Moiré superlattice in a MoSe$_2$/hBN/MoSe$_2$ heterostructure: from coherent coupling of inter- and intra-layer excitons to correlated Mott-like states of electrons

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Two dimensional materials and their heterostructures constitute a promising platform to study correlated electronic states as well as many body physics of excitons. Here, we present experiments that unite these hitherto separate efforts and show how excitons that are dynamically screened by itinerant electrons to form exciton-polarons, can be used as a spectroscopic tool to study interaction-induced incompressible states of electrons. The MoSe$_2$/hBN/MoSe$_2$ heterostructure that we study exhibits a long-period Moiré superlattice as evidenced by coherent-hole tunneling mediated avoided crossings between the intra-layer exciton with three inter-layer exciton resonances separated by $\sim 3$ meV. For electron densities corresponding to half-filling of the lowest Moiré subband, we observe strong layer-paramagnetism demonstrated by an abrupt transfer of all $\sim 1500$ electrons from one MoSe$_2$ layer to the other upon application of a small perpendicular electric field. Remarkably, the electronic state at half-filling of each MoSe$_2$ layer is resilient towards charge redistribution by the applied electric field, demonstrating an incompressible Mott-like state of electrons. Our experiments demonstrate that optical spectroscopy provides a powerful tool for investigating strongly correlated electron physics in the bulk and pave the way for investigating Bose-Fermi mixtures of degenerate electrons and dipolar excitons. Van der Waals heterostructures incorporating transition metal dichalcogenide (TMD) bilayers open up new avenues for exploring strong correlations using transport and optical spectroscopy. In contrast to similar structures in III-V semiconductors, these heterostructures exhibit possibilities for exotic material combinations, creation of Moiré superlattices exhibiting narrow electronic bands$^{1-3}$, and strong binding of spatially separated inter-layer excitons$^{5-9}$. Recently, ground-breaking transport experiments in twisted bilayer graphene demonstrated a fascinating range of strongly correlated electron physics in a single system$^{10-16}$: by varying the filling factor $\nu$ of the lowest energy Moiré from 0 to 1, the ground-state of the interacting electron or hole system could be reversibly changed from a superconductor for a large range of $\nu$ to a Mott insulator at $\nu = n/4$ ($n = 1, 2, 3$) or a band insulator at $\nu = 1$. In fact, this system realizes a two-dimensional (2D) Fermi-Hubbard model on a triangular lattice with a fully-tunable electron density – a paradigmatic example of a strongly correlated electronic system with many open questions.

In parallel, optical spectroscopy in van der Waals heterostructures have revealed the prevalence of many-body hybrid light-matter states, termed exciton-polarons$^{17,18}$, in the excitation spectra of electron or hole doped monolayers. Advances in material quality and device fabrication has lead to the observation of Moiré physics of non-interacting excitons in MoSe$_2$/WSe$_2$$^{19,20}$, MoSe$_2$/WS$_2$$^{21}$, and WS$_2$/WSe$_2$$^{22}$ heterobilayers. Potential of this new system for investigating many-body physics was recently revealed in a remarkable demonstration of a long-lived inter-layer exciton condensate$^{23,24}$. Here, we describe experiments in a heterostructure incorporating a MoSe$_2$/hBN/MoSe$_2$ homobilayer that in several ways combine the principal developments in these two fields to demonstrate interaction-induced incompressible states of electrons. We provide an unequivocal demonstration of hybridization of inter- and intra-layer excitons mediated by coherent hole tunneling$^{4,21,25-27}$ between the two MoSe$_2$ layers: the avoided crossings that we observe in optical reflection not only show the formation of dipolar excitons with a strong optical coupling but also reveal the existence of at least 3 Moiré bands of indirect excitons. We then demonstrate that intra-layer exciton-polaron resonances provide a sensitive tool to investigate correlated electronic states in the bulk. Equipped with this spectroscopic tool, we observe strong layer-paramagnetism$^{28,29}$ and an incompressible Mott-like state of electrons when each layer has half filling.

I. DEVICE STRUCTURE AND BASIC CHARACTERIZATION

We show the schematic of the device structure in Fig. 1a. By using a double gate structure, we can control the electric field and the chemical potential of the device independently. Few-layer graphene serves as transparent gates, top and bottom hBN serve as gate insulators, and middle hBN (monolayer) serves as a tunnel barrier. The crystal axis of the MoSe$_2$ layers are aligned to be close to 0 degree using the tear-and-stack technique$^{30}$. Both MoSe$_2$ layers are grounded via few-layer graphene contacts. Fig. 1d shows the optical microscope image of the device. Fig. 1b is a schematic image of a dipolar exciton formed by coherent coupling of inter-layer exciton (IX) and intra-layer exciton (X) via hole tunneling. Fig. 1c shows a schematic image of the electrons in a Moiré lat-
tice probed by intra-layer exciton in low electron density regime.

Figure 1e shows a spatial map of total photoluminescence (PL) from the device. Here, both top and bottom gate voltages are kept at zero Volts.

We observe PL from regions with monolayer MoSe$_2$, but not from bilayer MoSe$_2$, where two MoSe$_2$ flakes are in direct contact (the area around the point indicated by the white arrow in Fig. 1e). On the other hand, the MoSe$_2$/hBN/MoSe$_2$ area shows bright PL. This indicates that the heterostructure becomes a direct band gap system owing to the reduction of the inter-layer hybridization of the valence bands at the $\Gamma$ point, due to the presence of monolayer hBN. Typical PL spectra of the monolayer MoSe$_2$ and the MoSe$_2$/hBN/MoSe$_2$ area are shown in the inset of Fig. 1e: there are pronounced intra-layer exciton luminescence peaks in both areas. Even though the heterostructure is fabricated from a single MoSe$_2$ layer using tear-and-stack technique, we observe two distinct intra-layer exciton peaks in the MoSe$_2$/hBN/MoSe$_2$ region. We observe that this strain-induced energy difference between the PL from the top and bottom layers varies across the sample (see Supplementary Information S2).

