Electrically tunable Feshbach resonances in twisted bilayer semiconductors

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Moiré superlattices in transition metal dichalcogenide bilayers provide a platform for exploring strong correlations with optical spectroscopy. Despite the observation of rich Mott-Wigner physics stemming from an interplay between the periodic potential and Coulomb interactions, the absence of tunnel coupling—induced hybridization of electronic states has ensured a classical layer degree of freedom. We investigated a MoSe2 homobilayer structure where interlayer coherent tunneling allows for electric field—controlled manipulation and measurement of the ground-state hole-layer pseudospin. We observed an electrically tunable two-dimensional Feshbach resonance in exciton-hole scattering, which allowed us to control the strength of interactions between excitons and holes located in different layers. Our results may enable the realization of degenerate Bose-Fermi mixtures with tunable interactions.

The ability to tune the sign and the strength of interactions between particles is key for realization of quantum simulators (7). In ultracold-atom experiments, the requisite tunability of interactions is achieved by using Feshbach resonances (2–5); this has enabled many of the impressive developments in the field, including the preparation of ultracold molecules and the observation of BEC (Bose-Einstein condensate) to BCS (Bardeen-Cooper-Schrieffer state) transition (6–9). As a consequence of their rich internal energy level structure, two cold atoms prepared in a given internal state with total spin \( S \) can be brought into resonance with a bound molecular state of the two atoms with \( S ≠ S \) by applying an external magnetic field. Finite hyperfine coupling allows two atoms in a low-energy scattering state (open channel) to hybridize with a bound molecular state (closed channel), resulting in fully tunable scattering phase shifts in the open channel.

Recently, twisted bilayers of two-dimensional (2D) materials have emerged as a new platform for investigating the physics of strongly correlated electrons (10–17). In these synthetic quantum materials, partial quenching of kinetic energy in emerging flat bands and strong Coulomb interactions play a central role. The role of optical excitations, by contrast, has been mainly restricted to spectroscopy, revealing features such as incompressibility and emergent charge order in Mott-Wigner states of the interacting electron system (14–16, 18). Enhancing the strength of exciton-electron interactions could drastically enrich the physics of this system by allowing for photoinduced correlated states in degenerate Bose-Fermi mixtures.

Here, we demonstrate an electrically tunable Feshbach resonance in a twisted homobilayer MoSe2 heterostucture, providing the basis for tunable interactions between bosonic (exciton or polariton) and fermionic (electron or hole) particles. While the presence of a layer pseudospin degree of freedom allows us to obtain open and closed channels that can be tuned in and out of resonance, coherent hole tunneling between the layers provides the counterpart of hyperfine interaction that leads to hybridization of scattering and bound molecular states.

The interplay among strong Coulomb interactions, the moiré superlattice, and coherent interlayer hole tunneling is key to understanding the rich physics of the twisted MoSe2/hBN (hexagonal boron nitride)/MoSe2 heterostructure that we study. To unravel its properties, we use the electric field \( E_z \) and hole density dependence of the exciton-polaron spectrum (19, 20). We find a striking moiré site filling factor dependence of the many-body system: For unity filling of the moiré superlattice \( (\nu = 1) \), we observe Mott-like correlated insulator states in the top or bottom layers, as evidenced by the observation of an umklapp resonance (18). More generally for \( \nu ≤ 1 \), the hole states show no evidence for layer hybridization. For \( \nu > 1 \), however, the excess holes occupy layer hybridized sites in the moiré lattice, leading to strong \( E_z \) dependence of the associated attractive exciton-polaron resonance energy. Upon further increasing \( E_z \) for \( \nu > 1 \), we find that the umklapp resonance disappears, which suggests that the holes are no longer confined to high-symmetry points of the moiré lattice. Precisely in this parameter regime, we observe an asymmetric avoided crossing in the optically excited state; this demonstrates the hybridization of the top-layer exciton with a continuum of scattering states composed of a top-layer exciton and a bottom-layer hole. This observation provides unequivocal evidence for a 2D Feshbach resonance (21) and the presence of attractive and repulsive polaron branches associated with the emerging interlayer Feshbach molecule.

