A tunable bilayer Hubbard model in twisted WSe$_2$

Yang Xu$^{1,2}$, Kaifei Kang$^1$, Kenji Watanabe$^3$, Takashi Taniguchi$^4$, Kin Fai Mak$^{1,5,6}$ and Jie Shan$^{1,5,6}$

Moiré materials with flat electronic bands provide a highly controllable quantum system for studies of strong-correlation physics and topology. In particular, angle-aligned heterobilayers of semiconducting transition metal dichalcogenides with large band offset realize the single-band Hubbard model. Introduction of a new layer degree of freedom is expected to foster richer interactions, enabling Hund’s physics, interlayer exciton condensation and new superconducting pairing mechanisms to name a few. Here we report competing electronic states in twisted AB-homobilayer WSe$_2$, which realizes a bilayer Hubbard model in the weak interlayer hopping limit for holes. By layer-polarizing holes via a perpendicular electric field, we observe a crossover from an excitonic insulator to a charge-transfer insulator at a hole density of $\nu = 1$ (in units of moiré density), a transition from a paramagnetic to an antiferromagnetic charge-transfer insulator at $\nu = 2$ and evidence for a layer-selective Mott insulator at $1 < \nu < 2$. The unique coupling of charge and spin to external electric and magnetic fields also manifests a giant magnetoelectric response. Our results establish a new solid-state simulator for the bilayer Hubbard model Hamiltonian.

The interlayer coupling in transition metal dichalcogenide (TMD) homobilayers is strongly dependent on the twist angle between the layers$^{1-4}$. In the vicinity of the valence band edge located at the K and $\Gamma$ points of the Brillouin zone, the bands of each monolayer are spin-polarized with large spin splitting, and the spin and valley are locked. For near-0°-twisted (or AA-stacked) homobilayers the layers are strongly hybridized$^{5-8}$. Experimental studies demonstrate a correlated insulating state at a doping density of one hole per moiré unit cell$^{9-11}$. It is argued that the single-band Hubbard model well approximates the system. On the other hand, for a natural homobilayer that is 60° twisted (or AB-stacked) the single-particle interlayer tunnelling is spin forbidden$^{12,13}$. In this limit, the layer degree of freedom can be considered as layer pseudo-spin; together with spin it forms SU(4)-spin (Fig. 1c,d). Recent theoretical studies propose emulating SU(4) Hubbard models in twisted TMD AB-homobilayers$^{14,15}$, which can potentially host chiral spin liquid and exciton supersolid phases. However, experimental realization of the model and characterization of the charge, layer and spin correlations remain elusive.

Here we show that twisted WSe$_2$ AB-homobilayers realize SU(4) Hubbard models (or bilayer Hubbard models in the weak interlayer hopping limit). We investigate the phase diagram by characterizing the charge order, layer polarization and magnetization of the moiré bilayer using the exciton sensing and magneto circular dichroism (MCD) measurements. The twist angle is kept within 2°–3° of AB-stacking to maintain flat bands for strong correlation effects. We demonstrate results from a 58° twisted bilayer. This bilayer forms a triangular moiré lattice with a period of about 10 nm and a moiré density of $n_M \approx 1.1 \times 10^{11}$ cm$^{-2}$. Scanning tunnelling microscopy and spectroscopy reveal that the first moiré flat band for holes is localized on the MX site (M = W and X = Se) in the moiré unit cell (Fig. 1a). The sample is encapsulated in two nearly symmetric gates that are made of hexagonal boron nitride (hBN) gate dielectrics and graphite gate electrodes; it is grounded through a graphite contact (Fig. 1b). The two gates allow independent tuning of the doping density ($\nu$) and perpendicular electric field ($E$) in the sample.

A local dielectric sensor is integrated into the right half of the device. It consists of a WSe$_2$ monolayer that is electronically decoupled from the sample by an ~1-nm-thick hBN spacer. The sensor’s excited exciton resonances are highly sensitive to the screening effect$^{14-16}$, either from the emergence of correlated insulating states in the nearby sample or a change in the sample–sensor separation. The latter provides access to the sample layer polarization ($P = \frac{\nu_i - \nu_b}{\nu_i + \nu_b}$) with $\nu_i$ and $\nu_b$ denoting the charge density on the top and bottom layers of the moiré bilayer, respectively. We also probe the sample magnetization ($M$) and magnetic susceptibility ($\chi$) by MCD spectroscopy of the moiré exciton under an out-of-plane magnetic field $B$. Details on the device fabrication and optical measurements are provided in Methods. All results are obtained at 1.7 K unless otherwise specified.

Insulating states

Figure 1e shows the reflectance contrast spectrum of the right half of the device as a function of gate voltage (bottom axis) and doping density (top axis) in the moiré bilayer. The sensor layer is kept charge neutral. The resonance feature near 1.725 eV that saturates the colour scale and the feature near 1.85 eV are, respectively, the 1s and 2s excitons of the WSe$_2$ monolayer sensor. The feature that is red-shifted from the sensor 1s exciton by 30–50 meV is the moiré exciton, whose energy is lowered by the interlayer coupling in the WSe$_2$ moiré bilayer. The reflectance contrast spectrum of the left half of the device is similar, minus the sensor features. A series of insulating states are revealed by the sensor 2s exciton as abrupt energy blueshift, correlated with reflectance contrast enhancement$^{14}$. The strongest insulating state is observed for the charge-neutral sample ($\nu = 0$). Upon electron doping, additional insulating states emerge at $\nu = 1$, 2, 3, 4, and several fractional filling factors. Upon hole doping, only the $\nu = 1$ and 2 insulating states