II. COHERENT INTER-LAYER HOLE TUNNELING AND DIPOLAR EXCITONS

We first analyze the electric field ($E_z$) dependence of the elementary optical excitations of the MoSe$_2$/hBN/MoSe$_2$ section in the absence of itinerant electrons or holes. To this end, we scan the top and bottom gate voltages (along L4 indicated in Fig. 3c) together to change $E_z$ while keeping the homo-bilayer system in the charge neutral regime. The PL spectrum we thus obtain is depicted in Fig. 2a: using the top ($V_{tg}$) and bottom ($V_{bg}$) gate voltage dependence, we determine that the PL spectra around 1.632 eV and 1.640 eV stem from intra-layer exciton in top and bottom layer ($X_{\text{top}}$ and $X_{\text{bot}}$), respectively. For high values of $|E_z|$ depicted in the top and bottom parts of the color-coded PL spectrum, we observe PL lines with a strong $E_z$ dependence: we identify these PL lines as originating from inter-layer excitons with a large dipole moment leading to a sizeable Stark shift.

The spectra for positive (negative) $V_{tg}$ regime corresponds to the inter-layer exciton $X_{\uparrow}$ ($X_{\downarrow}$) which has a hole in the bottom (top) layer and an electron in the top (bottom) layer. The associated dipole-moment of the inter-layer exciton changes its polarity for $V_{tg} \sim 0$. By extrapolating the $X_{\uparrow}$ and $X_{\downarrow}$ PL lines and finding their crossing point, we estimate the energy difference between the inter- and the intra-layer exciton resonances at $E_z = 0$, which allows us to determine their binding energy difference to be $\sim 100$ meV.

Figure 2b shows the differential reflectance ($\Delta R/R_0$) spectrum obtained for the same range of gate voltage scan as that of Fig. 2a. Here, $\Delta R/R_0 \equiv (R - R_0)/R_0$, with $R$ and $R_0$ denoting the reflectance signal from the MoSe$_2$/hBN/MoSe$_2$ region, and background reflectance, respectively. In accordance with the PL data (Fig. 2a), we find $X_{\text{top}}$ and $X_{\text{bot}}$ resonances around 1.632 eV and 1.640 eV, respectively. Moreover, for $V_{tg} \gtrsim 7.5$ V ($V_{tg} \lesssim -7.5$ V), we observe $IX_{\uparrow}$ ($IX_{\downarrow}$) hybridizing exclusively with $X_{\text{top}}$ ($X_{\text{bot}}$). Figures 2c and 2d show the magnified plots of the regions highlighted with blue and green dashed lines in Fig. 2b, confirming avoided crossing of an intra-layer exciton line with multiple inter-layer excitons. We first note that the observation of a sizeable reflection signal from $IX_{\uparrow}$ away from the avoided crossing suggests that it is possible to resonantly excite long-lived inter-layer excitons in these structures. The hybridization of $IX_{\uparrow}$ lines with $X_{\text{top}}$ together with the lack of an avoided crossing with $X_{\text{bot}}$ in Fig. 2c, unequivocally shows that avoided crossings originate exclusively from coherent hole tunneling schematically shown in Fig. 2e. Our observation, proving that the hole tunnel coupling is much larger than that of the electron, is consistent with the band alignment expected from first principle band-structure calculations. This conclusion is also confirmed by the data depicted in Fig. 2d, where avoiding crossing originates from coherent-hole-tunneling induced hybridization of $IX_{\downarrow}$ and $X_{\text{bot}}$ schematically shown in Fig. 2f.

One of the most remarkable features of the spectra depicted in Figs. 2c and 2d is the existence of multiple avoided crossings associated with three inter-layer exciton resonances separated in energy by $\sim 3$ meV. This inter-layer exciton fine-structure demonstrates the existence of a Moiré superlattice, originating from a small twist angle between the two MoSe$_2$ layers. The presence of an hBN tunnel barrier strongly suppresses the strength of the associated Moiré potential, rendering it sizeable only for the inter-layer excitons.

III. CHARGE CONFIGURATION DETECTION BY EXCITON-POLARON SPECTROSCOPY

It is well established that presence of itinerant charges drastically alters the optical excitation spectrum. Recent theoretical and experimental work established that the modified spectrum originates from dynamical screening of excitons by electrons or holes, that lead to the formation of a lower energy attractive polaron (AP) branch. Concurrently, the exciton resonance evolves into a repulsive polaron (RP) (see Supplementary Information S1). The particularly strong sensitivity of the RP resonance energy to changes in electron density renders it an ideal spectroscopic tool for monitoring, or sensing, the electron density $n$ in the same layer. The strain induced resonance energy difference between $X_{\text{top}}$ and $X_{\text{bot}}$, ensuring different energies for the corresponding RP$_{\text{top}}$ and RP$_{\text{bot}}$, together with the much weaker sensitivity of RP$_{\text{top}}$ (RP$_{\text{bot}}$) on $n$ in the bottom (top) layer, allows us to determine the charging configuration of the
two layers simultaneously. Since we are predominantly interested in the low carrier density regime where the quasi-particle (bare-exciton) weight of the RP resonance is close to unity, we will refer to it as the exciton resonance.

Figures 3a and 3b show the gate voltage dependence of $\Delta R/R_0$ at $E = 1.632$ eV and $E = 1.640$ eV, which correspond to the top ($X_{\text{top}}$) and bottom ($X_{\text{bot}}$) intra-layer exciton resonance energy in the charge neutral regime, respectively. The inset to these figures show a line cut through the dispersive neutral exciton reflection spectrum, indicating the exciton energies at which we monitor $\Delta R/R_0$. Since a small increase of $n$ from $\sim 0$ to $1 \times 10^{11}$ cm$^{-2}$ results in a change of $\Delta R/R_0$ from $\sim -1$ to $\sim 0$, the blue areas in Figs. 3a and 3b correspond to the charge neutral regime of each layer. The red and white areas in turn, correspond to the electron or hole doped regime of each layer. This all-optical determination of the electrically resolved charge map of the bilayer provides an invaluable tool for monitoring the bulk properties of 2D materials which is not directly accessible in transport measurements.

