Exciton density waves in Coulomb-coupled dual moiré lattices

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Strongly correlated bosons in a lattice are a platform that can realize rich bosonic states of matter and quantum phase transitions1. While strongly correlated bosons in a lattice have been studied in cold-atom experiments2–4, their realization in a solid-state system has remained challenging5. Here we trap interlayer excitons—bosons composed of bound electron–hole pairs, in a lattice provided by an angle-aligned WS2/bilayer WSe2/WS2 multilayer. The heterostructure supports Coulomb-coupled triangular moiré lattices of nearly identical period at the top and bottom interfaces. We observe correlated insulating states when the combined electron filling factor of the two lattices, with arbitrary partitions, equals \( \frac{1}{3} \), \( \frac{2}{3} \), \( \frac{4}{3} \) and \( \frac{5}{3} \). These states can be interpreted as exciton density waves in a Bose–Fermi mixture of excitons and holes6,7. Because of the strong repulsive interactions between the constituents, the holes form robust generalized Wigner crystals8–11, which restrict the exciton fluid to channels that spontaneously break the translational symmetry of the lattice. Our results demonstrate that Coulomb-coupled moiré lattices are fertile ground for correlated many-boson phenomena12,13.

Moiré materials have emerged as a highly controllable quantum system for exploring a plethora of correlated electronic states14–18. Mott insulators10,19–21 and generalized Wigner crystals8–11 have been demonstrated for electrons in a moiré lattice formed in semiconducting transition metal dichalcogenide bilayers. However, strongly correlated excitons are generally not achievable in a single moiré lattice22,23 because of the short exciton lifetime. Two symmetric moiré lattices separated by a thin barrier have been proposed to realize exciton fluids in a lattice6 (Fig. 1a–c). Excitons here are the bound states of electrons in one lattice and empty sites (‘holes’) directly above (or below) in the other lattice; both are in the lowest moiré minibands formed from the conduction (or valence) bands of the host semiconductor. Strong binding is expected when the electronic correlation is strong in each lattice, and the lattice separation, \( d \), is small compared to the moiré period, \( a_M \). Similar to the case of Coulomb-coupled electron–hole double layers without a lattice6–12, the spatially separated double lattice structure substantially suppresses interlayer electron tunnelling and allows studies of strongly correlated excitons in thermal equilibrium. Many bosonic phases, including excitonic Mott insulators, Wigner solids, superfluids and supersolids, have been predicted12,13.

Here, we demonstrate two Coulomb-coupled moiré lattices of nearly identical period in angle-aligned WS2/bilayer WSe2/WS2 multilayers (Fig. 1b,c). In contrast to the recently studied excitons in a moiré lattice Coulomb coupled to a lattice-free monolayer33,34, both electrons and holes here are in the flat moiré minibands, and much stronger correlations are expected. We observe new excitonic insulating states at a combined electron filling factor of the two lattices, \( \nu = \frac{1}{3}, \frac{2}{3}, \frac{4}{3} \) and \( \frac{5}{3} \) in addition to that at \( \nu = 1 \) reported in the literature33,34. These states are distinct from the generalized Wigner crystals in a single moiré lattice8–11. They possess a fluid of excitons and provide a rare realization of exciton density waves with exciton density spontaneously breaking the translational symmetry of the lattice6,12,13.
Because of the 4% lattice mismatch for the two transition metal dichalcogenides in the angle-aligned WS₂/bilayer WSe₂/WS₂ multilayer, triangular moiré lattices of period $a_w$ are formed at both the top and bottom WS₂/WSe₂ interfaces. Here, $a_w$ is insensitive to small twist angle variations, which makes the creation of two nearly identical moiré lattices possible. We focus on the case of electron doping to maximize the correlation effects since the conduction minibands are flatter in this system. Electrons are located in the WS₂ layers as shown by the band alignment in Fig. 1b. The WSe₂ bilayer separates the moiré lattices with a centre-to-centre distance $d = 2 \, \text{nm}$ and acts as a tunnel barrier with a barrier height around 130 meV (ref. 19; Methods for discussions on the band alignment). To further suppress the electronic coupling between the two lattices, we align the two WS₂ monolayers at 180°; interlayer electron tunnelling is spin forbidden due to spin–valley locking in each monolayer. The physics is dominated by interlayer Coulomb interactions. As shown in Fig. 1c, the coupled moiré lattices are grounded and encapsulated in a top and bottom gate made of hexagonal boron nitride (hBN) dielectrics and graphite gate electrodes. The two gates independently control the combined filling factor $v$ (or the total doping density) and the out-of-plane electric field $E$ in two moiré lattices. The latter tunes the relative band alignment and distribution of the electrons between the two lattices at a fixed $v$.

