Dipolar excitonic insulator in a moiré lattice

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Two-dimensional moiré materials provide a highly controllable solid-state platform for studies of quantum phenomena1–3. To date, experimental studies have focused on correlated electronic states, whereas correlated bosonic states in moiré materials have received less attention. Here we report the observation of a correlated dipolar excitonic insulator—a charge-insulating state driven by exciton formation—in a device where a WSe2 monolayer and WSe2/WS2 moiré bilayer are coupled via Coulomb interactions. The system is a Mott insulator when all the holes reside in the moiré layer. Under an out-of-plane electric field, the holes can be continuously transferred to the WSe2 monolayer, but remain strongly bound to the empty moiré sites, effectively forming an interlayer exciton fluid in the moiré lattice. We further observe the emergence of local magnetic moments in the WSe2 monolayer induced by the strong interlayer Coulomb correlation. Our result provides a platform for realizing correlated quantum phenomena described by bosonic lattice models in a solid-state system, complementary to cold-atom setups1.

We consider a heterostructure of a WSe2 monolayer and a WSe2/WS2 moiré bilayer that are separated by a thin hexagonal boron nitride (hBN) spacer (Fig. 1a). The spacer suppresses single-particle tunnelling between the monolayer and moiré layer, but is sufficiently thin (2–4 layers; thickness, d ≈ 1 nm) relative to the moiré period (a_m ≈ 8 nm); therefore, the interlayer Coulomb interaction is as important as the Coulomb interaction within the moiré layer. We discuss the case of hole doping: the concept also applies to the case of electron doping. The valence band edge of the WSe2/WS bilayer resides in the constituent WSe2 layer25; it is above the band edge of the WSe2 monolayer because of the moiré potential26 (Fig. 1b). The flat moiré bands in angle-aligned WSe2/WS2 bilayers have been shown to realize the triangular lattice Fermi–Hubbard model in the strong-correlation regime26–29. When the moiré layer is populated with one hole per moiré site and the monolayer is undoped, the system is an electronic Mott insulator (Fig. 1d, left). An out-of-plane electric field E, which shifts the relative band alignment, can be tuned so that some holes are transferred from the moiré layer to the monolayer with the total hole density fixed (Fig. 1d, middle). The holes in the monolayer are bound to the empty moiré sites to minimize the Coulomb repulsion energy. Under a particle–hole transformation in the moiré layer, the empty sites become ‘electrons’; excitons—bound states of holes in the monolayer valence band and electrons in the moiré Hubbard band—emerge. These interlayer excitons, which are strongly interacting, can hop around and form a fluid in the moiré lattice (Fig. 1d, right), in contrast to the Mott insulator in an isolated moiré layer in which the charges are frozen, and the reported exciton fluid is in a continuum30,31. It is also a zero-magnetic-field analogue of the exciton fluid in quantum Hall bilayers32, induced by the moiré flat bands instead of the dispersionless Landau levels.

The interlayer excitons are tightly bound in the limit of low exciton density and d ≪ a_m. Dissociation of an exciton corresponds to removing a hole from its pairing empty site and creating a doublon on another site. Ignoring extended Coulomb interactions, we estimate the exciton binding energy using the on-site Coulomb repulsion energy, namely, (ε_0 + e^2/4πε_0d) ≈ 50 meV (here, ε, ε ≈ 4.5 (ref. 9) and ε_0 denote the elementary charge, effective dielectric constant of hBN and vacuum permittivity, respectively; a_m approximates the Wannier-function size of a hole). The ground state of the fluid is charge incompressible but exciton compressible in general; perfect Coulomb drag is expected in the coupled monolayer and moiré layer5. The exciton density can be continuously varied by the electric field F. In the low exciton density or the Bose–Einstein condensation (BEC) regime, the exciton fluid is expected to transition from a normal fluid to a superfluid at sufficiently low temperatures and disorder densities5–9,20. The large exciton binding energy favours a high transition temperature19–22. In the high exciton density or the Bardeen–Cooper–Schrieffer (BCS) regime, superfluidity and electron–hole binding are expected to occur at nearly the same transition temperature5,9.

