Strongly correlated electronic states in a Fermi sea spatially pinned by a MoSe$_2$/WSe$_2$ moiré superlattice

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Two-dimensional moiré materials provide a highly tunable platform to investigate strongly correlated electronic states. Such emergent many-body phenomena can be optically probed in moiré systems created by stacking two layers of transition metal dichalcogenide semiconductors: optically injected excitons can interact with itinerant carriers occupying narrow moiré bands to form exciton-polarons sensitive to strong correlations. Here, we investigate the many-body interactions between excitons and a Fermi sea that is spatially pinned by the moiré superlattice of a molybdenum diselenide (MoSe$_2$) / tungsten diselenide (WSe$_2$) twisted hetero-bilayer. At a multitude of fractional fillings of the moiré lattice, we observe ordering of both electrons and holes into stable correlated electronic states. Magneto-optical measurements reveal extraordinary Zeeman splittings of the exciton-polarons due to exchange interactions between holes in the correlated phases, with a maximum close to the correlated state at one hole per site. The temperature dependence of the Zeeman splitting reveals antiferromagnetic ordering of the correlated holes across a wide range of fractional fillings. Our results illustrate the nature of excitons interacting with a spatially pinned Fermi sea and provide robust evidence for strongly correlated electronic states in MoSe$_2$/WSe$_2$ hetero-bilayers, unveiling the rich potential of this platform for investigations of Fermi-Hubbard and Bose-Hubbard physics.

INTRODUCTION

Two-dimensional (2D) materials have emerged as a new playground to investigate many-body interactions and strongly correlated electronic phenomena. For example, due to a direct bandgap [1], huge exciton binding energies [2], and straightforward control of carrier concentration [3], monolayer transition metal dichalcogenides (TMDs) provide a platform to probe the interaction of an exciton with a Fermi sea (2D electron or hole gas) described by the Fermi-polaron model [4,8]. By extension, TMD moiré heterostructures provide access to a highly tunable many-body physical system consisting of an exciton dressed by a spatially pinned Fermi sea forming charge-ordered electronic states [9].

Stacking two monolayer TMDs with either a lattice mismatch and/or relative twist angle forms a moiré superlattice with a periodicity that far exceeds the interatomic spacing of the constituent crystals. Itinerant electrons in a Fermi sea can be spatially localised by the moiré potential, leading to the formation of flat bands. The suppressed kinetic energy of the charge carriers relative to their on-site Coulomb repulsion energy, $U$, has led to theoretical predictions [10,16] as well as experimental optical [9,17,22] and transport [23,27] investigations of strongly correlated electron and hole phases for different TMD homo- and hetero-bilayer systems. In the simplest scenario, the highest flat valence band in a TMD moiré system can be mapped onto the 2D triangular Hubbard model [10,12,14]. So far however, evidence of Hubbard model physics has only been observed experimentally for angle-aligned WSe$_2$/WS$_2$ hetero-bilayers which form a moiré superlattice due to lattice mismatch [17].

While strongly correlated phenomena have yet to be observed in moiré hetero-bilayers formed from WSe$_2$ and MoSe$_2$, the system remains compelling: it is predicted to form flat conduction and valence minibands [28,29] and is an excellent candidate for the realisation of a wide range of strongly correlated states including Wigner crystals [12,13], Mott insulators [10,11,14] and charge-transfer insulators [16]. Unlike TMD heterobilayers with different chalcogen atoms, the energetic interplay between Coulomb repulsion and kinetic energy in the WSe$_2$/MoSe$_2$ system is highly tunable with relative twist angle due to the small (0.2%) lattice mismatch [30]. In addition, the moiré potential in WSe$_2$/MoSe$_2$ hetero-bilayers has led to the observation of trapping of interlayer excitons at specific atomic registries in the moiré lattice [31,32]. To date, hetero-bilayer WSe$_2$/MoSe$_2$ remains the only moiré system to conclusively exhibit exciton trapping.

