to the large exciton binding energy and strong correlation here, the bosonic Mott insulator persists to above 30 K, orders of magnitude higher than previous studies in cold atoms and quantum wells(24, 25). Our results highlight semiconducting moiré superlattices as an attractive platform for engineering novel bosonic phases at high temperature, as well as for constructing a complete picture of many-body physics with interacting fermions and bosons.

Figure 1b illustrates the pump-probe spectroscopy used in our study. The relatively strong pump light tunes the background exciton density, while the weak probe light injects a small number of additional excitons and detects their responses. The concept is analogous to electronic capacitance measurement of compressibility, where a DC “pump” gate voltage tunes the background charge density and a small AC “probe” voltage slightly modulates the charge density. In both cases, the exciton/charge compressibility can be directly obtained from the minimum energy it takes to add one more particle on top of a given background particle density. Importantly, a DC “pump” is necessary to maintain a stable background particle density and a well-defined ground state, while the “probe” needs to be AC modulated to isolate the responses of particles created by the probe. This scheme is distinctive from conventional optical pump-probe measurements that modulate the pump beam to investigate pump induced changes.

With the capability of tuning charge and exciton density through electrostatic gating and pump light respectively, we fully explore the phase diagram spanned by the charge filling ν_e and exciton filling ν_{ex}. We start with the axes, i.e., ν_{ex} = 0 or ν_e = 0. Figure 1c & e shows the PL and absorption spectra of the moiré bilayer at ν_{ex} = 0 (zero pump intensity) and ν_e ≥ 0 (see Fig. S1 for complete doping dependence). At charge neutrality, the PL features a single peak at 1.43eV from interlayer exciton emission(6, 8), while the absorption shows three peaks from moiré intralayer excitons(13). At ν_e = 1 and 2, the emission peak blueshifts suddenly, and the absorption peaks show a kink. These features originate from the emergence of insulating states
at integer electron fillings, which has been independently confirmed by capacitance and microwave impedance measurements(3, 6). While the exact mechanisms of these spectra changes are worth further discussion, which we will provide later, the PL energy jump at $\nu_e = 1$ can be intuitively understood from the emergence of a fermionic Mott insulator state where all available sites are occupied by one electron. Any additional excitons injected are therefore forced into a higher energy state.

Now we turn to the case of $\nu_e = 0$ by fixing the gate voltage $V_g$ at charge-neutral -0.5V. Fig. 1d & f show the dependence of pump-probe PL and absorption spectra on the pump light intensity, which effectively controls $v_{\text{ex}}$. To account for the non-linear dependence of $v_{\text{ex}}$ on pump intensity, we also show dipolar-interaction-induced interlayer exciton energy shift $\Delta_{\text{dipole}}$, which is approximately proportional to $v_{\text{ex}}$(26–28). Interestingly, a jump in the PL energy is also observed. This jump is well reproduced in another 60-degree-aligned moiré bilayer but absent in a slightly misaligned bilayer(26), indicating its origin from correlation effects. The qualitative similarity to the gate-dependent PL suggests a similar origin behind the exciton energy jump: the emergence of a particle lattice that occupies all available sites. The most natural candidates are interlayer excitons, which are the immediate products of pump light absorption and interlayer charge transfer in a type II heterojunction. To elucidate nature of the lattice, we compare its effects on PL and absorption spectra to those from an electron lattice. PL spectra probe interlayer exciton responses. While both the gate-injected electron lattice (Fig. 1c) and pump-injected lattice (Fig. 1d) lead to a jump in interlayer exciton energy in PL, the amplitudes are quite different: 35 meV and 15 meV, respectively. A more prominent difference is observed in the absorption spectra, which examine how intralayer excitons respond to the injected lattices. Electrons induce rich features, such as shifting, merging and splitting of exciton resonances (Fig. 1e). In contrast, the pump-injected particles have rather weak effects on intralayer excitons, only slightly decreasing their oscillator strength (Fig. 1f). These distinctive behaviors indicate that the pump-injected lattice is not formed by electrons.

To further confirm the exciton nature of the pump-injected lattice, we move away from the axes and investigate the phase diagram at $v_{\text{ex}} > 0$ and $\nu_e > 0$. Fig. 2a shows a set of pump intensity-dependent PL spectra at different gate voltages, corresponding to increasing electron density in the WS$_2$ layer. All plots have qualitatively similar behaviors – interlayer exciton energy shows a jump. Upon increasing $\nu_e$, the jump occurs earlier and earlier and eventually appears at zero pump intensity when $\nu_e = 1$ ($V_g = 0.8$ V), consistent with the fact that gate-injected electrons have already occupied all sites at $\nu_e = 1$. Based on these observations, the pump-injected particles will cooperate with electrons in WS$_2$ to occupy available sites and should therefore contain electrons. Besides an electron itself, which has been already excluded, the only other candidate is the interlayer exciton composed of an electron in the WS$_2$ layer and a hole in the WSe$_2$ layer.

We, therefore, conclude that the pump light is creating interlayer excitons and tuning $v_{\text{ex}}$. At charge neutrality, the observed jump in PL spectra then corresponds to a sudden increase in the interlayer exciton energy when increasing its density, i.e., an incompressible state of the interlayer exciton. Moreover, this state connects smoothly into the $\nu_e = 1$ electron Mott insulator, indicating one particle per moiré site along the entire path. Our observation of an incompressible exciton state at $v_{\text{ex}} = 1$ is a hallmark of a correlated insulator purely made of
excitons, which we assign to a bosonic Mott insulator (29, 30).

