Moiré coupling in transition metal dichalcogenides (TMDCs) superlattices introduces flat minibands that enable strong electronic correlation and fascinating correlated states, and it also modifies the strong Coulomb-interaction-driven excitons and gives rise to moiré excitons. Here, we introduce the layer degree of freedom to the WSe₂/WS₂ moiré superlattice by changing WSe₂ from monolayer to bilayer and trilayer. We observe systematic changes of optical spectra of the moiré excitons, which directly confirm the highly interfacial nature of moiré coupling at the WSe₂/WS₂ interface. In addition, the energy resonances of moiré excitons are strongly modified, with their separation significantly increased in multilayer WSe₂/monolayer WS₂ moiré superlattice. The additional WSe₂ layers also modulate the strong electronic correlation strength, evidenced by the reduced Mott transition temperature with added WSe₂ layer(s). The layer dependence of both moiré excitons and correlated electronic states can be well described by our theoretical model. Our study presents a new method to tune the strong electronic correlation and moiré exciton bands in the TMDCs moiré superlattices, ushering in an exciting platform to engineer quantum phenomena stemming from strong correlation and Coulomb interaction.

In a strongly correlated electronic system, Coulomb interactions among electrons dominate over kinetic energy. Recently, two-dimensional (2D) moiré superlattices of van der Waals materials have emerged as a promising platform to study correlated physics and exotic quantum phases in 2D, such as the correlated insulating states and superconductivity in graphene moiré superlattices\(^{1-16}\), the Mott insulator state at half band filling and various generalized Wigner crystal states at fractional fillings of the moiré superlattices based on transition metal dichalcogenides (TMDCs)\(^{15-23}\). The key to the strong correlation in these systems is the enhanced Coulomb interaction in
2D and greatly reduced kinetic energy in the flat moiré minibands. In TMDC-based moiré superlattices, the combination of large effective mass and strong moiré coupling renders the easier formation of flat bands and stronger electronic correlation, compared with graphene moiré superlattices. For example, the 0- or 60-degree angle-aligned WSe$_2$/WS$_2$ exhibits Mott insulating states with transition temperatures exceeding 150 K $^{18,19}$, the highest among all 2D moiré systems studied so far. It also hosts various correlated insulating states at fractional fillings of the moiré lattice$^{18,19}$, indicating strong and long-range electron interactions.

Meanwhile, the strong Coulomb interaction in 2D also leads to tightly bound excitons with large binding energy in TMDCs$^{24-27}$. The moiré coupling in the TMDC moiré superlattices is expected to generate excitonic flat minibands$^{28}$, beyond the single-particle electronic flat bands in the conduction and valence bands. Recently, the moiré excitons have been reported in the angle-aligned WSe$_2$/WS$_2$ heterojunction$^{17,18}$, in which correlated insulating states also occur$^{17-19}$. The excitonic flat band is promising for realizing topological exciton states and correlated exciton Hubbard model$^{17,18}$, ushering in exciting opportunities for engineered correlated quantum states. However, there are key questions that remained to be addressed. For example, how is the moiré coupling’s extension in the out-of-plane direction? How could one systematically tune both the electronic flat bands and moiré exciton bands in the TMDCs moiré superlattice?

In this work, we investigate these questions utilizing the layer degree of freedom, inspired by the layer-layer coupling in TMDCs that leads to the abrupt direct-to-indirect bandgap transition from monolayer to bilayer TMDCs$^{30,31}$. We demonstrate a general approach tuning both electronic and moiré exciton bands by increasing the layer number of WSe$_2$ in the angle-aligned WSe$_2$/WS$_2$ heterojunction. As the layer number of WSe$_2$ varies from monolayer (1L) to bilayer (2L) and trilayer (3L), the optical spectra of the moiré exciton change systematically in a way that suggests the moiré coupling is highly interfacial, strongly confined at the WSe$_2$/WS$_2$ interface and barely affects the next neighboring WSe$_2$ layer(s). However, the added WSe$_2$ layer(s) could modify moiré excitons in the WSe$_2$ layer interfacing WS$_2$, resulting in a significant increase in the resonance energy separations between moiré excitons. This observation can be well described by a phenomenological model. Our work, to our best knowledge, reports the first sensitive tuning of moiré excitons via layer degree of freedom.