In this study, we demonstrate the realization of equilibrium excitons in a lattice using the moiré heterostructure (Fig. 1a). Both 0°- and 60°-aligned WSe2/WS2 bilayers (moiré density, around 395 m) and monolayer (ν = 1) have been studied; they show similar results. The heterostructure is dual gated with nearly symmetric top and bottom gates made of hBN gate dielectric and graphite gate electrodes (Extended Data Fig. 1). The two gate voltages (V_a and V_b) independently tune the total hole density ν (measured in units of the moiré density) via (V_a + V_b) and the out-of-plane electric field E through (V_a – V_b). The holes are generally distributed in both moiré layer (ν_m) and monolayer (ν_s); ν = ν_m + ν_s. We probe the correlation effects as a function of (ν, E) by optical spectroscopies and determine the charge gap energy by capacitance measurements. We further demonstrate charge localization and emergence of local magnetic moments in the monolayer induced by interlayer Coulomb correlation through magnetic circular dichroism (MCD) measurements. Details of the device fabrication and measurement methods are provided in Methods. Unless specified otherwise, all the measurements are performed at 3.4 K.

Figure 1e,f illustrates the doping dependence of the optical reflection contrast and photoluminescence (PL) spectra of the heterostructure, respectively, at E = −34 mV nm⁻¹. In the reflection spectrum, we can identify resonances from moiré excitons25 and the 1s and 2s excitons of the monolayer (Fig. 1c shows the spectrum at zero doping). The PL at around 1.4 eV arises from the recombination of holes in WSe2 and electrons in WS2 of the moiré layer7 (linecuts at...
The 2/3 and 1/2) in the order of decreasing charge gap size. However, insulating states on doping in the monolayer. We clearly identify five insulating states in the moiré layer, including the band density \( \nu \) in the heterostructure. Schematic of the valence band alignment in the absence of an electric field. The dashed line denotes the Fermi level.

Fig. 1 | Semiconductor heterostructures for realization of a dipolar exciton fluid in a lattice. a, Schematic of a dual-gated heterostructure consisting of a WSe₂ monolayer and a WSe₂/WS₂ moiré layer that are separated by 2–4 layers of hBN. The top- and bottom-gate voltages \( V_t \) and \( V_b \) control the doping density \( \nu \) and out-of-plane electric field \( E \) in the heterostructure. b, Schematic of the valence band alignment in the absence of an electric field. The dashed line denotes the Fermi level. c, Reflection contrast spectrum at \( \nu = 0 \) and \( E = -34 \text{ mV nm}^{-1} \) from device 1, showing two moiré excitons and the 1s and 2s excitons of the monolayer. d, Left, electronic Mott insulator at \( \nu = 1 \) (one hole per moiré site) when all the holes reside in the moiré layer in the absence of an electric field. Middle, some of the holes are transferred to the WSe₂ monolayer under a positive electric field. They are tightly bound to the empty moiré sites to minimize the total Coulomb repulsion energy. Right, the empty sites in the moiré layer are equivalent to electrons under a particle–hole transformation. A dipolar exciton fluid in the moiré lattice emerges. e,f, Reflection contrast (e) and PL (f) spectrum of the heterostructure as a function of doping density at \( E = -34 \text{ mV nm}^{-1} \).

The correlated insulating states can also be identified by the PL of the moiré layer with suppressed emission intensity and abrupt energy shift. The latter is presumably related to the chemical potential jump with doping across a correlated insulating state. Further theoretical studies are required to fully understand the microscopic mechanism. The spectral responses below \( \nu \approx 1/3 \) are likely distorted by the nonlinear gating effect from the non-ideal electrical contact to the heterostructure at low doping densities and low temperatures. Below, we explore the \( (\nu, E) \) parameter space for new correlated states using PL as a probe.