To quantify the jump in interlayer exciton energy, we fit PL spectra with two Lorentzian peaks (26) and compute the exciton energy change \( \Delta E_{\text{ex}} = [(E_1 I_1 + E_2 I_2)/(I_1 + I_2) - E_3] \), which can be considered as an effective exciton chemical potential. Here \( E_1 \) (\( E_2 \)) and \( I_1 \) (\( I_2 \)) correspond to the energy and amplitude of the PL peak that dominates at low (high) pump intensity, labeled as peaks I and II, respectively. Fig. 2c summarizes the evolution of \( \Delta E_{\text{ex}} \) with pump intensity at representative gating, where a transition in exciton energy is clearly observed at all gate voltages. The transition appears not particularly sharp around charge neutrality \((V_g = -0.4V)\), largely due to the non-linear dependence of exciton density on pump intensity (Fig. 1d). In addition, the transition is broadened by spatial inhomogeneity in the exciton density that is expected to be much larger than the charge case (26). We determine the position of \( \nu_{\text{ex}} = 1 \) from the middle point of the \( \Delta E_{\text{ex}} \) transition, as labeled by green triangles in Fig. 2c. We also independently estimate the exciton density at this point to be \( \sim 10^{12} \text{ cm}^{-2} \) from \( \Delta_{\text{dipole}} \) (26–28). At finite electron density, the transition happens when electrons and excitons cooperate to occupy all available sites. The transition points therefore correspond to \( \nu_{\text{tot}} = \nu_e + \nu_{\text{ex}} = 1 \) and keep shifting to lower pump intensity until reaching \( \nu_{\text{ex}} = 0 \) when \( \nu_e = 1 \).

To separate \( \nu_e \) and \( \nu_{\text{ex}} \) better, we also measure absorption spectra of the moiré superlattice while varying gate voltage and pump intensity. Because intralayer excitons are only sensitive to \( \nu_e \) but not \( \nu_{\text{ex}} \) (Fig. 1, e and f), we use spectral changes in absorption to independently determine \( \nu_e \). Fig. 2b shows the gate-dependent absorption spectra of the moiré bilayer at different pump intensities. All plots show similar behaviors except for a slight shift in gate voltage. This can be seen more clearly in the gate-differentiated absorption spectra at representative pump intensity (Fig. 2d) (26). Blue triangles in Fig. 2d and 2b label the position of “kinks” that correspond to \( \nu_e = 1 \).

With both \( \nu_{\text{tot}} \) and \( \nu_e \) extracted, Fig. 3a summarizes the phase diagram spanned by \( \nu_e \) and \( \nu_{\text{ex}} \) at base temperature of 1.65 K. The color scale represents \( \Delta E_{\text{ex}} \), on which the boundary of \( \nu_{\text{tot}} = 1 \) and \( \nu_e = 1 \) are overlaid. The \( \nu_e = 1 \) line shifts slightly towards lower gate voltage at high pump intensity, presumably from photocarriers generated during the relaxation of excitons. Nevertheless, such effect is rather weak, and the \( \nu_e = 1 \) line deviates far from the \( \nu_{\text{tot}} = 1 \) line where transitions in \( \Delta E_{\text{ex}} \) are observed. This further confirms that the transition at \( \nu_{\text{tot}} = 1 \) is not from an electron lattice. Instead, it originates from a mixed lattice composed of both excitons and electrons. Our observations, therefore, suggest a mixed Mott insulator along the \( \nu_{\text{tot}} = 1 \) line, which smoothly connects into a bosonic (fermionic) Mott insulator at the end point of \( \nu_{\text{ex}} = 1 \) (\( \nu_e = 1 \)).

We further obtain the phase diagrams at 15 and 30 K (Fig. 3b and c, see Fig. S4-S7 for details). A sharp transition in \( \Delta E_{\text{ex}} \) is always observed at \( \nu_e = 1 \), consistent with the strong charge correlation and a high melting temperature of charge Mott insulator in WSe\(_2\)/WS\(_2\) moiré superlattices (3, 4). Similarly, the behavior at \( \nu_{\text{ex}} = 1 \) remains largely unchanged with temperature, indicating survival of the bosonic Mott insulator to above 30 K. In contrast, for the regions in between, the transition in \( \Delta E_{\text{ex}} \) becomes much slower than at base temperature. This can be seen clearly by comparing the \( \Delta E_{\text{ex}} \) evolution at charge neutral \((V_g = -0.5V)\) and finite electron density \((V_g = -0.1V)\), as shown in Fig. 3e-f. At base temperature the finite \( \nu_e \) case shows a sharper rise than charge neutral (Fig. 3e), which can be naturally explained by the smaller \( \nu_{\text{ex}} \).
needed to reach $v_{\text{tot}} = 1$ and therefore less inhomogeneous broadening from exciton density. Interestingly, at 30K the rise in $\Delta E_{\text{ex}}$ becomes smoother at $V_g = -0.1V$ compared to at charge neutral, indicating increasing exciton compressibility and partial melting of the mixed Mott insulator. This observation suggests that the mix Mott insulators is less stable than both components(26).