To enhance the sensitivity of the charge map to the transition between the undoped and doped regimes and to visualize the charge configuration of both layers at the same time, we first evaluate the derivative of $\Delta R/R_0$ with respect to energy at $E = 1.632$ eV and $E = 1.640$ eV, and then overlay $d(\Delta R/R_0)/dE$ obtained for both layers. The resulting charge map, depicted in Fig. 3c, is closely reminiscent of the charging plateaus used to characterize gate-defined quantum dots. Since $d(\Delta R/R_0)/dE$ is only sensitive to changes in $n$, the blue regions in Fig. 3c correspond to the regime where the charge configuration changes, allowing us to clearly separate the regions (t,b) where the top or bottom layer is neutral (t=i or b=i), electron doped (t=n or b=n) or hole doped (t=p or b=p).

We show typical gate voltage dependence of $\Delta R/R_0$ in Fig. 3d and 3e which are obtained when the two gate voltages are scanned in a coordinated manner along the lines L1 and L2, indicated in Fig. 3a and 3b, respectively. In both plots, we confirm the emergence of the AP resonance and the associated blue shift of the exciton / RP resonance energy around the charge configuration transition points, confirming the assignment obtained from $d(\Delta R/R_0)/dE$ in Fig. 3c.

In stark contrast to the case of monolayer MoSe$_2$ (Supplementary Information S1), we find that the top (bottom) gate dependence obtained by fixing the bottom (top) gate voltage, is not monotonic because of the screening of the applied gate voltage when one of the two layers is already doped. The observed responsivity to the applied gate voltages is consistent with the interpretation that the lower (higher) energy exciton resonance at $E = 1.632$ eV ($E = 1.640$ eV) is $X_{\text{top}}$ ($X_{\text{bot}}$). For example in Fig. 2b, by fixing $V_{\text{tg}} = 0$ V and sweeping $V_{\text{bg}}$ from negative to positive, we find drastic change of $\Delta R/R_0$ around $V_{\text{tg}} \approx 2$ V. On the other hand, by fixing $V_{\text{bg}} = 0$ V and sweeping $V_{\text{tg}}$ from negative to positive, we find much less change of $\Delta R/R_0$. From this asymmetry of the gate dependence, we can confirm that the resonance in Fig. 2b is originating from $X_{\text{bot}}$.

Figure 3f shows the gate voltage scan along L3, indicated in Fig. 3c, where we fixed $V_{\text{bg}}$ at 4 V and scanned $V_{\text{tg}}$. By sweeping $V_{\text{tg}}$ from negative to positive, we find that the bottom layer gets electron doped around $V_{\text{tg}} = -3$ V, and then gets depleted by increasing $V_{\text{tg}}$ further. This observation shows that electrons are transferred from the bottom layer to the top layer whilst electrons are introduced into the top layer and $V_{\text{tg}}$ is kept unchanged. This counter-intuitive dependence shows up as the curving of the lines separating the charge configurations (n,i) and (n,n) in Fig. 3c. Similar inter-layer charge transfer behavior was previously observed in transport experiments in bilayer semiconductor systems and was attributed to the negative compressibility. To the best of our knowledge, our experiments provide the first observation of negative compressibility, arising from dominance of intra-layer exchange interactions over kinetic energy, using optical spectroscopy.

IV. Interaction Induced Incompressible States

The results we present in Sec. II establish the existence of a Moiré superlattice for inter-layer excitons. On the one hand, the underlying periodic modulation of the electronic bands should lead to Moiré subbands for electrons (holes) in the conduction (valence) band. On the other hand, the absence of coherent electron tunneling indicates that the resulting subbands in the top and bottom layers do not hybridize. Taking into account the relatively strong conduction-band spin-orbit coupling in MoSe$_2$, the homobilayer system we are investigating realizes a rather unique system exhibiting flat bands with layer and valley-spin degree of freedom; while the degeneracy associated with the former can be tuned using a perpendicular electric field ($E_z$), the latter can be controlled using a magnetic field ($B_z$). Moreover, our observation of negative compressibility (Sec. III) indicates that the electron-electron interaction energy scale dominates over kinetic energy even at relatively high electron densities ($n \approx 1 \times 10^{12}$ cm$^{-2}$) where several Moiré bands in one layer are occupied. In this section, we explore electron correlation effects in the more interesting regime of low carrier densities by zooming in to the low-$n$ section of the charging map (Fig. 3c) where the (i,i), (i,n), (n,i) and (n,n) regions coalesce. The high sensitivity of the exciton/RP resonance energy, as well as the AP oscillator strength, to changes in electron density once again forms the backbone of our investigation.

Figures 4a and 4b show the gate voltage dependence of differential reflectance close to the $X_{\text{top}}$ and $X_{\text{bot}}$ resonance energy for $n = 0$ at $E_{X_{\text{top}}}^0 = 1.6320$ eV and $E_{X_{\text{bot}}}^0 = 1.6402$ eV, respectively. In these maps, a shift of the exciton resonance energy due to a change in $n$
is detected as a modification of the exciton reflectance. The specific choice of $E_{X_{\text{top}}}$ and $E_{X_{\text{bot}}}$, indicated by the magenta and cyan points in the insets of Fig. 4a and 4b, maximizes the sensitivity to $n$. Instead of showing the reflectance map as a function of the top and bottom gate voltages, we now choose the specific choice of $E_{0_{\text{top}}}$ and $E_{0_{\text{bot}}}$, indicated by the vertical ($V_g$ axis) and horizontal ($V_E$ axis) cuts through the reflectance map leave $E_z$ and $\mu$ unchanged respectively, where $\mu$ denotes chemical potential.