Figure 2a,b shows the doping dependence of the reflection spectrum of the monolayer 1s exciton and the PL spectrum of the moiré layer, respectively, at four representative electric fields. At \( E = -10 \text{ mV nm}^{-1} \), the spectra for \( \nu \leq 1 \) resemble those shown in Fig. 1e,f. For \( \nu > 1 \), the 1s exciton turns into an attractive polaron (redshifted branch, denoted as \( X^+ \)) and a repulsive polaron (blueshifted branch, not labelled), which signal hole doping into the monolayer. The reflection change at \( \nu = 1 \) is correlated with a local intensity minimum and an abrupt energy shift of the PL. As \( \nu \) increases further, the PL follows a smooth doping dependence till \( \nu \approx 1.8 \), whereas \( X^+ \) remains nearly unchanged. These results indicate that the holes are first doped into the moiré layer to completely fill the first Hubbard band; further doping introduces holes into the monolayer till \( \nu \approx 1.8 \), at which the chemical potential reaches the second Hubbard band of the moiré layer. The state at \( \nu = 1 \) is an electronic Mott insulator. For \( \nu \) between 1.0 and 1.8, the doping density in the moiré layer is fixed at \( \nu_m = 1.0 \).

At larger electric fields \( E = 15 \text{ and } 18 \text{ mV nm}^{-1} \), the polaron feature \( X^+ \) appears at smaller \( \nu \) (below 1) in the reflection spectrum; accordingly, the extended region of \( \nu_m = 1 \) emerges at larger \( \nu \) (above 1) in the PL spectrum. In addition, the electron crystal states can no longer be identified. These results support the fact that the holes are now shared between the monolayer and moiré layer before the density in the moiré layer reaches \( \nu_m = 1 \). The electron crystal states are quantum melted because the extended Coulomb repulsion in the moiré layer that induces these states is screened by the holes in the monolayer.

Remarkably, we observe an abrupt energy shift in the attractive polaron resonance as well as the PL at total filling \( \nu = 1 \). This signifies a chemical potential jump and emergence of a new correlated insulating state. Such a state is unexpected if the monolayer and moiré layer are uncoupled. In that case, both layers should be
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charge compressible. The state is consistent with a dipolar excitonic insulator induced by the strong interlayer Coulomb interaction (Fig. 1d): holes in the monolayer ($\nu_f$) and in the moiré layer ($\nu_m$) with $\nu_m + \nu_f = 1$ effectively form dipolar excitons with exciton density $\nu_X = \nu_f$. As the electric field increases, the exciton density increases, the binding energy decreases and the insulating state weakens. Above the exciton Mott density (around $1.4 \times 10^{12}$ cm$^{-2}$, measurement shown below), the exciton fluid turns into an electron–hole plasma$^{23}$. The state can no longer be observed at $E \gtrsim 30$ mV nm$^{-1}$.

The ($\nu, E$) parameter space is mapped out for the excitonic insulating state in Fig. 3a,b. It can be divided into four electrostatic doping regions. In region I, the monolayer is charge neutral ($\nu_f = 0$). In region II, the monolayer is doped and the moiré layer is at $\nu_m = 1$. 

Fig. 2 | Optical responses under an out-of-plane electric field. a, b, Optical reflection spectrum of the 1s exciton of the monolayer (a) and the PL spectrum of the moiré layer (b) versus doping density (device 1). From the top to bottom row, the out-of-plane electric field is –10, 15, 18 and 30 mV nm$^{-1}$. On doping in the monolayer, the 1s exciton feature turns into an attractive polaron (redshifted branch, $X^+$) and a repulsive polaron (blueshifted branch, not labelled). The PL of the moiré layer exhibits an abrupt energy shift and intensity suppression at the correlated insulating states. A new correlated insulating state is observed at $\nu = 1$ under $E = 15$ and 18 mV nm$^{-1}$; it can no longer be identified at $E = 30$ mV nm$^{-1}$. 

5,000 14,000
1.69 1.71 1.73 1.75
1.35 1.40 1.45 1.50
Energy (eV)
0 3,500
–10 mV nm$^{-1}$
15 mV nm$^{-1}$
18 mV nm$^{-1}$
30 mV nm$^{-1}$

0 3,500
0.5 2.0
1.0

Reflection (a.u.)

PL intensity (a.u.)