Discussion

Our experimental results are well captured by a two-component Hubbard model with both fermionic and bosonic species(26). Fig. 4a summarizes the predicted $\Delta E_{\text{ex}}$ in the large $U$ limit using $U_{\text{ex-ex}} < U_{\text{e-ex}} < U_{\text{e-e}}$, where $U_{\text{ex-ex}}$, $U_{\text{e-e}}$ and $U_{\text{e-ex}}$ are on-site repulsive energies between excitons, electrons and exciton-electron, respectively. The prediction matches well with the experiment (Fig. 3a). Along the horizontal axis ($v_{\text{ex}} = 0$), $\Delta E_{\text{ex}}$ shows a sudden jump from 0 to $U_{\text{e-ex}}$ at $v_e = 1$. This can be understood as excitons always avoid electron-occupied sites due to the on-site electron-exciton repulsion (Fig. 4b) until all sites are electron-occupied at $v_e = 1$, after which adding an exciton necessarily pays the additional energy cost of $U_{\text{e-ex}}$ (Fig. 4d). The prediction matches well with experimental results (Fig. 1c), allowing us to determine $U_{\text{e-ex}}$ to be 35meV. Such doping dependence is distinctively different from that in monolayer TMDC or TMDC bilayers where strong correlation is not observed, such as WSe$_2$/MoSe$_2$(31, 32). In these systems, electron doping immediately leads to the formation of trion and a corresponding emission peak in PL at lower energy(31, 32). Here in WSe$_2$/WS$_2$, in contrast, the emission spectrum remains largely unchanged until $v_e = 1$, after which a higher energy state takes over.

Similarly, $\Delta E_{\text{ex}}$ remains 0 along the vertical axis until $v_{\text{ex}} = 1$, after which adding an exciton pays additional energy cost of $U_{\text{ex-ex}}$ (Fig. 4c). We thereby determine $U_{\text{ex-ex}}$ to be 15meV (Fig. 1d). Away from the axes, $\Delta E_{\text{ex}}$ remains 0 for $v_{\text{ex}} + v_e < 1$ (region I in Fig. 4a) since added excitons can find an empty site to avoid on-site repulsion. For $v_{\text{ex}} + v_e \geq 1$ but $v_e < 1$ (region II), added excitons will prefer to stay on an exciton site since $U_{\text{ex-ex}} < U_{\text{e-ex}}$, and the additional energy cost is $\Delta E_{\text{ex}} = U_{\text{ex-ex}}$. For $v_e \geq 1$ (region III), all sites are already occupied by electrons and added exciton can only reside an electron site; therefore $\Delta E_{\text{ex}} = U_{\text{e-ex}}$.

The above phase diagram ignores the hopping term or the two pseudospins of excitons from the $K$ and $K'$ valleys(23). Nevertheless, it captures all salient features of the experiments. Our results demonstrate semiconducting moiré superlattices as a unique platform to investigate two-component Hubbard model with bosonic and fermionic species and pave the way for investigating novel bosonic phases based on moiré excitons, such as exciton valley pseudospin order and pseudospin liquid, bosonic Mott insulator - superfluidity transition(29) , exciton-mediated superconductivity(33) and topological excitons(18, 19).

References

1. D. M. Kennes, M. Claassen, L. Xian, A. Georges, A. J. Millis, J. Hone, C. R. Dean, D. N. Basov, A. N. Pasupathy, A. Rubio, Moiré heterostructures as a condensed-matter quantum simulator. Nature Physics. 17 (2021), pp. 155–163.
2. Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, P. Jarillo-Herrero, Unconventional superconductivity in magic-angle graphene superlattices. Nature. 556, 43–50 (2018).
3. E. C. Regan, D. Wang, C. Jin, M. I. Bakti Utama, B. Gao, X. Wei, S. Zhao, W. Zhao, Z. Zhang, K.
Yumigeta, M. Blei, J. D. Carlström, K. Watanabe, T. Taniguchi, S. Tongay, M. Crommie, A. Zettl, F. Wang, Mott and generalized Wigner crystal states in WSe2/WS2 moiré superlattices. *Nature*. **579**, 359–363 (2020).

4. Y. Tang, L. Li, T. Li, Y. Xu, S. Liu, K. Barmak, K. Watanabe, T. Taniguchi, A. H. MacDonald, J. Shan, K. F. Mak, Simulation of Hubbard model physics in WSe2/WS2 moiré superlattices. *Nature*. **579**, 355–358 (2020).

5. Y. Shimazaki, I. Schwartz, K. Watanabe, T. Taniguchi, M. Kroner, A. İmamoğlu, Strongly correlated electrons and hybrid excitons in a moiré heterostructure. *Nature*. **580**, 472–477 (2020).

6. S. Miao, T. Wang, X. Huang, D. Chen, Z. Lian, C. Wang, M. Blei, T. Taniguchi, K. Watanabe, S. Tongay, Z. Wang, D. Xiao, Y. T. Cui, S. F. Shi, Strong interaction between interlayer excitons and correlated electrons in WSe2/WS2 moiré superlattice. *Nature Communications*. **12** (2021), doi:10.1038/s41467-021-23732-6.

7. X. Huang, T. Wang, S. Miao, C. Wang, Z. Li, Z. Lian, T. Taniguchi, K. Watanabe, S. Okamoto, D. Xiao, S. F. Shi, Y. T. Cui, Correlated insulating states at fractional fillings of the WS2/WSe2 moiré lattice. *Nature Physics*. **17**, 715–719 (2021).

8. E. Liu, T. Taniguchi, K. Watanabe, N. M. Gabor, Y. T. Cui, C. H. Lui, Excitonic and Valley-Polarization Signatures of Fractional Correlated Electronic Phases in a WSe2/WS2 Moiré Superlattice. *Physical Review Letters*. **127** (2021), doi:10.1103/PhysRevLett.127.037402.