Here we optically investigate the Coulomb and magnetic interactions arising in a Fermi sea pinned by a moiré superlattice in a MoSe$_2$/WSe$_2$ hetero-bilayer. First, in the absence of Fermi sea dressing, we observe a splitting of the MoSe$_2$ A 1s intralayer exci-
ton into two moiré excitons due to miniband formation. Then, as the Fermi level is tuned, we observe ordering and re-ordering of itinerant carriers into a multitude of correlated states as evidenced by abrupt changes in the oscillator strength, energy, and linewidth of the exciton-polarons. We observe these correlated states at positive (electron) and negative (hole) fractional fillings (ν) of the moiré lattice, including: ν = ±1/6, ±1/3, ±1/2, ±2/3, ±5/6, ±1, ±5/4, where |ν| = 1 represents a single carrier per moiré site. We assign the ±1 state to be a Mott [17, 18, 21] or charge-transfer [19] insulator state and the rest to be generalised Wigner tons dressed by a spatially pinned Fermi sea in optical Fermi-hole sea. Our results highlight the role of excitations of highly tunable 2D Fermi-Hubbard or Bose-Hubbard models [39].

DEVICE STRUCTURE, FERMI-POLARONS, AND MOIRÉ EXCITONS

Figure 1a shows a sketch of our dual-gated heterobilayer device, consisting of a monolayer MoSe2 and a monolayer WSe2 vertically stacked with a twist angle (Δθ) of ~57°. The relative twist angle from perfect 2H stacking (i.e., Δθ = 60°), estimated from the optical micrograph of the heterobilayer (see Supplementary Section 1) and confirmed by our gate dependent measurements (described later), is beyond the proposed critical angle for lattice reconstruction [40, 41]. The hetero-bilayer was encapsulated by hexagonal boron nitride (hBN) layers with nearly identical thicknesses (~18 nm). Graphene layers act as electrical contacts for the top, bottom, and hetero-bilayer gates (see Ref. 34 for more details). Moreover, the combination of the layer twist and the lattice mismatch between MoSe2 and WSe2 results in the formation of a triangular moiré superlattice in our device (see sketch in Fig. 1b) with a period of ~6 nm. This causes a periodic variation in the interlayer hopping that results in a flattening of the conduction and valence bands in the type-II band structure characteristic of TMD heterobilayers [42, 43] (see Fig. 1c). In addition to the intralayer excitons of the constituent TMDs, which can be probed via absorption spectroscopy, the type-II band alignment enables the formation of spatially indirect interlayer excitons that are best probed by photoluminescence (PL) (see sketch in Fig. 1c) [44, 45]. The interlayer excitons can be confined by the moiré potential at specific atomic registries giving rise to quantum-dot-like light emission [32, 34], as demonstrated for both neutral and charged interlayer excitons in our device [35, 36, 37].

The moiré lattice carrier concentration is tuned via the application of a gate voltage (Vg) between the top/bottom graphene contacts and the hetero-bilayer. To investigate doping-dependent phenomena, we perform differential reflection contrast (∆R/R0) spectroscopy as a function of Vg, where ∆R = Rs − R0, and R0 (R0) is the intensity of the light reflected by the hetero-bilayer (substrate). First, we monitor the doping dependence of the intralayer excitons in the individual constituent monolayers in the device away from the hetero-bilayer region. Figure 1b shows the ∆R/R0 spectrum of the 1s state of A excitons in hBN-encapsulated ML MoSe2 (left) and ML WSe2 (right) regions of the sample as a function of Vg. When Vg is positive (negative) the sample is doped with additional electrons (holes). At charge neutrality (Vg = 0 V) only the neutral A-excitons are present in both MoSe2 (X0 Mo) and WSe2 (X0 W). When additional holes are injected into the monolayers (Vg < 0 V), the positive attractive polarons AP+ Mo and AP+ W are formed at lower energies than X0 Mo and X0 W, respectively. With increasing hole doping, the neutral exciton and positive attractive polaron peaks in both materials blue-shift, in agreement with the formation of repulsive and attractive exciton polarons, respectively [28, 46, 47]. In a similar manner, electron doping (i.e., Vg > 0 V) leads to the formation of negative attractive polarons at lower energies than the respective neutral excitons. In this regime, the spin-ordering of the conduction bands in WSe2 results in a fine-structure splitting of the attractive polarons into spin-singlet AP− S (intravalley) and spin-triplet AP− T (inter valley) configurations [3], while the spin-ordering of the conduction bands in MoSe2 only results in the formation of intervalley negative attractive polarons AP− Mo. Similar to the hole doping regime, further electron doping leads to an energy blue-shift of both the neutral excitons and negative attractive polarons.