$^1$School of Applied and Engineering Physics, Cornell University, Ithaca, NY, USA. $^2$Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing, China. $^3$Research Center for Functional Materials, National Institute for Materials Science, Tsukuba, Japan. $^4$International Center for Materials Nanoarchitectonics, National Institute for Materials Science, Tsukuba, Japan. $^5$Department of Physics, Cornell University, Ithaca, NY, USA. $^6$Kavli Institute at Cornell for Nanoscale Science, Ithaca, NY, USA. $^7$e-mail: yang.xu@iphy.ac.cn; kinfai.mak@cornell.edu; jie.shan@cornell.edu

934
are clearly visible. This suggests flatter moiré conduction bands because many of these states arise from the correlation effect. In addition, the insulating states for the electrons are not sensitive to the out-of-plane electric field $E$, indicating strong interlayer hybridization of the states (which are from the $Q$ points of the Brillouin zone). In contrast, the hole states strongly depend on $E$ (see below), indicating weak interlayer hybridization. We will focus on the case of hole doping to investigate the correlation effects in the weak interlayer coupling limit.

**Electric-field control of layer polarization**

We examine the electric-field effect on layer polarization at a fixed hole density in the right region of the moiré bilayer. Figure 2a illustrates the sensor $2s$ spectrum at $\nu=1$. The $2s$ exciton is discernible for $E < 20 \text{ mV nm}^{-1}$, above which the sensor is doped as a result of charge transfer from the moiré bilayer and the $2s$ exciton is quenched (Extended Data Figs. 1 and 2). As the field sweeps from 20 to $-30 \text{ mV nm}^{-1}$, the $2s$ exciton exhibits a blueshift over a small field range around 0 mV nm$^{-1}$; the response is flat outside this range. The blueshift signals reduced screening of the sensor exciton when charge transfers from the bottom to the top layer in the moiré bilayer (that is, further away from the sensor). Above the critical fields (white dashed lines), the charges are fully layer-polarized ($P_\pm = \pm 1$); below, they are shared between the layers. We summarize the doping dependence of the critical field, $E_c$, in Fig. 2c. For large doping densities, only the crossover to the $P = \pm 1$ state is accessible because doping into the sensor limits its operating electric-field range.

A complementary moiré exciton probe supports the interpretation. Figure 2b shows the field-dependent moiré exciton resonance measured at $\nu = 2$ from the left region of the device (see Extended Data Fig. 3 for other densities; the twist angle in the two regions differs by $-0.1^\circ$). The spectrum shows two distinct features (indicated by the black dashed lines) under small $E$'s and a single prominent feature above $|E|$ (white dashed lines). A similar effect is observed in MoSe$_2$ bilayers separated by a thin hBN layer. The single prominent feature is interpreted as the strong response of the charge-neutral layer after the bilayer is fully layer-polarized. The two spectral features under small $E$s presumably arise from layer asymmetry and/or a small interlayer mixing when both layers are doped.

**Fig. 1** Twisted WSe$_2$ AB-homobilayer. **a**, Twisted WSe$_2$ bilayer exhibiting three types of high-symmetry stacking sites MX, MM and XX (M=W and X=Se) for twist angle $\delta \approx 60^\circ$. **b**, Schematic of a dual-gated WSe$_2$ moiré bilayer device. The right half contains an exciton (bound electron (e)-hole (h) pair) sensor that is made of WSe$_2$ monolayer and separated from the sample by a thin hBN spacer. **c**, Hole-spin alignment at the $-K$ and $K$ valleys in the top (t) and bottom (b) layers of WSe$_2$ AB-homobilayers. The dashed line indicates the Fermi level. Spin and valley are locked in each layer. Within each valley spins are anti-aligned in two layers. Interlayer tunnelling is spin-forbidden. **d**, Illustration of the bilayer Hubbard model with intralayer hopping much smaller than intra- and interlayer on-site repulsions. The spin and layer degrees of freedom are addressable in each layer. **e**, Hole–spin alignment at the $-K$ and $K$ valleys in the top (t) and bottom (b) layers of WSe$_2$ AB-homobilayers. The dashed line indicates the Fermi level. Spin and valley are locked in each layer. Within each valley spins are anti-aligned in two layers. Interlayer tunnelling is spin-forbidden. **d**, Illustration of the bilayer Hubbard model with intralayer hopping much smaller than intra- and interlayer on-site repulsions. The spin and layer degrees of freedom are addressable by an out-of-plane magnetic (B) and electric field (E), respectively. The arrows denote the positive field directions. **e**, Reflectance contrast ($\Delta R/R_0$) spectrum of the device with the sensor versus top-gate voltage. The back gate is fixed at 0 V. The corresponding doping density is shown on the top axis. The $2s$ and $1s$ exciton resonances of the sensor and the moiré exciton resonances of the twisted bilayer are shown in descending order of energy. A series of insulating states, manifested as blueshift of the sensor $2s$ resonance at integer and some fractional filling factors, are observed for both electron and hole doping.
We map the layer polarization as a function of \((\nu, E)\) in Fig. 2d by tracing the reflectance contrast averaged around the single prominent spectral feature \((\approx 1.7 \text{ eV})\). Regions with low (high) reflectance contrast correspond to partial (full) layer polarization. The exact value for \(|P| < 1\) cannot be calibrated from this measurement. The result is fully consistent with Fig. 2c. The threshold field increases linearly with \(\nu\) in each of the three doping regions, \(\nu < 1\), \(1 < \nu < 2\) and \(\nu > 2\); and the slope increases from the low- to high-density regions. There is also a notable jump in \(E_c\) at \(\nu = 1\) and 2, reflecting the charge gap of the states. Intriguingly, the observed \(E_c\) for \(\nu < 1\) is more than an order of magnitude smaller than the estimate from electrostatics using the parallel-plate capacitor model (blue line in Fig. 2c; Methods). This reflects strong correlation effects that favour layer-polarized states. However, ferroelectric instability (including spontaneous layer polarization and electric-field hysteresis) is not observed. At high doping densities \((\nu > 2)\), the slope for \(E_c\) versus \(\nu\) approaches the value of the electrostatics model, reflecting much weaker correlation effects.