9. C. Jin, Z. Tao, T. Li, Y. Xu, Y. Tang, J. Zhu, S. Liu, K. Watanabe, T. Taniguchi, J. C. Hone, L. Fu, J. Shan, K. F. Mak, Stripe phases in WSe2/WS2 moiré superlattices. *Nature Materials*. **20**, 940–944 (2021).

10. A. Ghiotto, E. M. Shih, G. S. S. G. Pereira, D. A. Rhodes, B. Kim, J. Zang, A. J. Millis, K. Watanabe, T. Taniguchi, J. C. Hone, L. Wang, C. R. Dean, A. N. Pasupathy, Quantum criticality in twisted transition metal dichalcogenides. *Nature*. **597**, 345–349 (2021).

11. T. Li, S. Jiang, L. Li, Y. Zhang, K. Kang, J. Zhu, K. Watanabe, T. Taniguchi, D. Chowdhury, L. Fu, J. Shan, K. F. Mak, Continuous Mott transition in semiconductor moiré superlattices. *Nature*. **597**, 350–354 (2021).

12. T. Li, S. Jiang, B. Shen, Y. Zhang, L. Li, Z. Tao, T. Devakul, K. Watanabe, T. Taniguchi, L. Fu, J. Shan, K. F. Mak, Quantum anomalous Hall effect from intertwined moiré bands. *Nature*. **600**, 641–646 (2021).

13. C. Jin, E. C. Regan, A. Yan, M. Iqbal Bakti Utama, D. Wang, S. Zhao, Y. Qin, S. Yang, Z. Zheng, S. Shi, K. Watanabe, T. Taniguchi, S. Tongay, A. Zettl, F. Wang, Observation of moiré excitons in WSe2/WS2 heterostructure superlattices. *Nature*. **567**, 76–80 (2019).

14. K. L. Seyler, P. Rivera, H. Yu, N. P. Wilson, E. L. Ray, D. G. Mandrus, J. Yan, W. Yao, X. Xu, Signatures of moiré-trapped valley excitons in MoSe2/WSe2 heterobilayers. *Nature*. **567**, 66–70 (2019).

15. K. Tran, G. Moody, F. Wu, X. Lu, J. Choi, K. Kim, A. Rai, D. A. Sanchez, J. Quan, A. Singh, J. Embley, A. Zepeda, M. Campbell, T. Autry, T. Taniguchi, K. Watanabe, N. Lu, S. K. Banerjee, K. L. Silverman, S. Kim, E. Tutuc, L. Yang, A. H. MacDonald, X. Li, Evidence for moiré excitons in van der Waals heterostructures. *Nature*. **567**, 71–75 (2019).

16. E. M. Alexeev, D. A. Ruiz-Tijerina, M. Danovich, M. J. Hamer, D. J. Terry, P. K. Nayak, S. Ahn, S. Pak, J. Lee, J. I. Sohn, M. R. Molas, M. Koperski, K. Watanabe, T. Taniguchi, K. S. Novoselov, R. v. Gorbachev, H. S. Shin, V. I. Fal’ko, A. I. Tartakovskii, Resonantly hybridized excitons in moiré superlattices in van der Waals heterostructures. *Nature*. **567**, 81–86 (2019).
17. L. Zhang, F. Wu, S. Hou, Z. Zhang, Y. H. Chou, K. Watanabe, T. Taniguchi, S. R. Forrest, H. Deng, Van der Waals heterostructure polaritons with moiré-induced nonlinearity. *Nature*. 591, 61–65 (2021).

18. H. Yu, G.-B. Liu, J. Tang, X. Xu, W. Yao, Moiré excitons: From programmable quantum emitter arrays to spin-orbit–coupled artificial lattices. *Science Advances*. 3 (2017), doi:10.1126/sciadv.1701696.

19. F. Wu, T. Lovorn, A. H. Macdonald, Topological Exciton Bands in Moiré Heterojunctions. *Physical Review Letters*. 118 (2017), doi:10.1103/PhysRevLett.118.147401.

20. X. Wang, C. Xiao, H. Park, J. Zhu, C. Wang, T. Taniguchi, K. Watanabe, J. Yan, D. Xiao, D. R. Gamelin, W. Yao, X. Xu, Light-induced ferromagnetism in moiré superlattices. *Nature*. 604, 468–473 (2022).

21. J. Gu, L. Ma, S. Liu, K. Watanabe, T. Taniguchi, J. C. Hone, J. Shan, K. F. Mak, Dipolar excitonic insulator in a moiré lattice. *Nature Physics*. 18, 395–400 (2022).

22. Z. Zhang, E. C. Regan, D. Wang, W. Zhao, S. Wang, M. Sayyyad, K. Yumigeta, K. Watanabe, T. Taniguchi, S. Tongay, A. Zettl, M. P. Zaletel, F. Wang, Correlated interlayer exciton insulator in double layers of monolayer WSe2 and moiré WS2/WSe2 (2021) (available at http://arxiv.org/abs/2108.07131).

23. P. Rivera, H. Yu, K. L. Seyler, N. P. Wilson, W. Yao, X. Xu, Interlayer valley excitons in heterobilayers of transition metal dichalcogenides. *Nature Nanotechnology*. 13, 1004–1015 (2018).

24. S. Sugawa, K. Inaba, S. Taie, R. Yamazaki, M. Yamashita, Y. Takahashi, Interaction and filling-induced quantum phases of dual Mott insulators of bosons and fermions. *Nature Physics*. 7, 642–648 (2011).