Our device consists of two MoSe2 layers encapsulated between two thick hBN layers and separated by a monolayer hBN tunnel barrier (Fig. 1A). Top- and bottom-gate voltages, \( V_{tg} \) and \( V_{bg} \), are applied using transparent thin graphene sheets while the two transition metal dichalcogenide (TMD) layers are grounded (16). Figure 1B shows a typical differential reflectance spectrum \( |\Delta R/R_0| = (R - R_0)/R_0 \) as a function of top-gate voltage with a fixed back-gate voltage \( V_{bg} = -4 \) V. In the range \( V_{tg} = ± 6 \) V, the excitation spectrum is dominated by the neutral exciton resonances of the top and bottom layers at \( E_{X0} = 1.632 \) eV and \( E_{X0} = 1.64 \) eV, indicating that both layers are charge-free. By setting \( |V_{tg}| > 6 \) V, the top-layer exciton evolves into the repulsive polaron (RP). In addition, an attractive polaron (AP) branch appears at lower energies \( E_{AP0} = 1.607 \) eV (19).

We use \( V_{tg} = 0.67 V_{tg} \) and \( V_{tg} = 0.25 V_{tg} + 0.472 V_{bg} \) to denote a linear combination of \( V_{tg} \) and \( V_{bg} \) that results in a minimal change to \( E_z \) and to the chemical potential \( \mu \), respectively (22). Figure 1C shows \( V_{tg} \)-dependent \( |\Delta R/R_0| \) at \( \nu = 1 \left( V_{tg} = -4.985 \right) \) V. By scanning across \( V_{tg} = -0.2 \) V (corresponding to \( E_z = 0 \)), an AP branch abruptly disappears in conjunction with the appearance of an AP state. Similarly to the electron-doped regime (16), this abrupt transfer of all holes from one layer to the other indicates a first-order phase transition between incompressible Mott-like hole states in the top and bottom layers. This incompressible state results in a periodic potential for excitons and the emergence of an umklapp band (18). Figure 1D shows the derivative of \( |\Delta R/R_0| \) with respect to energy as a function of \( V_{tg} \) at \( \nu = 1 \). The umklapp resonances appear 2.8 meV above the RPs and are labeled \( U_{top} \) and \( U_{bot} \). We emphasize that the dominant AP resonance energy remains, to first order, insensitive to the emergence of a periodic charge distribution inherent in the Mott-like correlated state in weak moiré potentials. This observation is consistent with the interpretation of an AP quasiparticle as a
collective trion excitation out of electrons or holes in the ground state (23); the ground-state charge mobility does not have a measurable effect on the energy of the AP resonance.

Increasing the carrier density to \( n \geq 1 \) reveals a qualitative difference between electron and hole doping: In contrast to changes in electron densities in which a periodic structure of incompressible states appears in the spectra (16), upon increasing the hole density, both AP\(_{\text{top}}\) and AP\(_{\text{bot}}\) split and signatures of an avoided crossing appear in one of their branches. Figure 2A shows \( V_E\)-dependent \( \Delta R/R_0 \) of AP resonances at \( n = 3 \) (\( V_E = -5.325 \) V). Varying \( V_E \) from negative to positive, the AP\(_{\text{top}}\) splits into two branches at \( V_E = -0.7 \) V, where the energy of one branch remains unchanged and the other branch blue-shifts. These observations show that a subset of ground-state holes occupy layer-hybridized states, whereas the others are localized within a layer. A complementary picture is observed for the AP\(_{\text{bot}}\) by scanning \( V_E \) in the opposite direction. Figure 2B shows the \( V_E\)-dependent photoluminescence (PL) spectra at \( n = 3 \), where the avoided crossing structure is fully visible in both AP\(_{\text{top}}\) and AP\(_{\text{bot}}\) with a coupling strength of \( 2\Gamma = 6 \) meV. The two radiative decay channels, ensuring the observation of both PL branches in the avoided crossing, provide clear evidence for the existence of hybridized states. The absence of the lower-energy branch in \( \Delta R/R_0 \) in turn allows us to conclude that the higher-energy AP resonances exhibiting strong \( V_E\)-dependence (AP\(_{\text{top}}^h\) and AP\(_{\text{bot}}^h\)) originate from the holes occupying the lower-energy hybridized state. Figure 2C and D, shows the calculated spectra as function of energy detuning between the top and bottom layer for reflection and PL, respectively (22).