Figures 4a and 4b show a periodic modulation of the RP differential reflectance as a function of $V_{\mu}$, particularly in the low $n$ regime. Moreover, the modulation of the top and bottom layer reflectance are correlated and symmetric with respect to the $V_E = -1$ V axis, indicating for this value of $V_E$, the energy detuning of the top and bottom MoSe$_2$ layers is zero. To gain further insight into the structure of correlated changes, we first determine the excitonic resonance energy for the top and bottom layers ($E_{X_{\text{top}}}$ and $E_{X_{\text{bot}}}$) by fitting the reflectance spectrum with two dispersive Lorentzian lineshapes (see Supplementary Information S3) and then plot the derivative of $E_{X_{\text{top}}}$ and $E_{X_{\text{bot}}}$ with respect to $V_E$ in Figs. 4c and 4d. The resulting map shows a remarkable checkerboard pattern that is complementary for the top and bottom layers. Since the blue shift of $X_{\text{top}}$ and $X_{\text{bot}}$ resonance while increasing $V_{\mu}$ (positive $dE_{X_{\text{top}}}/dV_{\mu}$ and $dE_{X_{\text{bot}}}/dV_{\mu}$) corresponds to filling of electrons in the top and bottom layer respectively, the complementary checkerboard like pattern indicates a layer by layer filling of electrons. Note that a similar diagram has been reported in layer resolved capacitance measurement of Landau levels in bilayer graphene$^{42}$.

The observed periodicity in Fig. 4 evidences the existence of Moiré subbands for electrons. In anticipation of the subsequent discussion, we define a layer filling factor $\nu_L$ ($L = \text{"top" or } \text{"bot"}$ indicating top or bottom layer) so that $\nu_L = 1/2$ corresponds to 1 electron per Moiré unit cell of a single layer, and a total filling factor $\nu$ as $\nu = \nu_{\text{top}} + \nu_{\text{bot}}$. From a capacitive model of our device, we determine that $\nu = 1/2$ coincides with a remarkably low electron density of $n = 2 \times 10^{11}$ cm$^{-2}$. At this low electron density, $r_s$ parameter, which describes the ratio of interaction energy to kinetic energy, is estimated to be $r_s \approx 14$. The density periodicity corresponds to a Moiré superlattice lattice constant of $\lambda_{\text{Moire}} = 24$ nm by assuming a triangular superlattice. We indicate the values of $\nu$ corresponding to $\nu = 1/2$, 1, 3/2, 2 with blue dashed lines in Fig. 4c.

Figures 5a and 5b show the $V_E$ dependence of the differential reflectance spectrum for fixed $V_n$ where $\nu = 1/2 (n = 2 \times 10^{11}$ cm$^{-2}$) and $\nu = 1 (n = 4 \times 10^{11}$ cm$^{-2}$), respectively. In Fig. 5a, we find an abrupt shift of exciton energy together with complete oscillator strength transfer between $\text{AP}_{\text{top}}$ and $\text{AP}_{\text{bot}}$. This shows that the electrons are completely and abruptly transferred from one layer to the other layer upon changing $E_z$. Figs. 4c and 4e show the extracted $X_{\text{bot}}$ and $X_{\text{top}}$ energies around $\nu = 1/2$. Remarkably, the abrupt jump in excitonic resonance is pronounced at $\nu = 1/2$, and smeared out for both lower ($\nu = 0.35$) and higher filling factors ($\nu = 0.65$). These measurements show that abrupt transfer of practically all of the $\sim 1500$ electrons within the region we monitor optically is linked to the emergence of an interaction-induced incompressible state in the lowest Moiré subband at $\nu = 1/2$ filling. As the filling factor is increased (decreased) towards $\nu = 1/2$, the electronic system shows an ever stronger layer-paramagnetism, due to enhanced role of interactions, but otherwise exhibits a continuous inter-layer transfer of electrons as a function of $E_z$ that would be expected from a compressible state. Close to $\nu = 1/2$, there is a phase transition to an incompressible state that can be accommodated either in the top or bottom layer (see Fig. 5g).

Figure 5b shows that for $\nu = 1$, the polaron reflectance spectrum as a function of $E_z$ is characterized by 3 plateau-like regions. We attribute the abrupt jumps in the excitonic resonance energy to the transition from ($\nu_{\text{top}}, \nu_{\text{bot}}$) = (0, 1), through (1/2, 1/2), to (1, 0) configurations (see Fig. 5h). This explanation is confirmed by the corresponding changes in the oscillator strength of the AP resonances of the top and bottom layers. In (1, 0) and (0, 1) configurations, we measure a reflectance signal either from $\text{AP}_{\text{top}}$ or $\text{AP}_{\text{bot}}$, consistent with full layer polarization. In the (1/2, 1/2) configuration, we find the oscillator strength of $\text{AP}_{\text{top}}$ and $\text{AP}_{\text{bot}}$ to be identical and equal to half the value obtained under (1, 0) for $\text{AP}_{\text{top}}$. The extracted excitonic resonance energy $X_{\text{bot}}$ and $X_{\text{top}}$ around $\nu = 1$ is shown in Figs. 5d and 5f, respectively. The plateau structure of the (1/2, 1/2) state with abrupt jumps in $n$ for $V_E = -1.2$ V and $V_E = -0.7$ V is clearly visible at $\nu = 1$, but is smeared out for both lower ($\nu = 0.85$) and higher fillings ($\nu = 1.15$). We conclude that as the total $n$ is changed away from $\nu = 1$, both top and bottom layers become compressible, showing smooth changes in layers occupancy as a function of small $E_z$ values ($-1.1 \text{V} < V_E < 0.8 \text{V}$). The emergence of the stabilized (1/2, 1/2) plateau at $\nu = 1$ strongly suggests that there is mutual stabilization of the incompressible electronic state due to the inter-layer electron-electron interactions. If the $\nu_{\text{top}} = 1/2$ or $\nu_{\text{bot}} = 1/2$ had been incompressible for $\nu = 0.85$, we would have observed corresponding plateaus in the excitonic reflectance map. The reflectance data for higher fillings ($\nu = 3/2$ and $\nu = 2$) are shown in the supplementary information (Fig. S5): in stark contrast to the (1/2, 1/2) configuration at $\nu = 1$, a plateau at the (1, 1) electron configuration is missing at $\nu = 2$ filling, indicating that the state with the corresponding integer fillings is not sufficiently stabilized by the inter-layer interactions.