25. C. Lagoin, S. Suffit, K. Baldwin, L. Pfeiffer, F. Dubin, Mott insulator of strongly interacting two-dimensional semiconductor excitons. *Nature Physics*. 18, 149–153 (2022).

26. See Methods and Extended Data Figures.

27. P. Nagler, G. Plechinger, M. v. Ballottin, A. Mitioglu, S. Meier, N. Paradiso, C. Strunk, A. Chernikov, P. C. M. Christianen, C. Schüller, T. Korn, Interlayer exciton dynamics in a dichalcogenide monolayer heterostructure. *2D Materials*. 4 (2017), doi:10.1088/2053-1583/aa7352.

28. Z. Wang, Y. H. Chiu, K. Honz, K. F. Mak, J. Shan, Electrical Tuning of Interlayer Exciton Gases in WSe2 Bilayers. *Nano Letters*. 18, 137–143 (2018).

29. M. P. A. Fisher, P. B. Weichman, G. Grinstein, D. S. Fisher, Boson localization and the superfluid-insulator transition. *Physical Review B*. 40, 546–570 (1989).

30. M. Greiner, O. Mandel, T. Esslinger, T. W. Hänsch, I. Bloch, Quantum phase transition from a superfluid to a Mott insulator in a gas of ultracold atoms. *Nature*. 415, 39–44 (2002).

31. E. Liu, E. Barré, J. van Baren, M. Wilson, T. Taniguchi, K. Watanabe, Y. T. Cui, N. M. Gabor, T. F. Heinz, Y. C. Chang, C. H. Lui, Signatures of moiré trions in WSe2/MoSe2 heterobilayers. *Nature*. 594, 46–50 (2021).

32. X. Wang, J. Zhu, K. L. Seyler, P. Rivera, H. Zheng, Y. Wang, M. He, T. Taniguchi, K. Watanabe, J. Yan, D. G. Mandrus, D. R. Gamelin, W. Yao, X. Xu, Moiré trions in MoSe2/WSe2 heterobilayers. *Nature Nanotechnology*. 16, 1208–1213 (2021).

33. F. P. Laussy, A. v. Kavokin, I. A. Shelykh, Exciton-polariton mediated superconductivity. *Physical Review Letters*. 104 (2010), doi:10.1103/PhysRevLett.104.106402.

34. L. Wang, I. Meric, P. Y. Huang, Q. Gao, Y. Gao, H. Tran, T. Taniguchi, K. Watanabe, L. M. Campos, D. A. Muller, J. Guo, P. Kim, J. Hone, K. L. Shepard, C. R. Dean, One-dimensional electrical contact to a two-dimensional material. *Science*. 342, 614–617 (2013).
35. S. L. Tomarken, Y. Cao, A. Demir, K. Watanabe, T. Taniguchi, P. Jarillo-Herrero, R. C. Ashoori, Electronic Compressibility of Magic-Angle Graphene Superlattices. *Physical Review Letters*. **123** (2019), doi:10.1103/PhysRevLett.123.046601.

36. Y. Jiang, S. Chen, W. Zheng, B. Zheng, A. Pan, Interlayer exciton formation, relaxation, and transport in TMD van der Waals heterostructures. *Light: Science & Applications*. **10**, 72 (2021).

37. E. Courtade, M. Semina, M. Manca, M. M. Glazov, C. Robert, F. Cadiz, G. Wang, T. Taniguchi, K. Watanabe, M. Pierre, W. Escoffier, E. L. Ivchenko, P. Renucci, X. Marie, T. Amand, B. Urbaszek, Charged excitons in monolayer WSe2: Experiment and theory. *Physical Review B*. **96** (2017), doi:10.1103/PhysRevB.96.085302.

38. M. Barbone, A. R. P. Montblanch, D. M. Kara, C. Palacios-Berraquero, A. R. Cadore, D. de Fazio, B. Pingault, E. Mostaani, H. Li, B. Chen, K. Watanabe, T. Taniguchi, S. Tongay, G. Wang, A. C. Ferrari, M. Atatüre, Charge-tuneable biexciton complexes in monolayer WSe2. *Nature Communications*. **9** (2018), doi:10.1038/s41467-018-05632-4.

39. Z. Li, T. Wang, Z. Lu, C. Jin, Y. Chen, Y. Meng, Z. Lian, T. Taniguchi, K. Watanabe, S. Zhang, D. Smirnov, S. F. Shi, Revealing the biexciton and trion-exciton complexes in BN encapsulated WSe2. *Nature Communications*. **9** (2018), doi:10.1038/s41467-018-05863-5.
Fig. 1. **Bosonic Mott insulator.** 

**a.** Illustration of a bosonic Mott insulator consisting of interlayer excitons. Inset: type II band alignment of WSe$_2$/WS$_2$ heterostructure. 

**b.** Schematics of continuous wave pump probe spectroscopy. The exciton and electron density are independently controlled by pump light and electrostatic gate. 

**c,e.** Gate-dependent PL (c) and absorption (e) spectra of a 60-degree aligned WSe$_2$/WS$_2$ moiré bilayer at zero pump intensity. The PL peak shows a sudden blue shift at electron filling $\nu_e = 1$ and 2 (yellow arrows), where absorption spectrum also shows kinks and splitting. 