Doping density, $\nu$

Energy (eV)

Doping density, $\nu$

Energy (eV)

Doping density, $\nu$

Energy (eV)

Doping density, $\nu$

Energy (eV)
In region III, the doping density in both layers is less than 1 (region of interest). In region IV, the moiré layer is charge neutral ($\nu_m = 0$). We use two spectral responses to identify these regions. The peak amplitude of the 1$s$ exciton reflection contrast is sensitive to doping in the monolayer. We determine the boundary between the neutral and doped monolayer (dash-dotted lines) in Fig. 3b. The PL exhibits an intensity minimum when the Fermi level touches a Hubbard band (both top and bottom) of the moiré layer. We determine the boundaries of the first Hubbard band (dashed lines) in Fig. 3a. Details of the doping-region assignment and maps of other spectral responses, including the PL peak energy and attractive polaron reflection intensity, are provided in Methods and Extended Data Figs. 6 and 7.

The optical studies (Fig. 3a) suggest an excitonic insulator over a large range of electric fields and a narrow range of doping densities around total filling $\nu = 1$ in region III. We quantitatively characterize the state by capacitance measurements. The capacitance device is similar to that shown in Fig. 1a with a local bottom gate (Extended Data Fig. 4a). The top gate heavily dopes the contact region of the heterostructure to achieve good electrical contacts, whereas the local bottom gate tunes the doping density in the sample of interest. The accessible range of ($\nu, E$) is, thus, limited to maintain good electrical contacts. The differential gate capacitance $C$ is measured by collecting charges on the bottom gate as a small a.c. voltage is applied on the heterostructure. We expect $C \approx C_g$ if the heterostructure is charge compressible (conducting) and $C \leq C_g$ if it is charge incompressible (insulating), where $C_g$ denotes the geometrical bottom-gate-to-sample capacitance. Figure 3c shows the measured $C/C_g$ as a function of ($\nu, E$) at 20 K and 443 Hz. The boundaries of different doping regions (dashed lines) are obtained from optical measurements on the same device. In Region III, we observe $C < C_g$ around total filling $\nu = 1$ over an extended range of electric fields and $C \approx C_g$ in the background. The charge-incompressible state revealed by capacitance is fully consistent with that obtained...
from optical studies (Fig. 3a and Extended Data Fig. 7) (the larger electric field values here compared with Fig. 3a,b are due to a thinner hBN spacer in the capacitance device). The state persists up to around 120 K. Details of the capacitance measurements and results at other temperatures are included in Methods and Extended Data Figs. 4 and 5.

We evaluate the charge gap of the excitonic insulator by integrating the capacitance dip, that is, \( \int dV \chi (1 - \frac{\chi}{\chi_0}) \). Figure 3d summarizes the electric-field dependence of the charge gap at varying temperatures (in contrast, the charge gap of the electronic Mott insulator in bare WSe2/WS2 moiré bilayers is nearly independent of \( E \) (ref. 1)). The top axis shows the corresponding exciton density \( \chi_0 \) from electrostatics calculations. The smooth electric-field dependencies show that the exciton fluid is compressible (that is, the holes can be continuously transferred between the layers). The gap energy at a small exciton density and low temperature (around 30 meV) provides a measure of the exciton binding energy, which is consistent with our estimate based on the on-site repulsion energy. The charge gap vanishes at large values of \( \chi_0 \); at low temperatures, it is around 0.7 (1.4 \times 10^{12} \text{cm}^{-2}; Extended Data Fig. 7). This provides an estimate of the Mott density, beyond which the charge-incompressible excitonic insulator transitions into a charge-compressible electron–hole plasma at low temperatures. The large exciton binding energy, high Mott density and ionization temperature observed in our experiment are consistent.