To probe the insulating states in the coupled moiré lattices, we employ an optical sensing technique by placing a WSe₂ monolayer above the top WS₂ layer. A thin (~1–2 nm) hBN spacer is introduced to prevent direct electronic coupling between the sensor and the sample, but is sufficiently thin so that the charge compressibility or dielectric constant of the sample can be probed through dielectric screening of the 2s exciton resonance of the WSe₂ sensor. Unless otherwise specified, all results are obtained at a temperature of 3.5 K.

Figure 1d–f shows the reflectance contrast (RC) spectrum of the sensor 2s exciton as a function of gate voltage (lower axis) and $v$ (upper axis). The three examples correspond to electron doping solely in the bottom lattice (Fig. 1d), in the top lattice (Fig. 1e) and evenly in two lattices (Fig. 1f). The accessible doping range in these examples is limited (and also different) in order to keep the sensor charge neutral. An insulating state in the sample is identified when the 2s exciton resonance exhibits a blueshift and an enhanced RC. When the electrons are solely in one of the lattices (Fig. 1d,e), the observation is similar to the reported result in single WS₂/WSe₂ moiré structures. In particular, insulating states are observed at $v = \frac{1}{3} + \frac{2}{3}$ and $\frac{1}{2}$ Compared to a single moiré structure, fewer correlated insulating states at fractional fillings can be identified here; the weaker ones are presumably suppressed by the stronger dielectric screening and/or disorder effects in coupled moiré lattices. In addition, we observe an overall stronger renormalization of the 2s exciton resonance in Fig. 1e than in Fig. 1d. This is a manifestation of the higher sensor sensitivity to the top lattice because of its closer proximity.

When the electrons are doped evenly in the two lattices by setting $E = 0 \, \text{V nm}^{-1}$ (Fig. 1f), we observe additional insulating states at $v = \frac{4}{3}$ and $\frac{5}{3}$. These states are absent in Fig. 1d,e because of the substantially weaker correlations for the second Hubbard band in a single moiré structure. This example illustrates the sensitivity of the correlated states on the charge configurations in both lattices.

We map the dielectric constant of the coupled moiré lattices as a function of v and $E$ in Fig. 2a. We represent the dielectric constant by the amplitude of the 2s exciton RC in the sensor (Extended Data Fig. 1), as in a recent study; an enhanced $R_{2s}$ reveals a charge-incompressible (insulating) state. The map is dominated by the response of the top moiré lattice because it is closer to the sensor. As expected, the largest $R_{2s}$ is observed when the entire structure is charge incompressible, for instance, when both lattices have integer fillings. We also observe extended regions with enhanced $R_{2s}$ where the top lattice is charge incompressible and the bottom lattice is generally charge compressible (orange regions in Fig. 2b). They are assigned as $v_t = 0, 1$ and 2 with the chemical potential inside the semiconductor bandgap, the Mott gap and the first moiré bandgap of the top lattice, respectively ($v_t$ and $v_b$ denote the filling factor of the top and bottom lattices, respectively).
The regions are extended because a finite electric field is required to Stark shift the minibands in the two lattices to overcome the finite charge gap of the top lattice33,40. To complete the electrostatic phase diagram, we add in Fig. 2d the mirror image of the orange regions with respect to zero electric field (blue regions) to represent regions of a gapped bottom moiré lattice (νb = 0, 1 and 2) but a charge-compressible top lattice. The sensor 2s exciton is, however, effectively screened by the top lattice and no longer sensitive to the compressibility of the bottom lattice. The electrostatic phase diagram is fully consistent with an independent measurement based on the moiré excitons in WS2 (Fig. 2c). The RC of the fundamental moiré exciton in WS2, RWS2, decreases significantly when at least one of the WS2 layers is electron doped33 (Extended Data Fig. 2). The boundary between the high and low values of RWS2 agrees well with the combined boundary of the region of νt = 0 and νb = 0 (dashed lines). The Methods contain additional discussions on the electrostatic phase diagram.