Finally, we emphasize that our identification of $n = 2 \times 10^{11}$ cm$^{-2}$ yielding half-filling of a single-layer Moiré subband is supported by our measurements at $B_z = 7$ T: In Fig. S6 we observe that the plateau structure observed for $\nu = 2$ under full valley polarization of electrons is
identical to that observed for $B_z = 0$ T, even though the total number of electronic states per Moiré subband is halved due to giant valley-spin susceptibility of electrons in MoSe$_2$. This observation shows that the incompressibility is determined by filling of each Moiré site by a single electron, irrespective of its degeneracy.

V. DISCUSSION

The experiments we describe in Sec. IV demonstrate the existence of Mott-like incompressible electronic states for half-filling of the lowest Moiré subband. Unlike prior reports, our experiments are carried out for long Moiré superlattice lattice constant of $\lambda_{\text{Moiré}} = 24$ nm and $r_s \simeq 14$. The weakness of the Moiré potential stemming from the hBN layer separating the two MoSe$_2$ layers in turn ensures that the on-site interaction strength is comparable to, or possibly larger than, the depth of the Moiré potential. In this sense, the homolayer system realizes a rather unique regime of the Fermi-Hubbard model where some of the expectations such as an antiferromagnetic ground-state need not be applicable.

In addition to establishing twisted TMD homo-bilayers as a promising system for investigating Mott-Wigner physics originating from strong electronic correlations, our experiments open up new avenues for exploring interactions between dipolar excitons and electrons confined to flat bands. In particular, the structure we analyzed could be used to realize and study Bose-Fermi mixtures consisting of degenerate electrons strongly interacting with an exciton condensate generated by resonant laser excitation. The phase diagram of such a mixture is currently not fully understood but is expected to provide a rich playground for many-body physics, including but not limited to exciton-mediated superconductivity.

Note added After completion of this work, we became aware of two manuscripts, arXiv:1910.08673 and arXiv:1910.09047, reporting similar results in a different material system.

METHODS

All MoSe$_2$, graphene, and hBN flakes are obtained by mechanical exfoliation of bulk crystals. The flakes are assembled together using the dry transfer technique in an Ar filled glove box. The crystal axis of top and bottom MoSe$_2$ layers are aligned to be close to 0 degree using tear-and-stack technique. The metal electrodes to graphene layers are formed by Ti/Au (5 nm/145 nm). The contact to the bottom graphene gate is formed by Cr/Au (3 nm/147 nm) using the one-dimensional contact technique by etching the hBN layer with reactive ion etching using CHF$_3$/O$_2$ as mixture gas.

The photoluminescence measurements were performed using a HeNe laser (633 nm) as an excitation source. The reflectance measurements were performed using a single mode fiber coupled broadband LED with a center wavelength of 760 nm and a bandwidth of 20 nm. In both photoluminescence and reflectance measurements, we used a long working distance apochromatic objective lens with $N_A = 0.65$ (attocube LT-APO/LWD/VISIR/0.65). All optical spectroscopy measurements have been performed at cryogenic temperature ($T \sim 4$ K).

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AUTHOR CONTRIBUTIONS

Y.S. and I.S. carried out the measurements. Y.S. designed and fabricated the sample. M.K. helped to prepare the experimental setup. K.W. and T.T. grew hBN crystal. Y.S., I.S. and A.I. wrote the manuscript. A.I. supervised the project.

COMPETING INTERESTS

The authors declare no competing financial interests.

ADDITIONAL INFORMATION

Supplementary information for this paper is available online.