The doping dependence of the intralayer excitons in the MoSe2/WSe2 hetero-bilayer is markedly different to that observed for the individual layers. ∆R/R0 as a function of Vg for the hetero-bilayer is shown in Figure 1c. At charge neutrality, we observe three excitonic resonances: X0 W at higher energy, and two resonances separated by 36 meV in the spectral range corresponding to X0 Mo, which we label as X0 Mo,low (low energy) and X0 Mo,high (high energy). We assign the two MoSe2 peaks to be a consequence of the formation of conduction moiré mini-bands, arising from the band folding at the edges of the reduced Brillouin zone [48]. Following
FIG. 1. Device structure and characterisation. a, Sketch of the dual-gated WSe$_2$/MoSe$_2$ hetero-bilayer. Graphene layers are used as top, bottom, and hetero-bilayer electrical contacts, while 18-nm-thick hBN layers are used as dielectric spacers [34]. b, Illustration of the top view of a moiré superlattice with a twist angle $\Delta \theta$. c, Schematic type-II band structure of the 2H-WSe$_2$/MoSe$_2$ hetero-bilayer. Purple and orange curves denote bands from MoSe$_2$ and WSe$_2$, respectively. The vertical wavy arrows represent the photon absorption by intralayer excitons in each monolayer, while the wavy black arrow represents the photon emission from interlayer excitons in the type-II hetero-bilayer. d, Density plot of the reflection contrast $\Delta R/R_0$ of the 1s state of $X$ excitons in hBN-encapsulated ML MoSe$_2$ (left) and ML WSe$_2$ (right) regions of the sample as a function of the applied voltage gate $V_g$. When $V_g$ is positive (negative) the sample is electron (hole) doped. e, $\Delta R/R_0$ vs $V_g$ spectra from the WSe$_2$/MoSe$_2$ hetero-bilayer. f, PL spectrum of the moiré trapped interlayer excitons in the same location as panel e under 10 nW excitation power at an excitation energy of 1.63 eV.

The empirical model in Ref. [49], we estimate the energy splitting between the moiré-split $X^0_{Mo}$ excitons to be $\sim 38$ meV for $\Delta \theta \sim 57^\circ$ (see Supplementary Section 6), in good agreement with the experimental value. Further, with increasing electron (hole) doping $X^0_W$ ($X^0_{Mo,1}$) dominate the spectrum, as expected for a type-II band alignment [43, 50]. With increasing doping, the peaks undergo modulations in oscillator strength, transition energy, and linewidth which are not observed in the monolayer regions. Similar modulations of the excitonic transitions, observed in WSe$_2$/WS$_2$ hetero-bilayers [17, 20, 21, 51], have been attributed to the suppressed charge screening originating from the formation of correlated crystalline phases at different fractional fillings of the moiré superlattice. In order to corroborate the presence of a robust moiré lattice at the same spatial position in our sample, we measure the low-temperature (4 K) PL spectrum at charge neutrality using confocal spectroscopy. Figure 1f shows a low-excitation-power (10 nW) PL spectrum, revealing a series of discrete peaks with narrow line-widths ($<100 \mu$eV) that demonstrate the existence of an underlying moiré lattice responsible for the interlayer exciton trapping [32–34].