We now focus on the unfilled regions in the phase diagram, in which both lattices would generally be charge compressible in the absence of inter-lattice (or interlayer) correlations. Figure 3a is a closer image of the orange regions with respect to zero electric field (enclosed by dashed lines). Remarkably, we identify incompressible states from the enhanced RWS2 at ν = 1, 2, 1/3 and 2/3 for all electric fields in the given range; and the electric-field dependence of RWS2 is smooth for all states (Fig. 3b). Figure 3c shows a horizontal line cut at E = 0 V nm⁻¹ under varying temperatures. The incompressible states at fractional fillings disappear around 30–35 K, but the ν = 1 state persists up to much higher temperatures.

The insulating states outside the dashed box in Fig. 3a are well understood, with each lattice in an insulating state (independent of the other). For instance, the ν = 1 state corresponds to (νt, νb) = (0, 1) under large upward electric fields and (1, 0) under large downward fields. This is an `intralayer' Mott insulator stabilized by the onsite Coulomb repulsion within each layer. Similarly, the ν = 1/3 state corresponds to (νt, νb) = (0, 1/3) and (1, 1/3) that is, an `intralayer' Wigner crystal stabilized by the long-range Coulomb repulsion within each layer.

The nature of the insulating states inside the dashed box is completely different. Here, the electrons are continuously transferred from one lattice to the other by the electric field. They arise from inter-lattice correlations and can be viewed as excitonic insulators33,34. We first discuss the case of ν = 1 with one lattice unfilled and the other being a Mott insulator. Electrons are transferred from the Mott insulator to the empty lattice but remain bound to the empty sites in the original lattice by the inter-lattice `onsite' Coulomb repulsion, stabilizing an `interlayer' Mott insulator. These bound states can be viewed as interlayer excitons, which can hop around the lattice (Fig. 4a). The result is an excitonic insulator that supports an exciton fluid with a dipole–dipole repulsion of (V′ − V′). Here, V′ and V′ (V′ ≤ V in the limit d ≪ a) denote the long-range Coulomb repulsion in the same lattice and between the lattices, respectively (Methods for estimates of V and V′). The smooth electric-field dependence in Fig. 3b suggests that the exciton density can be continuously varied (that is, the state is exciton compressible). The ground state of the exciton fluid is expected to be a superfluid in the weak-disorder limit32,41. Similar to the intralayer Mott insulator, the exciton density preserves the translational symmetry of the lattice.

At total fractional fillings, we start with an empty lattice and a generalized Wigner crystal in the other lattice. We illustrate the cases of ν = 1/3 and 2/3 (Fig. 4b). The physics of ν = 1/3 and 2/3 are nearly identical. Electrons are transferred from the generalized Wigner crystal to the empty lattice but remain bound to the empty sites in the original lattice by V′, also forming interlayer excitons. To minimize the total intra- and inter-lattice Coulomb repulsions (V and V′), the electrons from both layers combine to form an ‘interlayer’ Wigner crystal; this is supported by the non-topological nature of the fractional insulating states (Extended Data Fig. 3).
In the weak-disorder limit, they are expected to possess finite superfluid wavefunctions of \( \rho = \frac{1}{3} \) and \( \frac{1}{4} \). The latter breaks the translational symmetry of the lattice.

In summary, we have observed excitonic insulating states at total fractional fillings of \( \nu = \frac{1}{3}, \frac{1}{4}, \frac{1}{5} \) and \( \frac{1}{6} \) (and also \( \frac{1}{2} \) in some devices) in two Coulomb-coupled moiré lattices. The observation of these states requires strong electronic correlation within each lattice and between the two lattices, which are closely spaced (\( d \approx a_0 \)) but electronically decoupled. Perfect alignment of the two lattices is not required as long as the relative displacements are uniform and below the size of the electron Wannier functions (\( 2 \approx 3 \) nm) in the moiré lattice (Methods for more discussions). Future direct imaging studies by scanning tunneling microscopy are desired. The coupled moiré lattices demonstrated here open new doors in the search for exotic many-boson phenomena. In particular, the observed correlated insulating states can be viewed as exciton density waves emerged from an exciton–hole Bose–Fermi mixture (Methods). In the weak-disorder limit, they are expected to possess finite superfluid densities in the ground state and are therefore supersolids. These states provide a route to realize exciton supersolidity through the demonstration of spontaneous exciton phase coherence in future thermal transport and charge counterflow experiments.