**Magnetic properties**

To gain more insight into the nature of the electronic states, we probe their magnetic properties as a function of \((\nu, E)\) by MCD.

The MCD spectrum of the left region of the device is averaged over a narrow spectral window (typically 5 meV) centred on the moiré exciton (Extended Data Fig. 4); the value is linearly proportional to magnetization\(^{18,19}\). Figure 3a–c shows the magnetic-field dependence of MCD at three representative doping densities, \(\nu = 1, 1.6\) and 2. The top and bottom panels correspond to the case of \(P = 0\) and 1, respectively. For layer-unpolarized holes at \(\nu = 1\), we observe a paramagnetic (PM) response at all temperatures, that is, the MCD increases from zero as \(B\) increases, but the rate decreases monotonically. We extract the magnetic susceptibility, \(\chi\), from the MCD slope near zero magnetic field (Fig. 3d). This follows the Curie–Weiss law, \(\chi^{-1} \propto T - \theta\), with Weiss temperature \(\theta \approx 1 \text{ K}\) (blue line). This signals the emergence of magnetic local moments. The negative Weiss temperature reflects antiferromagnetic (AF) exchange interaction between the local moments. In addition, the MCD fully saturates at \(\approx 2 \text{ T}\) at low temperature. Similar behaviour is observed for full layer polarization with slightly weaker AF exchange interaction (Fig. 3d, black symbols).

For layer-unpolarized holes at \(\nu = 2\) (Fig. 3c), the magnetic response is similar to that at \(\nu = 1\). The Weiss temperature is about \(-2 \text{ K}\); the MCD fully saturates at \(\approx 2 \text{ T}\) at low temperature. In contrast, for layer-polarized holes the magnetic response is...
PM only at temperatures above ~10 K. Below 10 K, the response is metamagnetic, that is, the MCD increases slowly with $B$ for small fields, followed by a faster increase before reaching saturation at ~6 T. The high-temperature magnetic susceptibility follows the Curie–Weiss law with $\theta \approx -9$ K. Furthermore, $\chi$ shows a broad peak near $|\theta|$. These behaviours support AF ordering below 9 K (Fig. 3f).

For $1 < \nu < 2$ (Fig. 3b,c), the magnetic response is again PM for layer-unpolarized holes. It develops two components for layer-polarized holes: PM at small magnetic fields and metamagnetic at larger magnetic fields. Extended Data Fig. 5 illustrates the evolution as a function of electric field; the metamagnetic component appears only above $E_c$ (that is, on reaching full layer polarization). In addition, as a function of doping density the PM component continuously shrinks to zero when $\nu$ approaches 2 (Extended Data Fig. 6). The saturation magnetic field for the metamagnetic component increases slowly with density.

**Nature of the correlated electronic states**

Our experiment shows that the $\nu = 1$ and 2 states remain insulating for both layer-polarized and layer-unpolarized charges (Extended Data Fig. 7) under the largest magnetic field accessible in this experiment ($9$ T) and up to about $80$ K (Extended Data Fig. 8). Below we discuss the nature of the ground states and illustrate the most plausible charge/spin configuration in the insets of Fig. 3a–c.

The measurement shows that the $\nu = 1$ state with $|P| = 1$ is a PM insulator down to 1.7 K. In the limit of weak interlayer coupling, the situation is equivalent to half-filling in a TMD moiré heterobilayer with large band offset$^{16,23-24}$. The state is a Mott insulator or a charge-transfer insulator in the presence of more than one orbital (or stacking site with moiré potential minimum$^{22}$) as in this case. The holes are localized by the strong on-site Coulombic repulsion on the MX site of the moiré lattice in one of the WSe$_2$ layers (Fig. 3a). In the flat band limit, the local moments interact via the AF superexchange mechanism$^{17}$ ($\theta < 0$), which is weak for distant
moments (small \( |\theta| \)). The state thus remains PM down to 1.7 K; and a relatively small magnetic field (\( -2 \) T) is sufficient to align these ‘nearly isolated’ spins at low temperature. With \( |P| < 1 \), the holes are distributed in both layers. The holes in one layer are bound to the empty moiré sites in the other layer to minimize the (interlayer) on-site Coulombic repulsion, forming excitons. Such excitonic insulating state has been theoretically proposed for twisted TMD AB-bilayers and is observed in a related system of Coulomb coupled TMD monolayer–moiré bilayer heterostructures. Its charge gap is smaller than that of the Mott insulator, as evidenced by a weaker \( 2s \) contrast for \( |E| < E_c \) in Fig. 2a. Future capacitance studies should yield quantitative gap size measurements.