The blue-shifting branches of AP\(_{\text{top}}\) and AP\(_{\text{bot}}\) have comparable strength for \( V_E = -0.2 \) V. Upon increasing \( E_x \) we observe that the \( \Delta R/R_0 \) strength of AP\(_{\text{top}}^h\) decreases while that of AP\(_{\text{bot}}^h\) increases; likewise, upon decreasing \( E_x \), the \( \Delta R/R_0 \) strength of AP\(_{\text{bot}}^h\) decreases while that of AP\(_{\text{top}}^h\) increases. This observation can be understood as arising from the \( E_x \) dependence of the probability amplitude for finding the hybridized hole state in the top or bottom layer: For large and positive \( E_x \), the hybridized hole state is predominantly in the bottom layer, and for large and negative \( E_x \), the hybridized hole state is predominantly in the top layer. In the language of spins, the Bloch vector of the collective hole-layer pseudospin rotates from the equator to the south pole by increasing \( E_x \) and from the equator to the north pole by decreasing \( E_x \). By contrast, different AP\(_{\text{top}}\) and AP\(_{\text{bot}}\) resonance energies allow us to measure the projection of the collective layer pseudospin of holes occupying the hybridized states.

Fig. 1. Device structure and basic characteristics. (A) Schematic of the device structure. The applied electric field and the optical axis are along the \( z \) direction. (B) Top gate–dependent differential reflectance spectra at a fixed back-gate voltage \( V_{bg} = -4 \) V. \( X_{\text{top}}\) and \( X_{\text{bot}}\), top- and bottom-layer excitons [split as a result of strain (16)], RP\(_{\text{top}}^+\) and RP\(_{\text{top}}^-\), top-layer positive and negative repulsive polarons; AP\(_{\text{top}}\) and AP\(_{\text{bot}}\), top-layer positive and negative attractive polarons. (C) \( V_F\)-dependent differential reflectance spectra at a fixed chemical potential for unity filling of the moiré superlattice \( n = 1 \). RP\(_{\text{top}}^+\) and AP\(_{\text{bot}}\), bottom-layer positive repulsive and attractive polaron resonances. (D) \( V_F\)-dependent differential reflectance spectra differentiated with respect to photon energy at a fixed chemical potential for \( n = 1 \). \( U_{\text{top}}\) and \( U_{\text{bot}}\), top- and bottom-layer umklapp resonances. Blue dashed lines mark the energy of the repulsive polarons and the associated umklapp resonances.

Fig. 2. Coherent hole tunneling at moiré filling factor \( n = 3 \). (A) \( V_F\)-dependent differential reflectance spectra at a fixed chemical potential for \( n = 3 \), where each lattice site accommodates three holes. AP\(_{\text{top}}\) and AP\(_{\text{bot}}\), top- and bottom-layer attractive polarons; AP\(_{\text{top}}^h\) and AP\(_{\text{bot}}^h\), top- and bottom-layer attractive polarons originating from the hybridized hole state. (B) \( V_E\)-dependent PL spectra at a fixed chemical potential for \( n = 3 \). (C and D) Calculated reflectance spectra (C) and PL (D) at \( n = 3 \) as function of energy detuning between top and bottom layers. A.U., arbitrary units.
Figure 3A shows $V_E$-dependent $\Delta R/R_0$ at $\nu = 2$ ($V_E = -5.212$ V). For small detuning between the single-particle states of the two layers ($-0.5 V < V_E < 0$ V), $APH_{\text{top}}$ and $APH_{\text{bot}}$ are not visible, indicating that hybridized hole states are not occupied. This gap in the AP spectra is prominent for $1 < \nu \leq 2$ (Fig. 3B). In contrast, $APH_{\text{top}}$ and $APH_{\text{bot}}$ resonances can be observed for all $V_E$ for $\nu > 2$ ($V_E = -5.250$ V), as can be seen in Fig. 3C. The emergence of such a gap in the optical excitation spectrum provides clear evidence that the holes occupy different sites in the moiré lattice at large ($|V_E| > 0$ V or $-0.5 V > V_E$) and small ($-0.5 V < V_E < 0$ V) detunings for fixed $\nu$.