Online content
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Methods

Device fabrication and operation

The devices (Fig. 1c) were fabricated using the layer-by-layer dry transfer method described in earlier studies.\(^1\) In short, flakes of monolayer WS\(_2\), monolayer and bilayer WSe\(_2\), few-layer h-BN and few-layer graphite were exfoliated from bulk crystals onto silicon substrates with a 285 nm oxide layer. They were identified by the reflectance contrast under an optical microscope and stacked in the desired sequence. The finished stack was transferred onto pre-patterned Au electrodes on silicon substrates. In the WS\(_2\)/2L-WSe\(_2\)/WS, moiré structure, one WS\(_2\) monolayer was angle aligned and the other was anti-aligned with the WSe\(_2\) bilayer. The two WS\(_2\) monolayers were cut from the same monolayer flake using an atomic force microscope tip. Optical second-harmonic generation was used to determine the crystal orientations prior to transfer. Bilayer WSe\(_2\) was used as a tunnel barrier rather than monolayer and trilayer because monolayer is too thin to effectively suppress interlayer tunnelling and trilayer would undesirably weaken the interlayer exciton binding. The monolayer WSe\(_2\) sensor was placed above the moiré structure with a 1–2 nm h-BN spacer and was not angle aligned with the moiré structure. The sensor and the moiré structure were grounded by the same few-layer graphite electrode. For the device shown in the main text, the thickness of the h-BN gate dielectric for the top and bottom gates was determined by atomic force microscopy to be \(d_{fg} = 25 \pm 0.3\) nm and \(d_{fg} = 28 \pm 0.3\) nm, respectively. The top and bottom gate voltages (\(V_{fg}, V_{fg}\)) independently tune the out-of-plane electric field and the doping density in the moiré structure. The field, \(E = \left(\frac{d_{fg}}{d_{fg}} - \frac{d_{fg}}{d_{fg}}\right)/2\), is defined as positive when it points from the top to the bottom moiré lattice. The normalized gate voltage, \(V_g = V_{fg} + (d_{fg}/d_{fg})V_{bg}\), is proportional to the doping density and is used in Fig. 1f. Using the gate voltages for the assigned insulating states, we determined the moiré density to be \((1.98 \pm 0.10) \times 10^{12}\) cm\(^{-2}\) for the top and bottom lattices, respectively. The two lattice periods are identical within the uncertainties.

Optical measurements

The optical RC was measured with the devices in a closed-cycle optical cryostat (attoDRY1000) at temperatures down to 3.5 K. A tungsten halogen lamp was used as the light source, and the incident power on the devices was kept below 0.8 nW. The spectrum of the reflected light from the devices was collected. The RC spectrum is defined as \((I-I_0)/I_0\), where \(I_0\) is the reference spectrum and \(I\) is the signal spectrum for a fixed doping density and out-of-plane electric field in the moiré structure. To obtain a RC near the 2s exciton resonance of the sensor, we kept the sensor charge neutral; we used the spectrum measured with a heavily electron-doped sensor as the reference, for which the 2s exciton is quenched. To obtain a RC near the WS\(_2\) moiré exciton resonances, we used the spectrum measured with a heavily electron-doped moiré structure as the reference, for which the moiré exciton resonance is nearly quenched. Extended Data Fig. 2 shows the RC spectrum of device SI, including both the 1s and 2s exciton resonances in the WSe\(_2\) sensor and the moiré excitons in WS\(_2\) as a function of gate voltages. The WSe\(_2\) moiré exciton resonances from the moiré structure have lower energies and are outside the spectral window. Extended Data Fig. 1 illustrates several horizontal line cuts from Extended Data Fig. 2 centred on the sensor 2s exciton and the WS\(_2\) moiré exciton. Multilayer thin-film analysis is required to describe the detailed line shape. For simplicity, we use the peak-to-peak variation of the features, \(R_{2s}\) and \(R_{1s}\), to denote the spectral weight of the 2s and the moiré exciton resonances, respectively. The moiré exciton resonances are relatively broad and the contribution from the two different WS\(_2\) layers cannot be spectrally resolved. The moiré exciton RC in Fig. 2c therefore includes contributions from both layers.