Our MCD results show that the \( \nu=2 \) state with \( |P|=1 \) is an AF-ordered insulator below 9 K. The state is not compatible with a simple band insulator, in which two holes with antiparallel spins occupy the same orbital. In such an insulator, the spin gap would, following the Pauli exclusion principle, equal the charge gap. Contrarily, here the charge gap, estimated from twice the gap-closure temperature of 80 K, is substantially larger than the spin gap, estimated from the Weiss temperature of 9 K. The conclusion is further supported by the robust insulating state observed beyond magnetic saturation (Extended Data Fig. 8). The most natural scenario is therefore a charge-transfer insulator with a Néel-type AF order. Two holes with antiparallel spins occupy two different stacking sites (probably MX and MM) in each moiré unit cell that form a honeycomb lattice (Fig. 3c). The closer proximity of local moments in this configuration explains the enhanced \( |\theta| \), and the requirement for a much higher magnetic field (\( -6 \) T) to overcome the exchange interaction to fully polarize the spins. The removal of geometric frustration associated with triangular lattices also helps to stabilize AF ordering.

On the other hand, the insulating state at \( \nu=2 \) with \( P \approx 0 \) is practically two copies of the \( \nu=1 \) state with \( |P|=1 \). It is a charge-transfer insulator; the holes in two different layers are probably localized on two different stacking sites of the triangular lattice to minimize the (interlayer) on-site Coulombic repulsion. The transition from the \( |P|=0 \) to the \( |P|=1 \) state is rather abrupt near \( E_c \) (see below).

Whenever the density in a monolayer exceeds 1 for \( 1 < \nu < 2 \), the excess spins prefer to anti-align with their neighbours because of the enhanced AF exchange interaction; they form ‘AF clusters’ or Zhang-Rice singlets in a honeycomb lattice (Fig. 3b). The presence of both nearly isolated moments and AF clusters or spin singlets gives rise to the observed two-component magnetic responses for \( 1 < \nu < 2 \). The composition of the PM and metamagnetic responses is electric-field tunable. This picture is clearly illustrated in the \( (\nu, E) \) dependence of the MCD at \( B = 2 \) T (Fig. 4a). At \( 2 \) T, the nearly isolated moments are fully saturated, but the AF clusters or spin singlets remain largely unperturbed and their contribution to the MCD is small. The MCD thus mostly reflects the density of isolated moments. Figure 4b presents two extreme cases with \( |P|=0 \) and 1. For full layer polarization (black symbols), the density of isolated moments increases linearly with \( \nu \) from zero until reaching \( \nu = 1 \); it then decreases linearly for \( \nu > 1 \) because the nearly isolated moments are continuously replaced by the AF clusters or spin singlets; the nearly isolated moments are completely exhausted at \( \nu = 2 \). On the other hand, for \( P = 0 \) (blue symbols) the same dependence appears at twice the density because now both layers are involved.

The unprecedented gate control of the competing electronic phases in twisted TMD AB-bilayers opens up a new way to realize strong correlation effects and applications. In particular, because of the robust Mott gap and the flat Hubbard bands in each strongly correlated monolayer, a layer-selective Mott insulator, in which one layer is a Mott (or charge-transfer) insulator and the other layer contains itinerant electrons, can be achieved by adjusting the electric field for \( \nu > 1 \). The sudden charge transfer near \( E_c \) (Extended Data Figs. 2 and 3) supports the emergence of a layer-selective Mott insulator for \( 1 < \nu < 2 \). This configuration with metallic states interacting with a lattice of local moments can potentially be used to study the Kondo lattice model. Furthermore, an electric field can switch the system from \( \nu=2 \) to a layer-unpolarized PM insulator to a layer-polarized AF insulator, which results in a drastic change in magnetization under a finite magnetic field (Fig. 4 and Methods). This effectively is a giant magnetoelastic (ME) effect (that is, control of the magnetization by an electric field and vice versa) and is of a purely electronic origin. The electronic ME effect is sought after for energy-efficient, high-speed sensing and information technology, but has remained elusive in conventional solid-state materials. With a variety of competing electronic orders driven by strong correlation, and with charge and spin weakly coupled to the lattice, moiré materials will probably shed new light on electronic magnetoelectrics.

**Conclusions**

We have revealed an abundance of correlated electronic phenomena and competing states in AB-twisted WSe$_2$. We expect similar physics and/or even richer phenomenology in other twisted
TMD AB-homobilayers (for example, twisted MoSe₂ or MoTe₂), angle-aligned heterobilayers with a small band offset\(^{12,13}\) (for example, MoSe₂/WS₂) and more complex multilayer TMD heterostructures\(^{14,15}\). The platform also provides an opportunity to realize other exotic states of matter, such as quantum spin liquids and excitonic supersolids, as envisioned by recent theoretical studies\(^{16,17}\).

Online content
Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41565-022-01180-7.