Finally, we demonstrate charge localization and emergence of local magnetic moments in the monolayer induced by the strong interlayer Coulomb correlation in the excitonic insulating state. We measure the magnetic response of the monolayer under an out-of-plane magnetic field by MCD measurements near its repulsive polaron feature (Methods, Extended Data Fig. 3), which has been shown to be proportional to the sample magnetization. Figure 4a shows the magnetic-field dependence of the MCD at \( \nu = 1 \) and \( \nu_l = \nu_s = 0.3 \). The behaviour at other temperatures is discussed in Methods (ref. 8). The top axis shows the corresponding exciton density as a function of temperature at \( \nu_s = 0.30 \) and 0.45. The error bars are estimated from the linear fit to the low-field data in a. The magnitude of the Weiss constant follows the Curie–Weiss dependence (solid lines) at high temperatures with small negative Weiss constants (\( x \) intercepts). The magnitude of the Weiss constant is compatible with weak antiferromagnetic exchange interactions between the distant local moments.

\[ \theta \text{ denote the sample temperature and Weiss constant, respectively.} \]

\[ \text{Such a behaviour is in contrast to that of an isolated monolayer at comparable hole-doping densities but compatible with that of local magnetic moments induced by excitation formation.} \]

The Weiss constant obtained from the fits (solid lines) changes from approximately 0 to approximately 0.3 to 0.45. It suggests a weak antiferromagnetic exchange interaction between the distant local moments in the monolayer. This result highlights the strong interlayer correlation effect in this system.

In conclusion, we have realized equilibrium exciton fluids in a lattice using the platform of semiconductor moiré heterostructures. Future studies involving Coulomb drag and counterflow measurements are required to establish the bosonic ground states, investigate the BCS–BEC crossover and understand the effects of mismatched electron and hole masses. Intermediate hBN spacer thickness (2 nm), which suppresses single-particle interlayer tunneling as strong interlayer Coulomb correlation is maintained, is a promising parameter region to explore. Our results pave the path for the realization of exotic correlated bosonic states, such as bosonic Mott insulator, Wigner solid, supersolid and FFLO state under finite electron–hole density imbalance.

**Online content**

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Methods

Device fabrication. We fabricate dual-gate devices of transition metal dichalcogenide (TMD) heterostructures using the reported layer-by-layer dry transfer technique. Details have been reported in earlier studies[12,13]. The capacitance device characteristics described throughout this Letter are obtained from bulk crystals grown by the chemical vapour transport method (HQ Graphene). Angle alignment of the WSe₂ and WS₂ monolayers in the moiré bilayer is achieved with the assistance of the second-harmonic generation[14]. Both 0°- and 60°-aligned WSe₂/WS₂ bilayers are involved in this study; no noticeable difference is observed in the two cases. The Coulomb-coupled WS₂ monolayer is not angle aligned. The thickness of the hBN spacer between the moiré layer and WS₂ monolayer is determined by atomic force microscopy studies of the devices. No obvious dependence on the spacer thickness is observed, which is consistent with a suppression of interlayer tunnelling as well as negligible moiré printing on the monolayer. The top and bottom gates are nearly symmetric; they consist of hBN gate dielectric and few-layer graphite gate electrodes; the thickness of the gate dielectric is typically between 10 and 40 nm. Few-layer graphite is also used as contact electrodes in devices used for the optical studies. Pt electrodes are used to achieve better p-type contacts to the TMD heterostructures for the a.c. capacitance studies.

Optical measurements. The optical characteristics of the WSe₂/WS₂ heterostructure are described throughout this Letter. The optical reflection contrast is also observed across the moiré layer region, which is consistent with the reported results[15]. We integrate the MCD spectrum across the moiré layer as a function of magnetic field at several representative electric fields and fixed total doping density $\nu = 1$. We focus on the spectral range of the attractive and repulsive polaron states of the monolayer. MCD is not observed at zero magnetic field, and the monolayer’s band gap monotonically increases with the magnetic field and saturates at around 2–3 T. The MCD is resonantly enhanced near the energy of both repulsive and attractive polarons; it changes sign across the polaron resonances, which is consistent with the reported results[15]. We integrate the MCD spectrum over a narrow spectral window (boxed) near the repulsive polaron resonance (Fig. 4). The integrated MCD is proportional to the valley magnetization of the carriers in the WSe₂ monolayer[16]. For the PL measurements, we employ a He–Ne laser (633 nm) as the excitation source; the incident power is 30 mW.