Electrostatic phase diagram of two Coulomb-coupled moiré lattices

The electrostatic phase diagram of Fig. 2b,d can be qualitatively understood as follows. Inside the three orange-shaded regions, electron filling in the top layer \(v_t\) is fixed at either 0, 1 or 2 as indicated by the labels (Fig. 2b). We first consider the region with \(v_t = 0\). At \(v_t = 0\), the Fermi level is inside the large semiconductor bandgap of the heterostructure. With increasing \(v_t\), electrons start to dope into the bottom layer for an upward field \((E < 0)\); they dope into the top layer for a downward field \((E > 0)\) for \(v_t = 0\). At the boundary of the region (denoted by an orange line), the Fermi level is fixed at the conduction band edge of the top layer. As \(v_t\) increases across this line, the Fermi level sweeps through the lower and upper Hubbard bands of the bottom layer. Vertical jumps in \(E\) are observed for the Fermi level inside the Mott gap \((v_t = 1)\) and the moiré bandgap \((v_t = 2)\) of the bottom layer. The jump size multiplied by the top–bottom layer separation corresponds to the charge gap size.\(^{34,40}\) With the Fermi level inside the lower or upper Hubbard band of the bottom layer, the boundary shows a linear dependence between \(E\) and \(v_t\), the slope of which is determined by the thermodynamic density of states.\(^{34,40}\)

The two other regions \((v_t = 1\) and 2) can be understood similarly. In particular, the lower (upper) boundary for \(v_t = 1\) corresponds to the reference point where the Fermi level is fixed to the lower (upper) Hubbard band maximum (minimum) of the top layer. These two reference boundaries trace through the lower and upper Hubbard bands of the bottom layer in a way similar to the reference boundary for \(v_t = 0\). The vertical distance in the electronic field between these two boundaries is proportional to the Mott gap size of the top layer. The region \(v_t = 2\) has similar interpretations except that the Mott gap of the top layer is replaced by the moiré bandgap. Furthermore, because the WS\(_2\)/2L-WSe\(_2\)/WS, moiré structure is symmetric, the discussions above apply equally well to the blue-shaded regions with \(V_{tg} = 0, 1\) and 2. The orange- and blue-shaded regions are symmetric about the \(E = 0\) line (Fig. 2d).

The white regions in Fig. 2d correspond to the Fermi level inside the Hubbard bands of both layers. The electrons are added to the system along the filling-factor axis and are continuously transferred between the two layers along the electric-field axis. The system is in general charge compressible except at total fractional filling factors corresponding to the excitonic insulating states.

Determination of band alignment in the heterostructure

In order to determine the band alignment in the dual moiré heterostructure, we first note that the natural bilayer WSe\(_2\) is an indirect gap semiconductor with a valence band maximum from the K/K’ valley and conduction band minimum from the Q valley.\(^4\) The indirect optical gap (about 1.57 eV) can be determined by photoluminescence (PL) measurements (Extended Data Fig. 4). Such PL measurement on the dual moiré heterostructure allows us to determine the conduction band barrier height. The results are shown in Extended Data Fig. 4; we keep the heterostructure at charge neutrality in our measurement. The PL spectrum shows, in addition to the indirect optical gap at 1.56 eV for the natural bilayer WSe\(_2\), an interlayer exciton resonance at 1.43 eV for the heterostructure (the higher energy features near 1.6–1.65 eV are intralayer moiré exciton PL for WSe\(_2\)). With these energy scales determined, we can construct the band alignment in Extended Data Fig. 4. The type-II band alignment remains in order to be consistent with the experimental result, in particular, the lowest energy interlayer exciton emission at 1.43 eV. The energy difference between the indirect optical gap of the bilayer WSe\(_2\) and the interlayer exciton resonance gives a conduction band barrier height of about 130 meV. The large barrier height allows us to treat the bilayer WSe\(_2\) as a tunnel barrier for the conduction electrons in the monolayer WS\(_2\).