The correlated electronic structure is also sensitive to the number of layers in WSe$_2$. The Mott insulator state at the filling of one hole per moiré unit cell ($n = -1$) is found to have a transition temperature decreased from 150 K in 1L/1L WSe$_2$/WS$_2$ to 120 K in 2L/1L WSe$_2$/WS$_2$ and 60 K in 3L/1L WSe$_2$/WS$_2$. The correlated states at fractional fillings (fractional charge per moiré supercell) are significantly quenched in the 3L/1L WSe$_2$/WS$_2$ heterojunction. The reduced Mott transition temperature, however, is still significantly higher than that of graphene moiré superlattices (~4 K). Our study, therefore, also demonstrates a new knob to tune the strong electron correlation in TMDC moiré superlattices that can be further exploited for engineering new correlated quantum states.

Results and discussion

The back-gated angle-aligned WSe$_2$/WS$_2$ heterojunction device is schematically shown in Fig. 1a, which includes three different regions in the same device: 1L/1L WSe$_2$/WS$_2$, 2L/1L WSe$_2$/WS$_2$, and 3L/1L WSe$_2$/WS$_2$. The device was constructed through a dry pickup method described previously$^{13}$ (also see Methods), and the heterojunctions are encapsulated with layers of BN on both sides. The few-layer-graphene (FLG) works as the back gate electrode.
encapsulated with flakes of boron nitride (BN) and gated through few-layer graphene flake working as the back gate electrode. WSe$_2$ and WS$_2$ have a lattice mismatch of ~4%, resulting in a moiré superlattice with a periodicity of ~8 nm$^{1,23}$, when they are angle-aligned (0- or 60-degree twisted). Having the three regions (1 L/1 L WSe$_2$/WS$_2$, 1 L/2 L WSe$_2$/WS$_2$, and 3 L/1 L WSe$_2$/WS$_2$) in the same device is advantageous as these three regions have the same angle, so the WSe$_2$ for all these three regions are from the same flake. As a result, the moiré lattice constant in these three regions are about the same, and we can compare our measurements from these different regions directly.

The 1 L/1 L WSe$_2$/WS$_2$ heterojunction has a type-II band alignment, with the conduction band minimum located in the WS$_2$ layer and the valence band maximum in the WSe$_2$ layer$^2$. Strong moiré coupling leads to a band folding in the mini-Brillouin zone and generates moiré exciton bands$^3$, which will split the A exciton resonance of WSe$_2$ into three moiré exciton peaks, as demonstrated in the previous experiments$^{3,13}$. Here we measure the reflectance spectra in the three different regions of the WSe$_2$/WS$_2$ heterojunction as a function of the gate voltage, with results shown in Fig. 1c–e. There are two major differences between the moiré exciton spectra from the 1 L/1 L WSe$_2$/WS$_2$ and multilayer WSe$_2$/WS$_2$ (2 L/1 L WSe$_2$/WS$_2$ or 3 L/1 L WSe$_2$/WS$_2$) heterojunctions. First, it is evident that near the charge-neutral region (gate voltage ~0 V), there are three moiré exciton resonances in the 1 L/1 L WSe$_2$/WS$_2$ region (Fig. 1c) but four in both 2 L/1 L (Fig. 1d) and 3 L/1 L (Fig. 1e) WSe$_2$/WS$_2$ regions. Second, the moiré exciton energy difference between the lowest and highest energy moiré excitons increases significantly in both 2 L/1 L (Fig. 1d) and 3 L/1 L (Fig. 1e) WSe$_2$/WS$_2$ regions. These observations are better illustrated in Fig. 1b, which plots the differential reflectance spectra at the gate voltage of 0 V for the three different regions (line cuts at zero gate voltage in Fig. 1c–e). For the 1 L/1 L WSe$_2$/WS$_2$ region, we observe three moiré exciton peaks at ~1.662 eV ($X_{IA}^I$), 1.715 eV ($X_{IA}^{II}$), and 1.733 eV ($X_{IA}^{III}$. consistent with the previous reports$^2$. However, in the 2 L/1 L WSe$_2$/WS$_2$ region, one additional exciton resonance emerges, adding up to a total of four major excitons peaked at ~1.642 eV ($X_{IA}^I$), 1.693 eV ($X_{IA}^{II}$), 1.728 eV ($X_{IA}^{IV}$), and 1.793 eV ($X_{IA}^{V}$). In the 3 L/1 L WSe$_2$/WS$_2$ region, there are also four major exciton resonances at ~1.645 eV ($X_{IA}^I$), ~1.677 eV ($X_{IA}^{II}$), 1.730 eV ($X_{IA}^{III}$), and 1.785 eV ($X_{IA}^{IV}$). The largest moiré exciton energy difference, defined as the energy difference between $X_{IA}^{VI}$ and $X_{IA}^I$, is ~90 meV for the 1 L/1 L WSe$_2$/WS$_2$ region but ~150 meV for 2 L/1 L WSe$_2$/WS$_2$ region and ~140 meV for 3 L/1 L WSe$_2$/WS$_2$ region, an increase of more than 50%. Similar behaviors have been observed for all the devices we have studied (details in Supplementary Note 5).