Capacitance measurements. We follow the design reported elsewhere[8,9] to achieve good electrical contacts to the TMD heterostructure for the capacitance measurements. Extended Data Fig. 4a shows the schematic of the top gate (global gate) covers the entire TMD heterostructure; the bottom gate (local gate) covers it partially. The overlapped area is the region of interest; the non-overlapped area is the contact region. A negative voltage is applied to the global gate to introduce holes into the entire TMD heterostructure; the bottom local gate depletes holes in the region of interest (red box). The device structure limits the range of $(\epsilon, E)$ that can be accessed in the capacitance study because a finite negative top-gate voltage is required to maintain hole doping in the contact region to achieve good electrical contacts.

The TMD heterostructure is d.c. grounded. An a.c. excitation voltage (amplitude, 10 mV; frequency, 443–3,317 Hz) is applied to the TMD heterostructure. Charge is collected from the bottom gate and sent to an on-chip high-electron-mobility transistor amplifier and detected by a lock-in amplifier. The differential capacitance $C$ is the ratio of the collected charge to the excitation voltage. We verify that the result is largely independent of the excitation frequency. Extended Data Fig. 4b shows $C$ in the units of $C_0$ as a function of $(\epsilon, E)$ at 20 K, where $C_0$ is the geometrical capacitance defined by the thickness and dielectric constant of the hBN gate dielectric between the bottom gate and TMD heterostructure. The electrostatic doping regions are determined by optical responses (Extended Data Fig. 4c,d). Only regions III and IV can be accessed. The diagonal dashed green line corresponds to a constant top-gate voltage of $-1.4$ V. It marks the onset of charging into the device. Top-gate voltage above the green line is required to dope the contact region and turn on the channel for capacitance measurements.

Extended Data Fig. 4e shows the PL peak intensity from the moiré layer as a function of $(\epsilon, E)$. The PL intensity is sensitive to doping in the moiré layer[17,18]. The dashed line marks the boundary between regions III and IV, corresponding to the doped and neutral moiré layer, respectively. Extended Data Fig. 4d shows the peak reflectance contrast of the monolayer 1$s$ exciton, which is sensitive to doping in the monolayer[19,20]. The black dashed line marks the onset of hole doping in the monolayer. It is consistent with the onset of a finite capacitance signal (Extended Data Fig. 4b). A change in the $1$s$ reflection contrast is also observed across the green dashed line, which marks the onset of charging into the device. Detailed discussions on the electrostatic doping regions are provided below.

The measured differential capacitance accesses the quantum capacitance $C_Q$, which represents its electronic charge compressibility:

$$\frac{dC}{d\nu} \approx \frac{e^2}{\hbar C} + \frac{e^2}{\hbar Q}$$

We expect $C_C \ll C_Q$ for a charge-compressible state and $C_C \gg C_Q$ for a charge-incompressible state. A charge-incompressible state is clearly observed at $\nu = 1$ over an extended range of electric field. The state weakens with an increasing electric field. This is fully consistent with the optical results (Extended Data Fig. 7). The absence of a charge-incompressible state at the moiré filling $\nu = 1$ also shows negligible moiré printing onto the monolayer from the moiré layer. The characteristic temperature of the superconducting states in the moiré layer (Fig. 4) is an order of magnitude lower than the Fermi level level $k_B T_c \approx 10$ mK. The measured differential capacitance is consistent with the observed weak attractive polaron reflection (Extended Data Fig. 6b). The feature intensity of the $1s$ exciton since a charge-neutral monolayer gives rise to strong $1s$ exciton and weak attractive polaron reflection[21]; these spectral responses are reversed for a doped monolayer. This is consistent with the experimental results shown in Extended Data Fig. 6d.

Next, we examine the PL intensity from the recombination of holes in WSe₂ and electrons in WS₂ of the moiré layer (Extended Data Fig. 6a). A large PL intensity (red) is observed when the moiré layer is charge neutral ($\nu = 0$). A local PL intensity minimum emerges when the Fermi level touches a Hubbard band edge (Extended Data Fig. 6c). The PL intensity varies between two and four layers of stacked TMD, and the first Hubbard band is flatter than the second. These results are fully consistent with the reported properties of angle-aligned WSe₂/WS₂ bilayers[22].