Received: 15 February 2022; Accepted: 22 June 2022;
Published online: 1 August 2022

References
1. Naik, M. H. & Jain, M. Ultraflatbands and shear solitons in moiré patterns of twisted bilayer transition metal dichalcogenides. *Phys. Rev. Lett.* **121**, 266401 (2018).
2. Wu, F., Lovorn, T., Tutuc, E., Martin, I. & MacDonald, A. H. Topological insulators in twisted transition metal dichalcogenide homobilayers. *Phys. Rev. Lett.* **122**, 086402 (2019).
3. Zhang, Z. et al. Flat bands in twisted bilayer transition metal dichalcogenides. *Nat. Phys.* **16**, 1093–1096 (2020).
4. Pan, H., Wu, F. & Das Sarma, S. Band topology, Hubbard model, Heisenberg model, and Dzyaloshinskii–Moriya interaction in twisted bilayer WSe₂. *Phys. Rev. Res.* **2**, 033087 (2020).
5. Zhai, D. & Yao, W. Theory of tunable flux lattices in the homobilayer moiré of twisted and uniformly strained transition metal dichalcogenides. *Phys. Rev. Mater.* **4**, 094002 (2020).
6. Angeli, M. & MacDonald, A. H. Γ valley transition metal dichalcogenide moiré bands. *Proc. Natl. Acad. Sci. USA* **118**, e2021826118 (2021).
7. Devakul, T., Crépel, V., Zhang, Y. & Fu, L. Magic in twisted transition metal dichalcogenide bilayers. *Nat. Commun.* **12**, 6730 (2020).
8. Xiao, L. et al. Realization of nearly dispersionless bands with strong orbital anisotropy from destructive interference in twisted bilayer MoS₂. *Nat. Commun.* **12**, 5644 (2021).
9. Wang, L. et al. Correlated electronic phases in twisted bilayer transition metal dichalcogenides. *Nat. Mater.* **19**, 861–866 (2020).
10. Shimazaki, Y. et al. Strongly correlated electrons and hybrid excitons in a moiré heterostructure. *Nature* **580**, 472–477 (2020).
11. Ghiotto, A. et al. Quantum criticality in twisted transition metal dichalcogenides. *Nature* **597**, 345–349 (2021).
12. Zhang, Y.-H., Sheng, D. N. & Vishwanath, A. SU(4) chiral spin liquid, exciton supersolid, and electric detection in moiré bilayers. *Phys. Rev. Lett.* **127**, 247701 (2021).
13. Kennes, D. M. et al. Moiré heterostructures as a condensed-matter quantum simulator. *Nat. Phys.* **17**, 155–163 (2021).
14. Xu, Y. et al. Correlated insulating states at fractional fillings of moiré superlattices. *Nature* **587**, 214–218 (2020).
15. Raja, A. et al. Coulomb engineering of the bandgap and excitons in two-dimensional materials. *Nat. Commun.* **8**, 15251 (2017).
16. Xian, L. et al. Creation of moiré bands in a monolayer semiconductor by spatially periodic dielectric screening. *Nat. Mater.* **20**, 645–649 (2021).
17. Liu, G.-B., Xiao, D., Yao, Y., Xu, X. & Yao, W. Electronic structures and theoretical modelling of two-dimensional group-VIB transition metal dichalcogenides. *Chem. Soc. Rev.* **44**, 2643–2665 (2015).
18. Li, T. et al. Continuous Mott transition in semiconductor moiré superlattices. *Nature* **597**, 350–354 (2021).
19. Tang, Y. et al. Simulation of Hubbard model physics in WSe₂/WS₂ moiré superlattices. *Nature* **579**, 353–358 (2020).
20. Wu, F., Lovorn, T., Tutuc, E. & MacDonald, A. H. Hubbard model physics in transition metal dichalcogenide moiré bands. *Phys. Rev. Lett.* **121**, 026402 (2018).
21. Regan, E. C. et al. Mott and generalized Wigner crystal states in WSe₂/WS₂ moiré superlattices. *Nature* **579**, 359–363 (2020).
22. Zhang, Y.-H. & Vishwanath, A. Electrical detection of spin liquids in double monolayer WSe₂ and moiré WS₂/WS₂. Preprint at https://arxiv.org/abs/2108.07131 (2021).
23. Li, T. et al. Charge-order-enhanced capacitance in semiconductor moiré superlattices. *Nat. Nanotechnol.* **16**, 1068–1072 (2021).
24. Dalal, A. & Ruhman, J. Orbitally selective Mott phase in electron-doped twisted transition metal-dichalcogenides: a possible realization of the Kondo lattice model. *Phys. Rev. Res.* **3**, 043173 (2021).
25. Ajesh K., Hu, N. C., MacDonald, A. H. & Potter, A. C. Gate-tunable heavy fermion quantum criticality in a moiré Kondo lattice. Preprint at https://arxiv.org/abs/2110.11962 (2021).
26. Sivadas, N., Okamoto, S. & Xiao, D. Gate-controllable magneto-optic Kerr effect in layered colinear antiferromagnets. *Phys. Rev. Lett.* **117**, 267203 (2016).
27. Feibig, M. Revival of the magnetoellectric effect. *J. Phys. D* **38**, R123–R152 (2005).
28. Tang, Y. et al. Tuning layer-hybridized moiré excitations by the quantum-confined Stark effect. *Nat. Nanotechnol.* **16**, 52–57 (2021).
29. Tang, Y. et al. Dielectric catastrophe at the Mott and Wigner transitions in a moiré superlattice. Preprint at https://arxiv.org/abs/2201.12510 (2022).
30. Xian, L. et al. Engineering three-dimensional moiré flat bands. *Nano Lett.* **21**, 7519–7526 (2021).
31. Zhang, Y.-H. & Vishwanath, A. Electrical detection of spin liquids in double moiré layers. Preprint at https://arxiv.org/abs/2005.12925 (2020).