CORRELATED ELECTRONIC STATES

To visualise the formation of correlated states in more detail, in Fig. 2a, we plot the first derivative of the reflectance spectra of Fig. 1 with respect to photon energy ($d(\Delta R/R_0)/dE$). We employ the parallel plate capacitance model to estimate the dependence of the nominal
carrier concentration \( n \) on the applied \( V_g \). Using the density of moiré sites \( n_0 \) corresponding to \( \Delta \theta \sim 57^\circ \), we estimate the \( V_g \)-dependent nominal fractional filling \( \nu = n/n_0 \) of the moiré lattice (see Supplementary Section 4). The excitonic features shown in Fig. 2 exhibit strong modulations in their transition energy, linewidth and oscillator strengths for applied voltages close to the nominal \( V_g \) values corresponding to \( \nu = 0 \) and \( \pm 1 \). Moreover, Fig. 2 also reveals that each monolayer in the WSe\(_2\)/MoSe\(_2\) hetero-bilayer is capable of sensing the doping-induced changes in their dielectric environment originating from the fractional filling of the other layer, similar to the effects observed using a WSe\(_2\) sensor layer in proximity to a WSe\(_2\)/WS\(_2\) heterostructure [21]. Figure 2 shows an example of the sensing capabilities of the MoSe\(_2\) layer for hole doping of the WSe\(_2\) layer: the transition energies of \( X^0_{Mo,1} \) and \( X^0_{Mo,2} \) blue-shift and peak at \( \Delta V_g = -1.34 \) V, consistent with a decrease in the permittivity of the heterostructure arising from the formation of a correlated insulating state at 1 hole per moiré site in the WSe\(_2\) layer [17, 20]. In addition to the modulation in the transition energy, the
linewidth of $X_{Mo,1}^0$ also presents a clear minimum at $\nu \approx -1$ (see Fig. S2), which can be understood as the result of reduced charge disorder originating from a correlated insulating state [9]. These results demonstrate the potential of intralayer excitons as sensors that can probe the formation of correlated states in the adjacent layer (see sketch in Fig. S2) and corroborate the calibration of $\nu = \pm 1$ in our device. To estimate the $V_g$ values corresponding to other fractional fillings of the moiré lattice, we assume a linear dependence of $\nu$ with $V_g$ and extrapolate from the experimental $V_g$ values determined for one hole/electron per site, as shown in the right panel of Fig. 2b. To increase the sensitivity to doping-induced modulations of the reflectance signal, we plot the first derivative of $\Delta R/R_0$ with respect to $V_g$ ($d(\Delta R/R_0)/dV_g$) as a function of $V_g$ (see left panel of Fig. 2a). The $d(\Delta R/R_0)/dV_g$ spectrum highlights a series of abrupt changes in the reflectance signal at $\nu = 0, \pm 1/6, \pm 1/3, \pm 1/2, \pm 2/3, \pm 5/6, \pm 1, \pm 5/4$, (as indicated by the horizontal lines in Fig. S2), suggesting the formation of correlated states at these fractional fillings of the triangular lattice. These results reveal symmetrical loading of carriers, with an identical $\Delta V_g = \pm 1.34$ V required to fill the moiré superlattice with either one electron ($V_g = 1.34$ V) or one hole ($V_g = -1.34$ V) per site, respectively. We tentatively assign the stable phases at $\nu = \pm 1$ to be either Mott [17,18,21] or charge-transfer [16] insulator states and the remaining states to be generalised Wigner crystals [13,18,20,21,27].

To gain deeper insight into the strength of the electronic correlations in our system, we investigate the melting temperature of the different correlated states. Supplementary Figure S8 shows the dependence of $d(\Delta R/R_0)/dV_g$ on $V_g$ for temperatures ranging from 4 K to 90 K. With increasing temperature, the abrupt changes in the $d(\Delta R/R_0)/dV_g$ spectrum (indicative of correlated state formation in both the electron and hole doping regimes, see Fig. 2b) progressively smooth out until they can no longer be observed at 90 K. We quantitatively estimate a melting temperature of $\approx 55$ K for the correlated state at one hole per moiré site (see Supplementary Section 10).
We note that $RP_B = 1$.

definitions in AP, the solid lines are fits of the experimental spectra in Fig. 4a reveal a clear positive which we estimate the energy of the resonances. The model described in Supplementary Section 3, from the different effective hole doping in the magnetic fields. Such spin polarisation originates from the different effective hole doping in the $\pm K$ valleys induced by the large Zeeman splitting.