The map of the attractive polaron response is expected to be complementary to that of the $1s$ exciton since a charge-neutral monolayer gives rise to strong $1s$ exciton and weak attractive polaron reflection[21]; these spectral responses are reversed for a doped monolayer. This is consistent with the experimental results shown in Extended Data Fig. 6d.

Electrostatic phase diagram. Extended Data Fig. 6 shows the electrostatic phase diagram with a larger parameter range for $(\epsilon, E)$ than that in Fig. 3. Extended Data Fig. 6a–c shows the (peak) PL intensity of the moiré layer, the (peak) reflection intensity of the $1s$ exciton and the attractive polaron of the WSe₂ monolayer, respectively. The dashed lines outline the boundaries of the different electrostatic doping regions. Below, we describe how these boundaries are determined from the spectral responses.

We first examine the $1s$ exciton reflection (Extended Data Fig. 6b). The feature is sensitive to doping in the monolayer[19,20]. We can draw the boundary between the charge-neutral (red) and hole-doped region (blue) based on the $1s$ response. A schematic of the boundary is shown in Extended Data Fig. 6d (blue line). Along the line, the monolayer remains charge neutral ($\nu = 0$) and the holes are solely doped into the moiré layer ($\nu = 1$). The line traces the valence band edge of the monolayer as it shifts across the Hubbard bands of the moiré layer under an applied electric field. It, therefore, probes the chemical potential as a function of doping density of the moiré layer. In particular, the boundary is vertical over a range of electric fields, $\Delta E$, when the monolayer band edge is located inside a charge gap of the moiré layer. The product of $\Delta E$ and the separation between the monolayer and moiré layer, $d$, approximately corresponds to the charge gap energy. When the monolayer band edge overlaps with a Hubbard band, the boundary line gains a finite slope, which is inversely proportional to the thermodynamic density of the states of the particular Hubbard band. Our data shows that the $\nu = 1$ Mott gap is $= 1.3$, and that the $\nu = 2$ Mott gap is $= 2.3$, and the first Hubbard band is flatter than the second. These results are fully consistent with the reported properties of angle-aligned WSe₂/WS₂ bilayers[22].

The map of the attractive polaron response is expected to be complementary to that of the $1s$ exciton since a charge-neutral monolayer gives rise to strong $1s$ exciton and weak attractive polaron reflection[21]; these spectral responses are reversed for a doped monolayer. This is consistent with the experimental results shown in Extended Data Fig. 6c. We summarise the distinct regions of the electrostatic phase diagram (Extended Data Fig. 6f) as follows:

- Region I: $\nu = 0$ and $\nu = \nu_C$. No electric field dependence is seen here because all the holes reside in the moiré layer. Electron crystal states can be clearly identified as vertical straight lines at fractional filling factors. The PL features below $\nu \approx 1/3$ are probably artifacts from the nonlinear charging effect.
• Region II: \( \nu_m = 1 \) and \( \nu_f = \nu - 1 \). Here the Fermi level is inside the Mott gap of the moiré layer.
• Region III (region of interest): \( \nu_f > 0 \) and \( \nu_m < 1 \). Both layers are hole doped and the applied electric field transfers holes between the layers. The electron crystal states disappear in this region because of hole screening from the WSe\(_2\) monolayer.
• Region IV: \( \nu_m = 0 \) and \( \nu = \nu_f \).

Similarly, we can assign regions I', II' and III', which are the second Hubbard band copies of regions I, II and III, respectively. They correspond to (\( \nu_f = 0 \) and \( \nu = \nu_m \)), (\( \nu_m = 2 \) and \( \nu_f = \nu - 2 \)) and (\( \nu_f > 0 \) and \( 1 < \nu_m < 2 \)), respectively.

### Data availability
Source data are provided with this paper. Additional data that support the findings of this study are available from the corresponding authors upon reasonable request.