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

© The Author(s), under exclusive licence to Springer Nature Limited 2022
Methods

Device fabrication. Flakes of few-layer graphite, hBN and monolayer WSe₂ are mechanically exfoliated from bulk crystals on silicon substrates with a 285 nm SiO₂ layer. They are selected by colour contrast under an optical microscope. The top and back hBN gate dielectrics are chosen to have nearly identical thickness (~40 nm). The heterostructure is assembled layer-by-layer using a dry transfer method described elsewhere. The twisted WSe₂ bilayer is fabricated by the ‘‘pear-and-stack’’ method, similar to that developed for twisted bilayer graphene. Namely, part of a large WSe₂ monolayer is picked up and stacked on the remaining part with twist angle θ that is precisely controlled by a rotary stage. The target angle is 58°. The twist angle variation is ±0.1° across the sample, and is calibrated by the density of the insulating states at integer fillings locally (Extended Data Fig. 10).

An additional WSe₂ monolayer is introduced into one part of the device as a sensor. It is separated from the twisted bilayer by a thin hBN layer (1−2 nm).

Optical measurements. The optical measurements are performed in a closed-cycle cryostat (Attocube, Attodyno 2100) at temperatures down to 1.7 K and magnetic fields up to 9 T. The details of the reflectance spectrum measurement are described in ref. The reflectance contrast, k_B/R(ν) = (R−R)/R, is obtained by comparing reflectance k to reference R measured at the same sample location under large negative top- and back-gate voltages (V_top = −8 V). The MCD = |E(ν)−E(ν+δ)|/E(ν) is obtained using the reflectance spectrum excited by left and right circularly polarized light, R_+ and R_−, respectively. An example is shown in Extended Data Fig. 8. The reported MCD in this work is the average MCD over a small spectral range (typically 5 meV) centred near the moiré exciton resonance. To be more efficient, in some B-dependent studies we measure the reflectance spectrum with circularly polarized light of only one handedness (for example, R_+). We infer the spectrum of the other handedness based on R_− = R_+ (−B).

Two regions on the device are studied. One region has the sensor layer; the twist angle is ±0.04° and the layer separation is 58.2°. The sensor 2ν exciton is utilized to detect the correlated insulating states and the layer polarization. The second region lacks the sensor layer; the twist angle is 58.1°. The moiré exciton response and MCD are measured. The out-of-plane electric field is calculated from the gate voltages using E = (C_y V_y − C_x V_x)/2ε0/ε0 − E_v. Here C_y (C_x) is the geometrical capacitance per unit area of the top (back) gate, ε0 = 8.85 × 10⁻¹² F/m is the dielectric constant of hBN, ε_v is the vacuum permittivity, and E_v = −6 mV nm⁻¹ is a small built-in electric field (probably arising from the small layer asymmetry of the structure). The latter is introduced so that the layer polarization is symmetric about 0°. The top- and back-gate capacitances are individually calibrated by analysing the gate dependence of the Landau levels in a WSe₂ monolayer that is attached to the moiré bilayer under a perpendicular magnetic field of 9 T. The Landau level is detected optically by measuring the polaron energy/intensity oscillation. The two gate capacitances differ by less than 10%.

Assignment of the charge neutrality point. In Fig. 1e, we identify the insulating state with the largest charge gap, which corresponds to the highest energy and strongest 2ν exciton resonance, as the υ = 0 state, that is, the intrinsic semiconductor band gap. This assignment is consistent with the temperature-dependence data in Extended Data Fig. 8, in which the υ = 0 state survives to the highest temperature in this study. However, the gate voltage span for the υ = 0 state appears to be smaller than the expected semiconductor gap size (1−1.5 V). This is probably related to the bad electrical contact to the near charge neutrality at low temperatures, which could cause a nonlinear gate voltage dependence of the doping density. This picture is supported by the temperature dependence in Extended Data Fig. 8. Specifically, the gate voltage span near υ = 0 increases to ~1−1.5 V at 80 K, which is close to the expected semiconductor gap size. Better contacts are formed with increasing temperature.

Estimate of E_v using electrostatics. Charge transfer between the two WSe₂ monolayers in the twisted bilayer can be modelled by the parallel-plate capacitor model. The critical electric field E_v required to achieve full layer polarization for charge density n_v is

\[ E_v = \frac{C_y}{C_x} \frac{2}{2} \frac{\nu e V}{\epsilon_0} \]

Here C_y is the quantum capacitance of each WSe₂ monolayer, C_x is the interlayer capacitance of the twisted bilayer, C_x = C_x (ε_x C_y) is the gate capacitance and ε_0 is the elementary charge. Equation (1) is plotted in Fig. 2c (blue line). For non-interacting electrons, we have C_x ≈ C_x C_y and \( E_v \approx \frac{\nu e V}{\epsilon_0} \). At \( \nu = 1 \), the value is ≈2.8 mV nm⁻¹ and about an order of magnitude larger than the observed \( E_v \), which demonstrates the importance of the electron–electron interactions. At \( \nu = 2 \), the discrepancy between experiment \( (E_v \approx 17 mV nm⁻¹) \) and model \( (E_v \approx 56 mV nm⁻¹) \) is reduced; the experimental slope for \( E_v \) versus \( \nu \) also approaches the expected slope at high doping densities. The results suggest that the interaction effect becomes weaker as the Fermi level is moved to higher-lying moiré bands. This is expected as the moiré bandwidth generally increases with the band index.