Next, we investigate the fractional-filling-dependence of the Zeeman splitting of the $RP^+_W$ and $AP^+_W$ resonances. Figure 4a shows $\sigma^-$- (blue) and $\sigma^+$-resolved (red) $\Delta R/R_0$ spectra at representative hole $\nu$ values at $B = 1$ T. The dots represent experimental data while the solid lines are fits of the experimental $\Delta R/R_0$ to the model described in Supplementary Section 3, from which we estimate the energy of the resonances. The spectra in Fig. 4a reveal a clear positive $\Delta E$ for all hole $\nu$, although with a magnitude that depends strongly on $\nu$. Figure 4b shows the $B$-field-dependent Zeeman splitting of $AP^+_W$ from $-1$ to $1$ T at representative hole $\nu$ values. The estimated Zeeman splitting exhibits a linear dependence with $B$ at small fields (i.e., $|B| < 1$ T). We note that $RP^+_W$ shows a similar positive linear dependence with $B$ at small fields, although it saturates at larger $B$ (see Suppl. Fig. S5). The linear evolution of $\Delta E$ at small $B$ can be associated to an effective exciton $g$-factor according to $\Delta E(B) = g\mu_B B$, where $\mu_0$ is the Bohr magneton. Figure 4c shows the evolution of the $g$-factor of $AP^+_W$ as a function of the hole $\nu$ extracted from linear fits of $\Delta E$ in the range $|B| \leq 1$ T (solid lines in Fig. 4b). As already inferred from the results in Fig. 4b, the $g$-factor of $AP^+_W$ shown in 4c exhibits a strong dependence on $\nu$, peaking around $\nu \approx -1$, where it reaches a maximum value of $g \sim 145$.

Figure 4d shows the Zeeman splitting of $AP^+_W$ measured for $|B| \leq 1$ T at $\nu = -1$ for different temperatures, where we observe the slope of the Zeeman splitting (and therefore the $g$-factor) decreases with increasing temperature. Figure 4e shows the evolution of the measured $g$-factor of $AP^+_W$ as a function of temperature for $\nu = -1$ in the temperature range in which the oscillator strength.
and linewidth of $\text{AP}_W^+$ enable a reliable estimate. We observe that the $g$-factor ($\chi$) decreases by a factor $\sim 5$ when the temperature increases from 4 K to 39 K. We assume that the interaction-induced enhancement of the attractive polaron $g$-factor is proportional to the magnetic susceptibility of the correlated states (e.g. $\chi \propto g$) and observe that the decrease of $g$-factor with increasing temperature follows a Curie-Weiss law $\chi^{-1} \propto T - \theta$ (red solid line in Fig. 4). From the fit in Fig. 4, we estimate a Weiss constant of $\theta = -4.6 \pm 0.9$ K, which suggests an antiferromagnetic behaviour of the interactions between the localised hole moments for $\nu = -1$. Supplementary Figure S10 shows the temperature dependence of the $\text{AP}_W^+$ $g$-factor for a range of hole filling factors from $\nu = -0.7$ to $\nu = -1.37$. The Weiss constants extracted from the Curie-Weiss fits are negative for all the explored hole filling factors, suggesting an antiferromagnetic phase for all correlated hole states. We note we observe no magnetic hysteresis in the Zeeman splitting at $\nu = -1$ when the magnetic field is swept from negative to positive values followed by a subsequent positive to negative sweep (see Supplementary Section 8). In contrast to the large attractive polaron $g$-factor enhancement observed for $\nu = 0$, $\text{AP}_W^+$ and $\text{RP}_W^+$ at low magnetic fields, the $g$-factor of $\text{AP}_W^+$ shows a significant decrease with increasing magnetic field for $\nu = -1$. This behaviour is indicative of the interaction-induced enhancement of the attractive polaron $g$-factor and provides further evidence for the antiferromagnetic phase of the correlated hole states.
hancement observed under hole doping, we only observe a modest g-factor enhancement in the electron doping regime (see Supplementary Section 9).

**DISCUSSION**

The extraordinary g-factors observed under hole doping can be understood by considering the effect of a magnetic field on a localised hole in the triangular moiré superlattice. As a result of exchange interactions with other holes in its environment, such a hole experiences an effective magnetic field which is the sum of the externally applied field and the field induced by the other holes which in turn is proportional to the magnetisation $M$ of the localised hole gas (i.e., $\propto \lambda_X M$, with $\lambda_X$ being a coupling constant).