**d,f.** Pump intensity-dependent PL (d) and absorption (f) spectrum at charge neutrality. Right axes show dipolar-interaction-induced interlayer exciton energy shift, which is approximately proportional to $\nu_{ex}$. While a qualitatively similar jump in interlayer exciton energy appears in PL at moderate pump intensity (green arrow), absorption spectrum only shows a slight decrease in the oscillator strength of intra–layer moiré excitons, indicating distinctive origin behind the exciton energy jumps in (c) and (d).
Fig. 2. **Mixed Mott insulator.** a, Pump intensity-dependent PL spectra at gate voltages $V_g$ from -0.4V (near charge neutrality) to 0.8V (electron-one-filling). b, Gate-dependent absorption spectra at pump intensities from 0 to 1.6μW/μm². Blue arrows mark the kink and splitting in absorption peaks at $\nu_e = 1$. c, Power dependent interlayer exciton energy change $\Delta \varepsilon_{\text{ex}}$ at representative gate voltages. Green triangles mark middle of the transitions, which appear at smaller pump intensity ($\nu_{\text{ex}}$) with increasing gate voltage ($\nu_e$), consistent with a mixed Mott insulator state at $\nu_{\text{tot}} = 1$. d, First order derivative of absorption spectra with respect to gate voltage at 1.76 eV under different pump intensities. Blue triangles denote $\nu_e = 1$. 
Fig. 3. Phase diagram. a-c, $\Delta E_{\text{ex}}$ with respect to the gate voltage and pump intensity at 1.65K (A), 15K (b) and 30K (c). The boundaries of $\nu_{\text{tot}} = 1$ and $\nu_e = 1$, as determined from the pump-probe PL and absorption measurements, respectively, are highlighted with green and blue triangles. The clear separation between the two boundaries confirms that the transition of $\Delta E_{\text{ex}}$ at $\nu_{\text{tot}} = 1$ is not from a charge Mott insulator state until close to $\nu_e = 1$. Regions of high pump intensity and/or high gating are not shown since peak I has already disappeared and cannot be fitted reliably(26). d-f, Vertical line-cuts of the phase diagrams for $V_g = -0.5V$ (charge neutrality) and -0.1V (electron doped) at 1.65K (d), 15K (e) and 30K (f). While at 1.65K $\Delta E_{\text{ex}}$ rises faster for $V_g = -0.1V$ than -0.5V, at 30K it rises slower for $V_g = -0.1V$ than -0.5V, suggesting partial melting and lower stability of the mixed Mott insulator state.
Fig. 4. **Mixed Hubbard model.** a, $\Delta E_{\text{ex}}$ with respect to $\nu_e$ and $\nu_{\text{ex}}$ predicted from a mixed Hubbard model in the large $U$ limit. Boundaries between region I, II and II, III correspond to $\nu_{\text{tot}} = 1$ and $\nu_e = 1$. b-d, Schematic plots showing the energy required to add one interlayer exciton (red) into the system for region I (b), II (c) and III (d) in the phase diagram. Additional energy cost of $\Delta E_{\text{ex}} = U_{\text{ex-ex}}$ and $U_{e-ex}$ is required in region II and III, respectively, from on-site repulsion between excitons and between electron-exciton.
Methods

Device fabrication and characterization: Monolayer WS$_2$, monolayer WSe$_2$, few-layer graphite and thin hexagonal boron nitride (hBN) flakes were exfoliated onto silicon substrates with a 285 nm silicon oxide layer. Polarization-resolved second harmonic generation (SHG) was used to determine the relative angles between the WSe$_2$ and WS$_2$ crystalline axes (see Fig. S10). The hBN flakes with a thickness of around 20 nm were used as the gate dielectrics. Few-layer graphite flakes were used as the contact electrode and gates. The WSe$_2$/WS$_2$ heterostructures with symmetric top and bottom gates were built using a layer-by-layer dry transfer method (34). The whole sample was then released to a 90nm Si/SiO$_2$ substrate. The stamps for picking up flakes were made of a film of polycarbonate on polydimethylsiloxane. The polycarbonate residue on the sample was removed by dissolving in chloroform, followed by a rinse in isopropyl alcohol. Contacts (~100 nm gold with ~5 nm chromium and ~15nm palladium adhesion layers) to the few-layer graphite were made by electron-beam lithography, plasma etching and electron-beam evaporation. Fig. S10a shows an optical image of a typical device. Since the top and bottom gates are nearly symmetric, we applied the same gate voltage $V_g$ to top and bottom gates to tune carrier concentration of the sample while keeping the out-of-plane electric field near zero. The sample was electron-doped with positive gate voltage.

Estimation of exciton density at charge neutrality: the exciton density under optical pumping can be estimated through the continuous blueshift of emission energy (Fig. 1d) from exciton dipole-dipole interaction. Since interlayer excitons in WSe$_2$/WS$_2$ heterostructure have a permanent dipole moment $p_{ex}$ perpendicular to the sample plane (Fig. 1a inset), they will couple to any out-of-plane electric field $E_{op}$ and show an energy shift

$$\Delta = -p_{ex}E_{op}$$

At an interlayer exciton density of $n_{ex}$, the WSe$_2$ and WS$_2$ layers accumulate a hole and electron density of $n_{ex}$, respectively, which effectively generates an out-of-plane electrical field of $E_{dipole} = -\frac{en_{ex}}{\varepsilon_r\varepsilon_0}$ in between. Here $\varepsilon_r$ is the out-of-plane dielectric constant of the heterobilayer and $\varepsilon_0$ is vacuum permittivity. From this picture, the blueshifts of exciton energy from dipole-dipole interaction is $\Delta_{dipole} = -p_{ex}E_{dipole} = \frac{e_n_{ex}}{\varepsilon_r\varepsilon_0}p_{ex}$ (Ref. 28). On the other hand, when using a single gate to dope the WSe$_2$/WS$_2$ bilayer with electron density $n_e$, the electric field at the sample is $|E_{Stark}| = \frac{en_e}{\varepsilon_r\varepsilon_0}$, resulting in a Stark shift of $|\Delta_{Stark}| = \frac{en_e}{\varepsilon_r\varepsilon_0}p_{ex}$ (Ref. 28). We can therefore estimate $n_{ex}$ by comparing $\Delta_{dipole}$ and $\Delta_{Stark}$ following