As discussed in the main text, there is a jump in \( E_v \) at \( \nu = 1 \) and 2. The jump is caused by the presence of the charge gap for these insulating states. Additional electric field is required to overcome the charge gap and fully transfer the charges from one layer to the other. However, the larger jump in \( E_v \) observed at \( \nu = 2 \) than at \( \nu = 1 \) is not well understood. It is correlated with the enhanced layer polarization at low doping densities, at which the interaction effect is most important. Future theoretical studies are required to quantitatively describe the density dependence of the layer polarization.

ME effect. The coupling of the layer and spin degrees of freedom to the electric and magnetic fields, respectively, and the sensitivity of the magnetic ground state to layer polarization in twisted bilayer WSe₂ provide a possibility to achieve a large ME effect of pure electronic origin. We investigate the case \( \nu = 2 \). Extended Data Fig. 9 shows the contour plot of the MCD as a function of \((E_v, B)\). The MCD normalized by its value at 9 T (above magnetic saturation) characterizes the magnetization \( M (\nu = 1) \) for a fully spin polarized state. Four different regions can be identified: (I) \( |M_\parallel| < 1 \), (II) \( |M_\parallel| < 1 \), (III) \( |M_\parallel| = 1 \), (IV) \( |M_\parallel| = 1 \). The boundaries of these regions are determined according to \( M_\parallel dB \) and \( M_\parallel dM \). The magnetization is sensitive to both electric and magnetic fields. Particularly at intermediate magnetic fields, for \( |M_\parallel| = 1 \), the strong intralayer AF exchange favours antiparallel spin alignments which reduce the magnetization substantially compared to the PM state for \( P = 0 \). An electric field can thus shift the system between region I and II and cause over 60% change in the sample magnetization. This is a giant ME effect. At high enough magnetic fields that overcomes the interlayer AB exchange, the electric field shifts the system between region III and IV, and the ME coupling diminishes. The ME effect is also doping dependent. Figure 4a shows magnetization as a function of electric field and doping density at a fixed magnetic field of 2 T. The magnetization changes significantly around the threshold electric field for filling \( \nu = 1 \). The ME effect is the largest around \( \nu = 2 \) and weakens for \( 1 < \nu < 2 \) and \( \nu > 2 \).

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. Source data are provided with this paper.

References

36. Kim, K. et al. van der Waals heterostructures with high accuracy rotational alignment. Nano Lett. 16, 1989–1995 (2016).
37. Cao, Y. et al. Superlattice-induced insulating states and valley-protected orbits in twisted bilayer graphene. Phys. Rev. Lett. 117, 116804 (2016).
38. Chung, T.-E., Xu, Y. & Chen, Y. P. Transport measurements in twisted bilayer graphene: electron–phonon coupling and Landau level crossing. Phys. Rev. B 96, 035425 (2018).

Acknowledgements

We thank L. Fu, Y. Zhang, M. Davydova, A. H. MacDonald, N. Wei and Y. Zeng for fruitful discussions. X.Y. acknowledges support from the National Key R&D Program of China (2021YFA1401300) for data analysis at the Chinese Academy of Sciences. This study was supported by the US Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), under award number DE-SC0022058 (optical characterization) and DE-SC0019481 (magnetic characterization), the National Science Foundation (NSF) under DMR-2114535 (Rydberg sensing measurement) and the US Office of Naval Research under award number N00014-21-1-2471 (device fabrication). The growth of hBN crystals was supported by the Elemental Strategy Initiative of MEXT, Japan, and CREST (JPM/R353). This work was made use of the Cornell NanoScale Facility, an NNCI member supported by NSF grant NNCI-2025233.

Author contributions

X.Y. and K.K. fabricated the devices. X.Y. performed the measurements and analysed the data. K.W. and T.T. grew the bulk hBN crystals. X.Y., K.F.M. and J.S. designed the scientific objectives, oversaw the project and co-wrote the manuscript. All authors discussed the results and commented on the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

This article is available at https://doi.org/10.1038/s41565-022-01180-7.

Supplementary information

The online version contains supplementary material available at https://doi.org/10.1038/s41565-022-01180-7.

Peer review information

Nature Nanotechnology thanks Angel Rubio and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

Reprints and permissions information is available at www.nature.com/reprints.
Extended Data Fig. 1 | Operating gate-voltage range of the sensor. a,b, Gate dependence of the reflectance contrast spectrum of the device with the sensor at $E=6\,\text{mV/nm}$ (a) and $-55\,\text{mV/nm}$ (b). The gate voltage $V_{\text{tot}} (= V_{\text{tg}} + V_{\text{bg}})$ is proportional to the total charge density in the moiré bilayer and the sensor layer. The red and blue dashed lines mark the limits of gate voltage beyond which the sensor becomes doped, as evidenced by the emergence of the polarons ($1.7-1.75\,\text{eV}$) and disappearance of the $2s$ state ($\sim 1.85\,\text{eV}$). c, Operating range of the gate voltage (shaded area). It is primarily determined by the back gate voltage ($-2\,\text{V}, 4\,\text{V}$). The sensor is closer to the back gate. The blue and red symbols are determined from the boundaries in a, b and similar measurements at other electric fields.
Extended Data Fig. 2 | Layer polarization probed by the sensor 2 s exciton. The reflectance contrast spectrum of the sensor 2 s exciton as a function of electric field at representative doping densities. The case of \( \nu = 1 \) is shown in Fig. 2a of the main text. The grey dashed lines denote the critical electric fields \( E_c \).
Extended Data Fig. 3 | Layer polarization probed by the moiré exciton. Reflectance contrast spectrum of the moiré exciton as a function of electric field at representative hole filling factors. The result for $\nu = 2$ is shown in Fig. 2b of the main text. The spectra are measured from the region without the sensor. The white dashed lines mark $E_c$. 