Our observations suggest that the moiré potential is highly localized at the WSe$_2$/WS$_2$ interface and has a limited extension along the out-of-plane direction. As a result, the moiré coupling only significantly modifies the first WSe$_2$ layer in contact with the monolayer WS$_2$. The newly developed exciton resonances in the 2 L/1 L and 3 L/1 L WSe$_2$/WS$_2$ heterojunctions ($X_{IA}^{IV}$ and $X_{IA}^{V}$), therefore, arise from the barely modified intralayer A exciton in the upper WSe$_2$ layers away from the interface (also see Supplementary Note 4). Our interpretation is supported by the fact that the energies of $X_{IA}^{IV}$ (1.693 eV) and $X_{IA}^{V}$ (1.677 eV) are close to the intralayer A exciton energy of monolayer WSe$_2$ (~1.70 eV), and it is further corroborated by the stronger reflectance intensity from the new moiré exciton in 3 L/1 L WSe$_2$/WS$_2$ ($X_{IA}^{VI}$) compared with that in 2 L/1 L WSe$_2$/WS$_2$ ($X_{IA}^{IV}$). Moreover, there is a redshift in the moiré exciton resonance $X_{IA}^I$ and blueshifts in $X_{IA}^{II}$ and $X_{IA}^{III}$ compared with those in 3 L/1 L WSe$_2$/WS$_2$ (Fig. 1b). And the shift of $X_{IA}^I$ and $X_{IA}^{II}$ is more significant in magnitude than that of $X_{IA}^{III}$.

Our results can be understood with a phenomenological model (details in Supplementary Note 2), considering the moiré excitons in the first WSe$_2$ layer interacting with a exciton state in the added WSe$_2$ layer(s) that has the resonance energy between $X_{IA}^I$ and $X_{IA}^{II}$. The resulting level repulsion naturally explains the redshift of $X_{IA}^I$ and blue shift of $X_{IA}^{II}$ in the 2 L/1 L (3 L/1 L) WSe$_2$/WS$_2$ compared with 1 L/1 L WSe$_2$/WS$_2$.
moiré exciton for the three different regions ($X_{1L}^I$, $X_{2L}^I$, and $X_{3L}$), which clearly shows intensity modulations at $n = -1$ and $+1$. On the other hand, the additional excitons in 2L/1L ($X_{1L}^II$) and 3L/1L ($X_{1L}^III$) WSe$_2$/WS$_2$ regions are barely affected by the formation of the Mott states at $n = \pm 1$ (Fig. 1d, e). These behaviors can also be explained by the interfacial nature of the moiré coupling, which confines the correlated electrons at the interface of WSe$_2$/WS$_2$. The modulation of the moiré excitons at the $n = \pm 1$ is likely due to the dielectric constant change and gap opening associated with the Mott insulator states. Due to the small radius of the strongly bound exciton\(^7\), only the moiré excitons in the first WSe$_2$ layer immediately interfacing with the WS$_2$ monolayer can sensitively detect the dielectric constant change at the interface. In the 2L/1L and 3L/1L regions, the additional excitons originated from intralayer excitons localized in the added layers, are thus barely affected.