Estimates of intralayer and interlayer Coulomb repulsions

The intralayer and interlayer Coulomb repulsions between two electrons separated by an intralayer distance \(r\) are \(V(r) = \frac{2\sqrt{\epsilon^2 e^4}}{4\pi\hbar c} \frac{2\hbar c}{\sqrt{m^* + 2m^*}}\) and \(\frac{e^2}{4\pi\epsilon_0 r}\), respectively.
and $V(r) \approx \frac{e^2}{4\pi\varepsilon_0 a} \sum_{k=-\infty}^{\infty} \frac{(-1)^k}{k^2 + (k^2 + d)^2}$, respectively. Here, $k$ is an integer, $e$ is the electronic charge, $\varepsilon_0$ is the vacuum permittivity, $\varepsilon = 4.5$ is the effective dielectric constant of hBN, $d = 56$ nm is the distance between the top and bottom graphite layers and $t = 2$ nm accounts for the separation between the two WS$\text{}_2$ layers in $V(r)$ (valid in the limit $d \gg t$). The formulas for $V(r)$ and $V'(r)$ take into account the screening effect from the graphite gates. Using $a_0 = 8$ nm, we can estimate the nearest-neighbour intralayer and interlayer Coulomb repulsions as $V_1 \approx 32.2$ meV and $V'_1 \approx 31.0$ meV, respectively. $V_1$ and $V'_1$ are similar in strength because $t = 2$ nm is small compared to $a_0 = 8$ nm. The nearest-neighbour exciton–exciton interaction is approximately $V_1 - V'_1 \approx 1.2$ meV.

**Effects of disorder in two Coulomb-coupled moiré lattices**

Compared to single moiré lattices, the coupled moiré lattice system introduces a new type of disorder involving the random variation in the relative displacement between the two lattices over a length scale that is long compared to $a_0$. The observed correlated states require a uniform relative displacement between the two moiré lattices over a sizable fraction of the probed area (about 1 μm in diameter). The current fabrication method does not have control over the moiré lattice alignment. Random displacement between the two moiré lattices can also arise from defects, unintentional strain and so on in each moiré layer. These effects are expected to limit our experiment. Indeed, results from different regions of the same device (Extended Data Fig. 5) and from different devices (Extended Data Fig. 6) show substantial variations compared to the single moiré samples. Whereas the excitonic insulating state at total filling $\nu = 1$ is observed in all areas and all devices, the insulating states at fractional fillings are observed only in about 20% of the sample areas (but in multiple devices). Future efforts are required to improve the uniformity of the coupled moiré lattices. Scanning tunnelling microscopy studies are also required to better understand the correlation between the inter-moiré disorders and the stability of the exciton density waves.

**Particle–hole transformation picture**

The exciton density waves at total fractional fillings can also be viewed from a particle–hole transformation perspective by ignoring the role of spins. Unlike the case of $\nu = 1$, particle–hole transformation in the bottom lattice cannot remove all of the fermionic degrees of freedom at total fractional fillings (Extended Data Fig. 7b,c); there are excess holes (open blue circles) after the formation of interlayer excitons, resulting in a Bose–Fermi mixture of excitons and holes. In this picture, the exciton density waves are defined by the repulsive interactions between the constituents including, in descending order of the energy scale, $V$ between the holes, $V'$ between the excitons and holes and $V - \nu$ between the excitons (as in the case for $\nu = 1$). In the strong correlation limit, where the interactions far exceed the hopping amplitude, the holes spontaneously form generalized Wigner crystals. The exciton–hole repulsion then guides the excitons to the channels defined by the hole Wigner crystals. A fluid of excitons that spontaneously break the translational symmetry of the lattice is formed.

**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.

**References**

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**Author contributions**

Y.Z., Z.X. and R.D. fabricated the devices. Y.Z. and Z.X. performed the measurements and analysed the data. K.W. and T.T. grew the bulk hBN crystals. Y.Z., Z.X., K.F.M. and J.S. designed the scientific objectives, measurements and analysed the data. K.W. and T.T. grew the bulk hBN crystals. Y.Z., Z.X., K.F.M. and J.S. designed the scientific objectives, measured the samples, performed the measurements and commented on the manuscript. All 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 | Reflectance contrast (RC) spectra. a, b, Representative RC spectra of the sensor 2s exciton (a) and the fundamental moiré exciton in WS₂ (b) of device S1 with lattice filling (ν_t, ν_b). The spectral line shape is given by the optical interference effect in the multiple layer structure on the Si/SiO₂ substrate.