**Image Description**: The image shows a series of graphs labeled with different filling factors ($\nu = 0.3$, $\nu = 1$, $\nu = 1.4$, $\nu = 2$, $\nu = 2.4$). Each graph plots energy (eV) against electric field (mV/nm) with color gradients indicating reflectance contrast ($\Delta R/R_0$). The graphs are sequential and demonstrate the variation in reflectance contrast with changes in filling factor.
Extended Data Fig. 4 | MCD measurement. **a**, Magnetic-field dependence of the MCD spectrum at $\nu = 2$ under zero electric field. **b**, Linecuts of **a** at $B = 5$ T (black line) and -5 T (red line). The MCD is determined as $\frac{R_{+}(B) - R_{+}(-B)}{R_{+}(B) + R_{+}(-B)}$, where $R_{+}(B)$ is the reflectance contrast of left circular polarized light under magnetic field $B$. 
Extended Data Fig. 5 | MCD at \( r = 1.6 \) under different electric fields. Magnetic-field dependence of MCD under electric field ranging from 0 to 128 mV/nm. The MCD is obtained by averaging the MCD spectrum over a spectral window of 5 meV centred at the exciton resonance (≈ 1.69 eV). The response is PM at small electric fields. A metamagnetic component emerges above about 8.5 mV/nm, which agrees well with the measured threshold electric field \( E_c \) for full layer polarization.
Extended Data Fig. 6 | MCD at different doping densities. Magnetic-field dependence of MCD for layer-polarized ($E = -55$ mV/nm) (a) and layer-unpolarized holes ($E = 0$ mV/nm) (b) at representative filling factors. The MCD is obtained by averaging the MCD spectrum over a spectral window of 5 meV centred at the exciton resonance ($\approx 1.69$ eV).
Extended Data Fig. 7 | Electric-field effect on the insulating states. a–f. Gate dependence of the reflectance contrast spectrum of the sensor 2\( s \) exciton at representative electric fields, including 67 mV/nm (a), 43 mV/nm (b), 18 mV/nm (c), 0 mV/nm (d), -18 mV/nm (e), and -43 mV/nm (f). Electrons and holes are introduced into the channel for \( V_{\text{tot}} \) above and below about -1 V, respectively. The electric field limits the operating voltage range of the sensor. In addition, notable 2\( s \) exciton energy shift is observed on the hole side, but not on the electron side, when the electric field switches sign. The result indicates strong layer hybridization for the conduction bands in twisted WSe\(_2\), the band edge of which is located at the Q point of the Brillouin zone.
Extended Data Fig. 8 | Temperature and magnetic-field effects on the insulating states. a-f, Top gate dependence of the reflectance contrast spectrum of the sensor 2 s exciton at 1.7 K and under magnetic field of 2 T (a) and 9 T (b), as well as under zero magnetic field at 10 K (c), 36 K (d), 55 K (e) and 80 K (f). The back gate is fixed at 0 V. The corresponding electric field varies for different doping densities. Particularly, the electric field is -43 mV/nm for \( \nu = 1 \) and -76 mV/nm for \( \nu = 2 \) on the hole side, under both of which the moiré bilayer is fully layer-polarized. The correlated insulating states for holes at \( \nu = 1 \) and 2 are robust against magnetic field for the entire accessible range (9 T) and against thermal melting up to about 80 K.
Extended Data Fig. 9 | Magnetoelectric (ME) effect at $\nu = 2$. a, Normalized MCD (by its value at 9 T) (a) and its derivative with respect to $B$ (c) and to $E$ (d) as a function of the electric and magnetic fields. The four regions (defined by the dashed lines) correspond to: (I) $|M|, |P| < 1; (II) |M| < 1, |P| = 1; (III) |M| = 1, |P| < 1; and (IV) $|M|, |P| = 1$. The boundaries are identified by the derivatives in c and d. The insets in a on the left and right illustrate, respectively, the spin and charge configuration at small magnetic fields and after magnetic saturation. On each side the three insets correspond to $P = -1, 0$ and 1 from top to bottom. b, Vertical linecuts of a at representative magnetic fields. Strong ME effect is observed for the intermediate magnetic fields, under which the electric field changes the magnetization by over 60%. 
Extended Data Fig. 10 | Spatial variation of moiré density. Top-gate voltage ($V_{tg}$) dependence of the reflection contrast spectrum of the sensor 2’s exciton at three sample locations (P1, P2, and P3) that have slightly different moiré densities. Only the position of the $\nu = 0$ state remains unchanged (illustrated by the yellow vertical dashed line) while the positions of the other states (such as the $\nu = 1,2$ states indicated by the black dashed lines) shift from P1 to P3. The same applies to the electron doping case with $V_{tg} > 0$. The results demonstrate the slight twist angle variation in the sample.