To better investigate the tuning of the electron correlation by the layer degree of freedom, we perform microwave impedance microscopy (MIM) measurements to study the correlated insulating states in the three different heterostructure regions (Fig. 4a). MIM probes the local conductivity of the sample and has been successfully employed to reveal a rich structure of correlated insulating states in the angle-aligned 1L/1L WSe$_2$/WS$_2$ device\(^8\). In the multilayer WSe$_2$/1L WS$_2$ device, we primarily focus on the features on the hole side, as the holes reside in the WSe$_2$ layer due to the type-II alignment, and we introduce the layer degree of freedom by modulating the layer number of WSe$_2$. At temperature $T = 10$ K, the MIM spectra in both 1L/1L and 2L/1L WSe$_2$/WS$_2$ regions show similar pronounced features at various fillings, including the Mott insulator states at $n = -1$, the generalized Wigner crystal states at fractional fillings of $n = -1/3 \& -2/3, -1/2, -1/4 \& -3/4$, etc. The 3L/1L WSe$_2$/WS$_2$ data show fewer and less pronounced dips: other than the Mott insulator state at $n = -1$, only two fractional fillings $n = -1/3$ and $-1/2$ can be resolved. There is also a small difference in the twist angle in the 3L/1L WSe$_2$/WS$_2$ region ($1.3°$) compared to that in the 1L/1L and 2L/1L regions ($0.9°$), which results in different gate voltage positions for these insulating states in the 3L/1L WSe$_2$/WS$_2$ region (details in method).

Since the formation of the correlated insulating states at fractional fillings depends on long-range Coulomb interaction among electrons in neighboring moiré unit cells, our results suggest that the inter-site electron interaction strength is weaker in 3L/1L than in 1L/1L or 2L/1L WSe$_2$/WS$_2$. The difference in the on-site interaction, corresponding to the Mott insulator state at $n = -1$, can be further revealed in its temperature dependence. As shown in Fig. 4b–d, as the temperature is raised, the features at fractional fillings disappear at $-30$ K in both 1L/1L and 2L/1L WSe$_2$/WS$_2$ regions and at $-15$ K in the 3L/1L WSe$_2$/WS$_2$ region. The Mott insulator state at $n = -1$ survives at much higher temperatures in 1L/1L WSe$_2$/WS$_2$, persisting to above $180$ K, the highest Mott transition temperature reported in all 2D moiré superlattice structures so far. In the 2L/1L WSe$_2$/WS$_2$ region, the Mott transition temperature is $-120$ K, while it is much lower, $-60$ K, in the 3L/1L region. As the correlation strength is determined by the ratio of the Coulomb interaction to the kinetic energy, the reduction of electron correlation strength from 1L/1L to 2L/1L WSe$_2$/WS$_2$ is likely due to the increased dielectric screening from the added WSe$_2$, which reduces the Coulomb interaction.  

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**Fig. 2 | Theoretical simulation of moiré excitons.** a, b are the energy bands of bare intralayer A excitons in 1L WSe$_2$ and moiré excitons in 1L/1L WSe$_2$/WS$_2$, respectively. a The A exciton bands are folded into the mini-Brillouin zone of the moiré superlattice to compare directly with that of moiré excitons in b. Inset of a shows the schematic of the mini-Brillouin zone and the label of high symmetry points. The WSe$_2$ bright intralayer A exciton state is marked by $X_A$ in a. The three bright Moiré exciton states are marked by $X_{1L}^I$, $X_{2L}^I$, and $X_{3L}$ in b. Here, we set the energy $E_X$ of $X_A$ as the energy reference. c–e Optical absorption spectra in 1L/1L, 2L/1L, and 3L/1L WSe$_2$/WS$_2$. The interlayer hybridization between moiré exciton and intralayer A exciton is considered in d and e, with details elaborated in Supplementary Information Note 2.
interaction at the interface. However, the further reduced correlation strength in the 3 L/1 L WSe₂/WS₂ is facilitated by the additional increase of kinetic energy, which arises from the increased bandwidth of the flat band, according to our calculation shown in Supplementary Information Fig. 2. We emphasize here that even the reduced electron correlation in the 2 L/1 L and 3 L/1 L WSe₂/WS₂ is still significantly stronger than that in graphene moiré systems, which has a Mott transition temperature ~4 K. As a result, the layer degree of freedom can be utilized for engineering new correlated states.

In summary, we have demonstrated a new moiré superlattice system based on multilayer TMDC heterojunctions. The added layers host additional intralayer excitons that interact with the moiré excitons residing at the moiré interface, and they can further modify the correlation strength of the correlated states. Considering the layer-valley-spin locking in TMDC₃⁸, these new TMDCs moiré superlattices provide an exciting platform to investigate emerging correlated valley and spin physics.