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Figure 1. **Device structure and the basic characteristics.** a Schematic image of the device structure. $V_{tg}$ ($V_{bg}$) is the applied voltage to the top (bottom) gate. b, c Schematic image of coupled inter- and intra-layer exciton (b) and electrons in the Moiré lattice probed by the intra-layer exciton (c). Purple planes correspond to MoSe$_2$ layers, and dashed lines indicate a Moiré unit cell. The pink (light blue) circles with + (−) sign indicate holes (electrons) forming excitons, and the electron-hole pair enclosed by the red (yellow) ellipse indicates intra-layer (inter-layer) exciton. The green double arrow in b indicates tunnel coupling of holes through the monolayer hBN barrier. The light green circles in c indicate electrons filling the Moiré lattice. d Optical microscope image of the device. The border of each flake is highlighted with dashed lines, and the material is indicated in the gray box with the corresponding color. (The abbreviation “Gr” stands for Graphene.) e Spatial map of the integrated photoluminescence from 1.59eV to 1.65eV. The blue and yellow dashed lines indicate the boundary of the area of monolayer MoSe$_2$ and MoSe$_2$/hBN/MoSe$_2$, respectively. The inset shows representative PL spectra of monolayer MoSe$_2$ and MoSe$_2$/hBN/MoSe$_2$ measured at the positions indicated with the blue and the yellow stars in the main figure, respectively.
Figure 2. **Electric field dependence of photoluminescence and differential reflectance at charge neutrality.** Gate dependence of photoluminescence \(a\) and differential reflectance \(b\) of MoSe\(_2\)/hBN/MoSe\(_2\). Top and bottom gate voltages are scanned together to tune the electric field at a constant chemical potential (scanned along the dashed line L4 shown in Fig. 3c). The intensity of the photoluminescence \(a\) is shown in log scale. \(c, d\) Magnified plots of \(b\). The corresponding area of \(c\) and \(d\) is indicated by the blue and green dashed rectangulars in \(b\), respectively. \(e, f\) Schematic of the energy bands and the exciton energy alignment under electric fields at the hole resonant conditions of excitons.
Figure 3. **Gate dependence of differential reflectance spectrum of MoSe$_2$/hBN/MoSe$_2$.** a, b Two gates dependence maps of differential reflectance around top (a) and bottom (b) intra-layer exciton resonances ($E = 1.632$eV and $E = 1.640$eV, respectively). The insets of a and b show the differential reflectance spectrum at ($V_{tg}$, $V_{bg}$) = (0V, 0V) (indicated with the white stars in the maps). The magenta and cyan dots in the insets indicate the points where $E = 1.632$eV and $E = 1.640$eV, respectively. c Charge configuration diagram obtained by derivative of the differential reflectance spectrum with respect to energy (sum of the derivatives at $E = 1.632$eV and $E = 1.640$eV). The charge configuration for each layer is indicated by p, i, n which correspond to hole doped, neutral, electron doped, respectively, and shown in the order of (top, bottom). d - f Gate dependence of differential reflectance along the dashed lines L1 (d), L2 (e), and L3 (f) shown in a, b and c. Magenta and cyan dashed lines indicate the top ($E = 1.632$eV) and bottom ($E = 1.640$eV) exciton resonance energies, respectively. AP$_{C}$ and RP$_{C}$ stand for intra-layer attractive and repulsive polarons, where L = "top" or "bot" stands for top or bottom layer, and C = + or − stands for hole or electron as Fermi sea carriers. Charge configuration is written in green together with the green dashed lines indicating the charge configuration transition point.
Figure 4. **Gate dependence of intra-layer exciton resonances in low electron density regime.** a, b Gate dependence maps of differential reflectance around top (a) and bottom (b) intra-layer exciton resonances ($E = 1.6320\text{eV}$ and $E = 1.6402\text{eV}$, respectively). The insets of a and b show the differential reflectance spectrum at $(V_E, V_\mu) = (-1\text{V}, 0.175\text{V})$ (indicated with the white stars in the maps). c, d Gate dependence maps of top (c) and bottom (d) intra-layer exciton resonance energy differentiated by $V_\mu$. 

Figure 5. Electric field dependence of differential reflectance spectrum in low electron density regime. a, b Electric field ($V_E$) dependence of differential reflectance spectrum for each fixed $V_\mu$ at $\nu = 1/2$ and $\nu = 1$. Charging configuration of top and bottom layer is indicated by $(\nu_{\text{top}},\nu_{\text{bot}})$ in green. c, d $V_E$ dependence of $X_{\text{bot}}$ resonance energy around each total filling of $\nu = 1/2$ (c), $\nu = 1$ (d). e, f $V_E$ dependence of $X_{\text{top}}$ resonance energy around each total filling of $\nu = 1/2$ (e), $\nu = 1$ (f). In c - f, the cyan curves are without offset and other curves are displaced by 0.5meV and 1.0meV. g, h Schematic picture of charge configuration with density of states at filling of $\nu = 1/2$ (g) and $\nu = 1$ (h).
Supplementary Information

S1  Differential reflectance and Photoluminescence of monolayer MoSe₂

Here we show gate dependence of differential reflectance and photoluminescence (PL) of monolayer MoSe₂. Fig. S1a shows the gate dependence of the differential reflectance at $E = 1.644\text{eV}$. This energy corresponds to the minimum of the exciton reflection spectrum, as shown in the inset of Fig. S1a. In contrast to the case of MoSe₂/hBN/MoSe₂ (Figs. 2a and 2b in the main text), the dependences on top gate and bottom gate are similar. The blue area where we have absorption from the exciton resonance is where MoSe₂ is neutral. In Fig. S1b, we plot the gate dependence of the differential reflectance spectrum along L0 in Fig. S1a. When MoSe₂ is doped, the exciton resonance around $E = 1.644\text{eV}$ becomes the repulsive polaron and blue shifts. This is accompanied by the emergence of the attractive polaron[1] resonance around $E = 1.619\text{eV}$. Due to this blue shift of the exciton resonance together with the dispersive shape of the exciton resonance as shown in the inset of Fig. S1a, we can detect the changes in the charge configuration as we show in Fig. S1a. In Fig. S1c, we plot gate dependence of the PL spectrum along L0 in Fig. S1a. We observe that exciton PL around $E = 1.644\text{eV}$ is prominent in the neutral regime, while attractive polaron PL around $E = 1.619\text{eV}$ dominates in the electron and hole doped regimes.
Figure S1: **Gate dependence of differential reflectance spectrum and PL of MoSe$_2$.** 

- **a** Two gates dependence map of differential reflectance around exciton resonance ($E = 1.644$eV). The charge configuration is indicated by p, i, n which correspond to hole doped, neutral, electron doped, respectively. The inset shows the differential reflectance spectrum at $(V_{tg}, V_{bg}) = (0V, 0V)$ (indicated with the white star in the map). The magenta dot in the inset indicate the point where $E = 1.644$eV.  
- **b** Gate dependence of differential reflectance along the red dashed line L0 shown in a. Magenta dashed line indicates the exciton resonance energy ($E = 1.644$eV).  
- **c** Gate dependence of PL along the red dashed line L0 shown in a.