We use the peak-to-peak amplitude of the RC between 1.836 and 1.853 eV ($R_{2s}$) as an indicator of the 2s exciton spectral weight (a), and between 1.95 and 2.05 eV ($R_{MX}$) for the moiré exciton (b).
Extended Data Fig. 2 | Optical reflectance contrast spectrum of device S1. a–c, Gate voltage dependence of the reflectance contrast spectrum covering the 1s and 2s resonances of the sensor and the WS₂ moiré excitons. Electrons are located in the bottom lattice solely (a), the top lattice solely (b) and equally in two lattices (c). The total filling factors \( \nu = 0, 1 \) and 2 are determined according to the sensor 2s exciton resonance.
Extended Data Fig. 3 | Magnetic-field dependence. Filling dependence of $R_{2S}$ at representative perpendicular magnetic fields. The curves are displaced vertically for clarity. The perpendicular electric field is 2 mV/nm. The black dashed lines mark total filling factor $\nu = 0, 2/3, 1$ and 2, at which insulating states are observed in the absence of the magnetic field. These states do not disperse with magnetic field and are therefore non-topological. The orange line at each magnetic field denotes the peak positions if the state were topological with Chern number 1.
Extended Data Fig. 4 | Determination of band alignment in the heterostructure. a, Left: Simplified band diagram of natural bilayer WSe₂. While the valence bands are from the K/K' valley, the conduction band minimum is from the Q valley. The indirect optical gap is 1.565 eV as determined by the emission peak in the PL spectrum (right). Right: PL spectrum of the heterostructure showing the interlayer exciton emission at 1.43 eV and the bilayer WSe₂ indirect optical gap at 1.56 eV.

b, Left: Type-II band alignment in the dual-moiré heterostructure. The dashed and solid lines represent spin-down and spin-up bands from the K valley of the monolayer TMDs. The energy scales are determined by PL measurements. Right: PL spectrum of the heterostructure showing the interlayer exciton emission at 1.43 eV and the bilayer WSe₂ indirect optical gap at 1.56 eV.
Extended Data Fig. 5 | Additional sample regions of device S1. a,b. Dependence of $R_\parallel$ on the total filling factor and electric field at two additional regions of the same device as in the main figures. Electrons are in both moiré lattices in the region enclosed by the dashed lines. The insulating state at $\nu = 1$ is robust in all regions. Fewer and less robust insulating states are observed here particularly in a. The features appear curved at certain electric fields because the large contact resistance causes nonlinear gating effects. The electric field offset at $\nu_t = \nu_b$ is likely caused by the layer asymmetry in these regions.
Extended Data Fig. 6 | Additional devices. a, b, Dependence of $R_2$ on the total filling factor and electric field for device S2 (a) and S3 (b). Electrons are in both moiré lattices in the region enclosed by the dashed lines. The insulating state at $\nu = 1$ is robust in both devices. The insulating states at total fractional fillings are observed only in device S3, including an insulating state at $\nu = \frac{1}{3}$. The features appear curved at certain electric fields because the large contact resistance causes nonlinear gating effects.
Extended Data Fig. 7 | Exciton density waves in a particle–hole transformation picture. a–c, Schematic representation of the correlated insulating states in coupled moiré lattices at total filling $\nu = 1$ (a), $\nu = \frac{1}{3}$ (b) and $\nu = \frac{2}{3}$ (c). Top, electrons in the top lattice (red) and bottom lattice (blue). Electrons in the top lattice are bound to the empty sites in the bottom lattice directly below them to minimize the Coulomb interactions. Middle, particle–hole transformation performed on the bottom lattice generates interlayer excitons (red-blue circles) and excess holes (empty blue circles). Bottom, the excess holes form generalized Wigner crystals (or charge-ordered states) because of the hole–hole repulsion $V$. The excitons are guided to the channels defined by the hole Wigner crystals by the exciton–hole repulsion $V'$. The exciton density distribution shows an exciton fluid at $\nu = 1$ and exciton density waves at $\nu = \frac{1}{3}, \frac{2}{3}$. The latter breaks the translational symmetry of the lattice.