### Code availability
The code that supports the plots within this paper is available from the corresponding authors upon reasonable request.

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### Author contributions
J.G. fabricated the devices, performed the optical measurements and analysed the data. J.G. and L.M. performed the capacitance measurements and analysed the compressibility data. S.L. and J.C.H. grew the bulk WSe\(_2\) crystals. K.W. and T.T. grew the bulk hBN crystals. J.G., J.S. and K.F.M. designed the scientific objectives and oversaw the project. All the authors discussed the results and commented on the manuscript.

### Competing interests
The authors declare no competing interests.

### Additional information
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Extended Data Fig. 1 | Optical micrograph of device 1. The constituent layers are outlined by lines of different colors and labeled. The effective device area is shaded in grey. The scale bar is 10 μm.
Extended Data Fig. 2 | PL spectrum at different hole doping densities. The data corresponds to the data in Fig. 1f at $E = -34$ mV/nm. The spectra are vertically displaced for clarity. The left axis denotes the scale bar for the PL intensity. The right axis denotes the hole doping density for each vertically displaced spectrum. An abrupt energy shift is observed at $\nu = 1$. 
Extended Data Fig. 3 | MCD spectrum as a function of magnetic field at ν = 1 and two representative electric fields. The spectra focus on the attractive and repulsive polaron resonances of the 1s exciton of the WSe₂ monolayer. The MCD is enhanced at the resonances. The box shows the spectral window, over which the MCD is integrated. The magnetic-field dependence of the integrated MCD is shown in Fig. 4 of the main text.
Extended Data Fig. 4 | Capacitance measurements. a. Schematic of device structure and electrical connections for the AC capacitance measurement. b–d. Differential capacitance (b), PL peak intensity of the moiré layer (c), and 1s exciton peak reflection contrast of the monolayer (d) as a function of doping density and electric field ($\nu$, $E$). Electrostatic doping region III and IV in b are determined by the optical responses in c and d. A charge-incompressible state is observed at $\nu = 1$ in region III, which weakens with increasing electric field. The dashed line in c marks the onset of doping into the moiré layer; it separates region III and IV. The black and green dashed line in d mark the onset of charging into the monolayer and in the device, respectively.
Extended Data Fig. 5 | Temperature dependence of differential capacitance. Differential bottom-gate capacitance as a function of (ν, E) at 20 K (a), 40 K (b), 60 K (c), 80 K (d), 120 K (e), and 150 K (f). The charge-incompressible state at ν = 1 disappears around 120 K.
Extended Data Fig. 6 | Electrostatics phase diagram. a-c, PL peak intensity of the moiré layer (a) and reflection peak intensity of the 1s exciton (b) and the attractive polaron (c) of the monolayer as a function of doping density and out-of-plane electric field (ν, E). The PL arises from recombination of the interlayer excitons in the moiré bilayer. The dashed lines are the boundaries that separate the different electrostatic doping regions (labeled in d) and are determined from the optical responses. The data in a-c are well correlated with one another. d, A schematic of the electrostatics phase diagram of the TMD heterostructure as described in Methods.
Extended Data Fig. 7 | PL signatures of excitonic insulator. The PL peak intensity (a) and peak energy (b) of the WSe₂/WS₂ interlayer moiré excitons as a function of doping density and out-of-plane electric field ($\nu$, $E$). The different electrostatic doping regions are labeled. The feature at $\nu = 1$ in region III corresponds to the correlated excitonic insulating state. c. Doping dependence of the PL peak energy at varying electric fields. The abrupt energy shift at $\nu = 1$ reflects a chemical potential jump for the correlated insulating state. The energy shift disappears above the critical field value around 30 mV/nm, at which the exciton Mott density (approximately 0.7 moiré density) is reached. d. Dependence of the PL peak shift at $\nu = 1$ as a function of electric field (bottom axis) and exciton density in the units of the moiré density (top axis). The Mott density is consistent with the value from the capacitance measurement. The smaller electric-field range here is due to a thicker hBN spacer compared to the capacitance device.