**S2 Differential reflectance and Photoluminescence of MoSe$_2$/hBN/MoSe$_2$ at a different spot**

In the main text, we show the spectroscopy data of MoSe$_2$/hBN/MoSe$_2$ at a specific spot. Here we show charge configuration detection and inter- and intra-layer exciton coupling at a different spot in the MoSe$_2$/hBN/MoSe$_2$ section of the sample.

Fig. S2a and S2b show gate dependence the differential reflectance at $E = 1.636$eV and $E = 1.641$eV, respectively. These energies correspond to minima of the exciton resonances in the charge neutral regime (see the insets of Fig. S2a and S2b). As we discussed in the main text and also in section S1, carrier doping induces the blue shift of the exciton resonances due to the formation of repulsive polaron, which results in the abrupt enhancement of the differential reflectance in Fig. S2a and S2b. From the gate dependence, we assign the exciton resonances at $E = 1.636$eV and $E = 1.641$eV to the
bottom and the top intra-layer excitons, respectively. Note that the energy order of top and bottom intra-layer excitons is opposite to the result shown in the main text. This is most likely caused by the inhomogeneity of the strain across the sample. To enhance the border of the transition and visualize the charge configuration of both layers at the same time, we calculated the derivative of the differential reflectance signal with respect to energy at $E = 1.636\text{eV}$ and $E = 1.641\text{eV}$, and added them together (Fig. S2c). In Fig. S2d, e, and f, we show the gate dependence of the differential reflectance spectrum along L’1, L’2 and L’3 (see Figs. S2a, S2b and S2c), respectively. The results are similar to the results we show in the main text including the signature of negative compressibility. The only qualitative difference with respect to the data in the main text is the opposite energy order of the top and the bottom intra-layer exciton resonances.

We also show the electric field dependence of PL (Fig. S3a) and differential reflectance (Fig. S3b) in the charge neutrality regime at this spot. For both measurements, the top and bottom gates are scanned along L’4 indicated in Fig. S2c to control the electric field $E_z$ while keeping the system at charge neutrality. In Fig. S3a, there are inter-layer exciton PL lines which show Stark shift in the high $E_z$ regime (top and bottom side of the figure). In Fig. S3b, we also observe differential reflection from the inter-layer excitons, exhibiting strong Stark shift, for high $E_z$. Fig. S3c and S3d show the magnified plots of the differential reflection for high $|E_z|$ where we observe the inter-layer exciton reflection, and we find avoided crossing structure in both plots. In Fig. S3c, we find the avoided crossings between the top intra-layer exciton ($E \sim 1.641\text{eV}$) and the inter-layer excitons with an electron in the top and a hole in the bottom layer, evidencing the coupling of these states via coherent hole tunneling (Fig. S3e). In Fig. S3d, we find the avoided crossings between the bottom intra-layer exciton ($E \sim 1.636\text{eV}$) and the inter-layer excitons with the opposite dipole, once again confirming that the coherent coupling of these states via
Figure S2: Gate dependence of differential reflectance spectrum of MoSe$_2$/hBN/MoSe$_2$ at a different spot. a, b Two gates dependence maps of differential reflectance around bottom (a) and top (b) intra-layer exciton resonances ($E = 1.636$eV and $E = 1.641$eV, respectively). The insets of a and b show the differential reflectance spectrum at ($V_{tg}, V_{bg}$) = (0V, 0V) (indicated with the white stars in the maps). The magenta and cyan dots in the insets indicate the points where $E = 1.636$eV and $E = 1.641$eV, respectively. c Charge configuration diagram obtained by derivative of the differential reflectance spectrum with respect to energy (sum of the derivatives at $E = 1.636$eV and $E = 1.641$eV). The charge configuration for each layer is indicated by p, i, n which correspond to hole doped, neutral, electron doped, respectively, and shown in the order of (top, bottom). d - f Gate dependence of differential reflectance along the red dashed lines L’1 (d), L’2 (e), and L’3 (f) shown in a, b and c. Magenta and cyan dashed lines indicate the bottom ($E = 1.636$eV) and top ($E = 1.641$eV) exciton resonance energies, respectively.

hole tunneling (Fig. S3f).

All of the results are fully consistent with the results we show in the main text, even though the energy order of top and bottom intra-layer excitons is opposite. Note that even the fine structure of the inter-layer exciton in Fig. S3c and Fig. S3d is similar to
the result which we show in the main text (Fig. 2c and 2d). This observation indicates that the origin of the fine structure is intrinsic to the system and is consistent with Moiré potential picture. In particular, it shows that multiple inter-exciton lines do not stem from random spatial inhomogeneities in the sample.

Figure S3: Electric field dependence of PL and differential reflectance at charge neutrality. Gate dependence of PL (a) and differential reflectance (b) of MoSe₂/hBN/MoSe₂ at the different spot along the dashed line L’4 shown in Fig. S2c. Top and bottom gate voltages are scanned together to tune the electric field but to fix the chemical potential. The intensity of the PL (a) is shown in log scale. c, d Magnified plots of b. The corresponding area of c and d is indicated by the blue and green dashed rectangulars in b, respectively. e, f Schematic image of the energy band and the exciton energy alignment under electric fields when the resonant conditions are achieved. e and f correspond to the situation of c and d, respectively.
S3  Detail of reflectance spectrum fitting and additional spectroscopy data in the low electron density regime

We define the differential reflectance as $\Delta R / R_0 \equiv (R - R_0) / R_0$ where $R_0$ is the background reflectance, measured in a region of the heterostructure without MoSe$_2$ flakes, and $R$ is the experimentally measured reflectance signal. In this work we are interested in the reflectance from MoSe$_2$/hBN/MoSe$_2$ layers. In order to account for reflections and losses from multiple layers of the heterostructure, we assumed a Lorentzian lineshape for the excitons $\chi(E) \propto \frac{\gamma}{E - E_0 + i\gamma/2}$ and effectively describe the total reflectance by $	ext{Im}[\chi(E) \exp(i\alpha)]$ [2]. Here, $\alpha$ is a phase shift that depends in energy and gate voltage. The total reflectance signal is given by

$$
\Delta R / R_0 = A \cos(\alpha) \frac{\gamma^2/2}{(E - E_0)^2 + \gamma^2/4} - A \sin(\alpha) \frac{\gamma(E - E_0)}{(E - E_0)^2 + \gamma^2/4} + C, \quad (S1)
$$

where we introduced the parameter $C$ to capture the broad background signal.