To calculate the induced field for a given fractional filling, we first determine the configuration of localised holes, and then determine the corresponding g-factor enhancement, $g^*/g$, of a localised hole are calculated within mean-field theory (see Supplementary Section 12 for details). To describe the spin response of this arrangement of charges, we consider a Heisenberg Hamiltonian with distance-dependent antiferromagnetic isotropic exchange interactions $J(r) = J_0 \exp(-r/r_0)$ with $J_0$ denoting the magnitude of the exchange coupling at the characteristic length scale $r_0$. For this Hamiltonian, the induced magnetic field and the corresponding g-factor enhancement, $g^*/g$, of a localised hole are calculated within mean-field theory (see Supplementary Section 12 for a detailed description). To obtain the effective g-factor of the attractive exciton-polaron which is probed in our experiments, we assume that its g-factor enhancement due to exchange interactions with localised holes is the same as that of a single hole, but that the "non-enhanced" g-factor $g$ can be different. The experimental value of this non-enhanced g is unknown since the g-factors of exciton-polarons are dependent on paramagnetic interactions and phase-space filling effects as carrier concentration is changed, even for monolayer TMDs [6]. Therefore, we treat $g$ as an adjustable parameter choosing its value such that the calculated and experimentally measured g-factors agree at $\nu = -1$.

Figure 4f shows that the filling dependence of the calculated g-factor is in good qualitative agreement with the experimental results. Specifically, it reaches a maximum at $\nu = -1$, where the average number of occupied moiré sites around the localised hole is largest and the strong exchange interactions between neighboring spins give rise to a large effective magnetic field. The model also captures the plateau-like feature between $\nu = -1/3$ and $-2/3$. In contrast to the experimental findings, however, the calculated g-factor is symmetric around $\nu = -1$. Further theoretical work is required to understand this discrepancy.

Finally, we estimate $U/t$ in our device. The antiferromagnetic coupling between neighbouring spins due to the kinetic exchange mechanism can be estimated as $J \approx -t^2/U$, where $t$ is the hopping amplitude between neighbouring moiré lattice sites. Using the value of $\theta$ at $\nu = -1$ we estimate $J \approx -0.4$ meV. By combining $J$ with the estimated melting temperature of the correlated state at $\nu = -1$ ($\sim 55$ K) we obtain $U/t \approx 3.5 \pm 0.4$. This experimental value agrees well with predicted values for MoSe$_2$/WSe$_2$ heterostructures with stacking angles $\sim 3^\circ$ and $\sim 57^\circ$ [10].

**CONCLUSION**

Our results illustrate the sensitivity of exciton-polarons to optically probe correlated states in moiré heterostructures. Using the changes in energy, oscillator strength, and linewidth of intra-layer excitons dressed by itinerant carriers occupying narrow electronic moiré bands in a MoSe$_2$/WSe$_2$ hetero-bilayer, we observe the formation of correlated electron and hole states at a multitude of fractional fillings of the moiré lattice. In addition, we observe the magnetic interactions within the correlated hole states via both the attractive and repulsive WSe$_2$ exciton-polarons, which exhibit enhanced Zeeman splittings due to exchange interactions among the moiré pinned carriers. Through temperature dependent measurements, the magnetic ordering of the correlated holes is shown to be antiferromagnetic in the range $\nu = -0.7$ to $\nu = -1.37$, and the $U/t$ ratio of our device is estimated to be $\sim 3.5$. Further investigations could exploit the small lattice mismatch between MoSe$_2$/WSe$_2$, which enables a highly tunable moiré period, to simulate condensed matter phase diagrams over a large range of $U/t$ ratios. Our observation of the formation of flat electronic bands compliments recent reports of moiré trapped interlayer excitons in MoSe$_2$/WSe$_2$ hetero-bilayer [32,38] and highlights the exciting prospects to investigate Fermi-Hubbard and Bose-Hubbard physics in this system.

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