$$\frac{n_{ex}}{n_e} = \frac{\Delta_{dipole}}{|\Delta_{Stark}|}$$

At charge neutrality, the jump in exciton energy happens at pump intensity around 0.7$\mu$W/μm$^2$.
(Fig. 1d), where the blueshift of peak I is $\Delta_{\text{dipole}} \approx 7$ meV compared to zero pump intensity (Fig. S9a). Meanwhile, using the bottom gate only, the exciton emission shows Stark shift $|\Delta_{\text{Stark}}| \approx 13$ meV from $v_e = 0$ to $v_e = 1$ (Fig. S9b), corresponding to an electron density $n_e = n_0$, $n_0 \approx 2.1 \times 10^{12} \text{cm}^{-2}$ is the density of one moiré filling for 1° twist(3, 4). We thereby estimate the incompressible state of interlayer exciton to emerge at $n_{ex} \approx 0.5 n_0 \sim 10^{12} \text{cm}^{-2}$. The above picture assumes a homogeneous sheet of charges/excitons and neglects any short-range correlation effects, therefore only offering an order-of-magnitude estimation. Nevertheless, the estimated exciton density of $\sim 10^{12} \text{cm}^{-2}$ is consistent with the Bosonic Mott insulator picture.

Inhomogeneous broadening in exciton density: an incompressible state ideally leads to a step function in chemical potential with respect to density. In practice, however, the step always has finite width due to inhomogeneous broadening. For example, broadening is universally observed in electrical capacitance measurements due to inhomogeneity in charge density across the device(35). Similarly, our pump-probe measurements collect signals from the entire probe-covered region, where spatial inhomogeneity in exciton density will broaden the transition of exciton energy. On the other hand, the inhomogeneous broadening of excitons is expected to be much larger since the exciton density depends on lifetime, which can vary by orders of magnitude depending on defects and strains(36). The electron density, in contrast, mainly depends on local dielectric environment and typically varies by only a few percent. This is consistent with our observation that the transition in $\Delta E_{\text{ex}}$ is sharper at $v_e = 1$ than at $v_{ex} = 1$.

Data analysis: The PL spectra were fitted using a two-Lorentzian model. We define exciton energy change $\Delta E_{\text{ex}} = (E_2 I_2 + E_1 I_1)/(I_1 + I_2) - E_1 = (E_2 - E_1) I_2/(I_1 + I_2)$. By taking the energy difference, we remove energy shifts from long-range dipolar interaction between interlayer excitons, which are universally reported to induce a continuous blueshift with increasing exciton density regardless of moiré effects(27, 28). Examples of fitting curves are shown in Fig. S8. At high pump power and/or high gate, peak II dominates and peak I largely disappears, making the fitted $\Delta E_{\text{ex}}$ unstable. In the phase diagram we only included regions where $\Delta E_{\text{ex}}$ can be reliably fit. To determine the electron one filling from absorption spectrum, we took first order derivative of reflection contrast with respect to gate voltage and focused on the intralayer moiré exciton peak around 1.76 eV (Fig. 2d), which shows a well-defined splitting/kink at one electron filling (Fig. 1e and Fig. 2b).

Two-component Hubbard model: Our experimental results are well captured by a two-component Hubbard model with both fermionic and bosonic species(24)

$$H = H_{\text{ex}} + H_e + U_{\text{ex}-e} \sum_{i, \sigma} n_{\text{ex} i \sigma} n_{e i \sigma}$$
\[ H_{\text{ex}} = -t_{\text{ex}} \sum_{\langle ij \rangle} b_i^{+} b_j + \frac{U_{\text{ex-ex}}}{2} \sum_i n_{\text{ex},i} (n_{\text{ex},i} - 1) \]

\[ H_{\text{e}} = -t_{\text{e}} \sum_{\langle ij \rangle, \sigma} c_{i,\sigma}^{+} c_{j,\sigma} + U_{\text{e-e}} \sum_{l, \sigma \neq \sigma'} n_{\text{e},l,\sigma} n_{\text{e},l,\sigma'} \]

Where \( \langle i,j \rangle \) represents the summation over nearest-neighbors and \( \sigma \) denotes electron spin. \( b_i^{+} \) (\( c_{i,\sigma}^{+} \)) and \( b_j \) (\( c_{j,\sigma} \)) are creation and annihilation operators for excitons and electrons respectively. \( n_{\text{ex},i} \) and \( n_{\text{e},i,\sigma} \) are number operators for excitons and electrons. \( U_{\text{ex-ex}} \), \( U_{\text{e-e}} \) and \( U_{\text{e-ex}} \) denote on-site repulsive energies between excitons, electrons and exciton-electron. \( t_{\text{ex}} \) and \( t_{\text{e}} \) are the nearest-neighbor-hopping energy.