In Fig. S4a and S4b we show once again the gate voltage dependent maps of differential reflectance around top and bottom excitons used in Fig. 4a and 4b of the main text (where the gate voltages axes are $V_E = (7/15)V_{tg} + (8/15)V_{bg}$ and $V_\mu = 0.5V_{tg} - 0.5V_{bg}$). We used eq. S1 to fit the data and plotted the exciton resonance energies as function of gate voltages in Fig. S4c and S4d. The structures observed in Fig. S4a and S4b are well reproduced in Fig. S4c and S4d, respectively. Figures S4e and S4f (and Fig. 4c and 4d in the main text) shows the derivative of the fitted exciton resonance from Fig. S4c and S4d with respect to $V_\mu$. The complementary checkerboard patterns are discussed in the main text, confirming the interpretation of layer by layer filling of electrons with changing chemical potential. Periodic structures also appear in the integrated PL intensity of top
and bottom layer attractive polaron (Fig. S4g and S4h, respectively), which is enhanced at the half integer filling of $\nu = 1/2, 1, 3/2$ and 2. We indicate the charge configuration with $(\nu_{\text{top}}, \nu_{\text{bot}})$, with $\nu_{\text{top}} + \nu_{\text{bot}} = \nu$.

Fig. S5f - g shows additional filling factors, complementing Fig. 5 of the main text; here, we plot $V_E$ dependence of differential reflectance spectrum for fixed $V_\mu$ where $\nu = 0, 1/2, 1, 3/2, \text{ and } 2$ ($\nu = 1/2 \text{ and } \nu = 1$ are presented in Fig 5. in the main text). The $V_\mu$ (or $\nu$) values corresponding to Fig. S5f - g are indicated by a blue dotted lines shown in Fig. S5a - e, showing $dE_{X_{\text{bot}}} / dV_\mu$ (Fig. S4f). For $\nu = 0$ (Fig. S5f), both layers are neutral and therefore $X_{\text{top}}$ and $X_{\text{bot}}$ resonances do not shift. In stark contrast to $(1/2, 1/2)$ state at $\nu = 1$ filling, at $\nu = 2$ filling the $(1, 1)$ state is missing, indicating that the state of integer filling in both layers is not sufficiently stabilized by the interactions.
S4 Differential reflectance spectrum in the low electron density regime under a perpendicular magnetic field

Here we show $V_E$ dependence of the differential reflectance spectrum in the low electron density regime (at $\nu = 2$) obtained under perpendicular magnetic field ($B_z$). Figs. S6b and S6c show the differential reflectance spectrum obtained at $B_z = 7T$ for $\sigma_-$ and $\sigma_+$ circular polarization, respectively. In Fig. S6b, attractive polaron (AP) signal is clearly visible, but is missing in Fig. S6c. $\sigma_+$ ($\sigma_-$) circularly polarized exciton-polaron is formed between K (-K) valley exciton and -K (K) valley Fermi sea electrons [3, 2]. The full $\sigma_-$ polarization of AP reflectance demonstrates that Fermi sea electrons are fully K-valley polarized at $B_z = 7T$, $\nu = 2$.

Remarkably, even though Fermi sea electrons are fully valley polarized and the degeneracy of the Moiré subbands is reduced by a factor of 2, the structure of the spectrum remains essentially the same as the case of $B_z = 0T$ (Fig. S6a). Our experiments therefore show that it is the number of electrons in Moiré unit cell that determines the structure, rather than the degeneracy of the quantum number. This observation is fully consistent with what we would expect from incompressible states induced by electron-electron interactions, where the degeneracy of Moiré subbands is lifted by strong interactions.
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Figure S4: Spectroscopy data of low electron density regime. a, b Gate dependence maps of differential reflectance around top (a) and bottom (b) intra-layer exciton resonances ($E = 1.6320\text{eV}$ and $E = 1.6402\text{eV}$, respectively). The insets of (a) and (b) show the differential reflectance spectrum at ($V_E, V_{\mu}$) = (−1V, 0.175V) (indicated with the white stars in the maps). c, d Gate dependence maps of top (c) and bottom (d) intra-layer exciton resonance energy extracted from fitting of differential reflectance data. e, f Gate dependence maps of top (e) and bottom (f) intra-layer exciton resonance energy differentiated by $V_{\mu}$. g, h Gate dependence maps of top (g) and bottom (h) attractive polaron PL integrated over 1.6000 to 1.6083eV and 1.6125 to 1.6167eV, respectively.
Figure S5: **Differential reflectance spectrum in low electron density regime.** a - e Diagrams indicating electron filling of bottom layer (Fig. S4f). f - j $V_E$ dependence of differential reflectance spectrum for each fixed $V_\mu$ indicated by the blue broken lines in a to e. Charging configuration of top and bottom layer is indicated by $(\nu_{\text{top}}, \nu_{\text{bot}})$. Green broken lines indicate the transition point of charging configuration, which are also indicated in a to e.
Figure S6: **Differential reflectance spectrum under perpendicular magnetic field.** a - c $V_E$ dependence of differential reflectance spectrum at $\nu = 2$ ($V_\mu = 1.627V$). a is the reflectance spectrum at $B_z = 0T$. b and c are the reflectance spectrum at $B_z = 7T$ in $\sigma_-$ and $\sigma_+$ circular polarization, respectively.