Methods

Heterostructure device fabrication

We use a dry pickup method²⁰,₃⁹ to fabricate the WSe₂/WS₂ heterostructures. We exfoliate monolayer WS₂, multilayer WSe₂, graphite, and BN layers on silicon substrate with a 285 nm thermal oxide layer. For angle-aligned heterostructures, we choose exfoliated WS₂ and WSe₂ layers with sharp edges, whose crystal axes are further confirmed by second harmonic generation measurements. We then mount the SiO₂/Si substrate on a rotational stage and clamp the glass slide with the WSe₂ flat band from the 1st layer WSe₂ labeled in red. d-f are differential reflectance intensity of the lowest energy moiré exciton (X₁) as a function of the back gate voltage (carrier density) in Fig. 1c-e, respectively (vertical dashed line cuts), with the 2D color plots showing the enhanced reflectance spectra near the moiré excitons X₁ and X₃.

between the two flakes. The final constructed device is annealed at 130 °C for 12 hours in a vacuum chamber. The pre-patterned Au contact electrodes are fabricated through standard electron-beam lithography and e-beam evaporation processes (see Supplementary Information Fig. 3 for the optical microscope image of the device used in the main text). More sample characterization details can be found in Supplementary Note 4.

Optical spectroscopy measurements

To perform differential reflectance contrast measurement, the samples were mounted in a helium flow-controlled cryostat with a quartz optical window and electrical feedthroughs. A super-continuum laser (YL Photonics) was used as the white light source. The laser was focused onto the sample with a ×50 objective (the typical laser spot size is ~2 μm). The reflected light was directed into a spectrograph and collected with a CCD camera (Princeton Instruments). The differential reflectance is calculated as δR = R – R₀, where R is the reflectance spectrum at the highest p-doping region as the reference R₀.

Microwave impedance microscopy measurements

The MIM measurement is performed on a homebuilt cryogenic scanning probe microscope platform. A small microwave excitation of about 0.1 μW at a fixed frequency -10 GHz is delivered to a chemically etched tungsten tip mounted on a quartz tuning fork. The reflected signal is analyzed to extract the demodulated output channels, MIM-I and MIM-Re, which are proportional to the imaginary and real parts of the admittance between the tip and sample, respectively. To enhance the MIM signal quality, the tip on the tuning fork is excited to oscillate at a frequency of around 32 kHz with an amplitude of ~8 nm. The resulting oscillation amplitudes of MIM-I and MIM-Re are then extracted using a lock-in amplifier to yield d(MIM-I)/dz and d(MIM-Re)/dz, respectively. The d(MIM)/dz signals are free of fluctuating backgrounds, and their behavior is very similar to that of the standard...
MIM signals. In this paper, we simply refer to d(MIM)/dz as the MIM signal.

Estimating the twist angle
Twist angles of the moiré superlattices can be estimated by the carrier density corresponding to the correlated insulating state at \( n = \pm 1 \), with a bottom hBN of thickness \( \sim 52 \) nm and dielectric constant 3.5. The 1L/1L WSe\(_2\)/WS\(_2\) and 2L/1L WSe\(_2\)/WS\(_2\) regions have a similar moiré periodicity of 7.4 nm and twist angle of 0.9°. For the 3L/1L WSe\(_2\)/WS\(_2\) region, the moiré periodicity is 6.0 nm and the twist angle is 1.3°. This difference is likely caused by a small distortion or wrinkle between WSe\(_2\) and WS\(_2\) layers.

Data availability
Source data are available for this paper. All other data that support the plots within this paper and other finding of this study are available from the corresponding author upon reasonable request.

Code availability
The source code for the numerical simulations is available from the corresponding author upon reasonable request.

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### Author contributions

S.-F. S. and Y.-T. C. conceived the project, D.C. and Z.L. fabricated the heterostructure devices and performed the optical spectroscopy measurements. X.H. performed the MIM measurements. M.R. helped with device fabrication. M.B. and S.T. grew the TMDC crystals. T.T. and K.W. grew the BN crystals. C.Z. and Y.S. performed the theoretical calculations. S.-F.S., Y.-T.C., C.Z., Y.S., D.C., Z.L., Z.W., X.H., and L.Y. analyzed the data. S.-F.S. and Y.-T.C. wrote the manuscript with inputs from all authors.

### Competing interests

The authors declare no competing interests.

### Additional information

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**Correspondence** and requests for materials should be addressed to Zenghui Wang, Chuanwei Zhang, Yong-Tao Cui or Su-Fei Shi.

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