Fig. 4a in the main text summarizes the predicted relative exciton chemical potential \( \Delta E_{\text{ex}} \) in the large \( U \) limit, which matches well with the experiment. Along \( \nu_{\text{ex}} = 0 \) and \( \nu_{\text{e}} \neq 0 \), our model provides a natural explanation to the widely observed yet not fully understood PL doping dependence in WSe\(_2\)/WS\(_2\)\((6, 8)\). At \( \nu_{\text{e}} > 1 \), the PL emission comes from a composite particle of electron and exciton on the same site, which may be considered as a correlated version of trion. However, owing to the strong correlation in the present system, it is fundamentally different from a conventional trion that has attractive interaction between the exciton and electron\((31, 32, 37)\).

The phase diagram along the vertical axis \( (\nu_{\text{e}} = 0, \nu_{\text{ex}} \neq 0) \) can be similarly understood. PL emission above \( \nu_{\text{ex}} = 1 \) originates from a composite particle of two excitons on the same site, i.e., a correlated version of biexciton. Again, it is fundamentally different from biexcitons in systems without strong correlation, which would appear at a lower energy compared to single exciton\((38, 39)\).

Along the line of \( \nu_{\text{tot}} = \nu_{\text{ex}} + \nu_{\text{e}} = 1 \), the jump of exciton energy with \( \nu_{\text{ex}} \) indicates that it is generally exciton incompressible until \( \nu_{\text{e}} = 1 \). Because here excitons and electrons are on an equal footing, we expect the \( \nu_{\text{tot}} = 1 \) line to be also charge incompressible. This mixed Mott insulator state can be considered as a mixture of fermionic and bosonic Mott insulator, and becomes less stable than both components at higher temperature (Fig. 3 a-c). As a potential mechanism, the mixed Mott insulating state involves total electron density of \( \nu_{\text{tot}} = 1 \) and hole density of \( \nu_{\text{h}} = \nu_{\text{ex}} < 1 \). While electrons have occupied all available sites, holes are at a general filling and may move around without energy penalty. At elevated temperature, holes can become mobile enough to drag electrons away and destabilize the mixed Mott insulating state.
Fig. S1. Complete PL and absorption spectra. a,b, Doping-dependent PL (a) and absorption (b) spectrum of a 60-degree aligned WSe$_2$/WS$_2$ moiré bilayer device D1 at zero pump intensity, from which all data in the main text is taken.
Fig. S2. Results from another 60-degree aligned moiré bilayer. a, Doping-dependent absorption (a) and PL (b) spectrum of another 60-degree aligned WSe$_2$/WS$_2$ moiré bilayer device D2. c, Pump intensity-dependent PL at charge neutrality. A qualitatively similar jump in interlayer exciton energy is observed, indicating emergence of a bosonic Mott insulator composed of interlayer excitons.
Fig. S3. Results from a misaligned sample. a, b, Doping-dependent PL (a) and absorption (b) spectrum of a few-degree-misaligned WSe₂/WS₂ bilayer device D3. c, Pump intensity-dependent PL at charge neutrality. No jump in the interlayer exciton energy is observed due to the lack of strong correlation. On the other hand, the continuous PL blueshift from long-range dipolar interactions is still observed.
Fig. S4. Pump intensity-dependent PL spectra at 15K for gate voltages from -0.5V (charge neutrality) to 0.8V (electron-one-filling).
Fig. S5. Gate-dependent absorption spectra at 15K for pump intensities from 0μW/μm² to 1.8μW/μm².
Fig. S6. Pump intensity-dependent PL spectra at 30K for gate voltages from -0.5V (charge neutrality) to 0.8V (electron-one-filling).
Fig. S7. Gate-dependent absorption spectra at 30K for pump intensities from 0μW/μm² to 2.5μW/μm².
**Fig. S8. Two-Lorentzian model fitting.**

**a,** experimental PL data and fitting curves under 1.65K, \( V_g = -0.5 \text{V} \) (charge neutrality) and 0.72\( \mu \text{W/}\mu \text{m}^2 \) pumping.

**b,** experimental PL data and fitting curves under 1.65K, \( V_g = -0.2 \text{V} \) and 0.9\( \mu \text{W/}\mu \text{m}^2 \) pumping.
Fig. S9. Estimation of exciton density. a, Pump-probe PL spectrum at charge neutrality and selected pump intensity. The low energy peak (peak I) shows a continuous blueshift with pump intensity from dipolar interaction between interlayer excitons, which can be used to estimate the exciton density generated by pump. b, PL spectrum at zero pump intensity and selected electron fillings tuned by a single gate. Peak I shows a continuous Stark shift with respect to the filling, which can be used to calibrate the relation between carrier density and PL shift.
Fig. S10. Optical image and SHG measurement of representative devices. **a**, optical image of a typical dual-gated 60-degree aligned WSe$_2$/WS$_2$ device D1. Yellow and green solid lines denote the sample edges of WSe$_2$ and WS$_2$, respectively. The scalar bar is 5$\mu$m. **b**, SHG measurement on monolayer WSe$_2$ (red circles), monolayer WS$_2$ (grey squares) and heterobilayer (blue triangles) region. Solid curves are fits to the data using $A + B\cos^2[3(\phi - \phi_0)]$, where $\phi$ is the excitation polarization angle, $\phi_0$ is the crystal orientation and $A$ and $B$ are fitting parameters. For WSe$_2$ and WS$_2$ monolayer, the crystal orientation $\phi_0$ is 6.7° ± 0.2° and 5.6° ± 0.1° respectively. The twist angle is determined to be 1.1° ± 0.3°. Since the SHG signal from the bilayer region is much weaker than that from the monolayers, we conclude that the WSe$_2$ and WS$_2$ are nearly 60-degree aligned.