The observations reported in Figs. 2 and 3 for small $E_z$ can be captured by a simple single-particle model that assumes that holes are subject to a superlattice potential with three minima, strong on-site repulsion, and vanishing hopping between the different sites within a layer. The three minima are attributed to different sites in the moiré superlattice for which the local stacking of the metal (M) and chalcogen (X) atoms of the two layers are denoted by MM, MX, and XM. We conclude, from the lack of avoided crossing signatures below $\nu \leq 1$, that the lowest single-particle moiré subbands are located at $XM^t$ ($MX^b$) for $V_E < -0.2$ V ($V_E > -0.2$ V) (24), with the superscripts indicating top- or bottom-layer holes. When $1 < \nu \leq 2$, the second moiré subband starts to fill: For large energy detuning between the layers, this subband is located at the hybridized MM site, and for small energy detuning, the two lowest-energy states are $XM^t$ and $MX^b$. For $\nu > 2$, the third moiré subband starts to fill up, leading to the closing of the gap in the avoided crossing in $\Delta R/R_0$ and indicating that the MM sites are occupied for all energy detunings between the layers. Figure 3E is a schematic of the model showing the energy of the hole states at the three high-symmetry points of the two layers for $E_z = 0$. Figure 3F shows the calculated hole energy levels using the simplified model (22).

We emphasize, however, that although the general spectral features for small $|E_z|$ are qualitatively captured by this model, the absence of an umklapp resonance for $\nu \geq 2$ at large $|E_z|$ indicates that when all holes reside in one layer, they do not form a periodic charge distribution (22). We speculate that delocalization of holes is attributable to finite hole hopping among MM, MX, and XM sites, together with screening of the moiré potential.

Having identified the ground-state properties arising from an interplay between Coulomb interactions and interlayer coherent hole tunneling using optical excitations as a spectroscopic tool, we address the Feshbach physics that emerges in the excited state. The relevant scattering process is illustrated in Fig. 4A. By changing the electric field, the potential energy difference of holes in the top and bottom layers can be adjusted. This in turn allows us to adjust the interlayer and intralayer exciton-hole potentials with respect to one another, as depicted schematically in Fig. 4B. We show a wide range of the $V_E$-dependent $\Delta R/R_0$ spectra at $\nu = 3$, covering both AP and RP resonances in Fig. 4C. For concreteness, we focus on the $APH_{\text{top}}$ branch that is associated with holes originally at the MM sites of the moiré lattice: As $V_E$ is increased, this branch continues to blue-shift while losing oscillator strength. The latter feature is a consequence of the fact that as $V_E$ is increased, the probability of finding a lowest-energy MM site hole in the top layer is decreased: Because the hole in the initial state of the optical transition is predominantly in the bottom layer, the photon energy required to create a top-layer AP increases linearly with $V_E$. Naturally, for large $V_E$, the probability of finding the hole in the
top-layer MM site is small, as evidenced by the very faint $A_{\text{top}}$ signal.

Remarkably, for $V_{\text{E}} \geq 1.3 \text{ V}$, the oscillator strength of the $V_{\text{E}}$-dependent top-layer AP transition starts to increase and exhibits a highly asymmetric anti-crossing with the top-layer exciton transition at $V_{\text{E}} = 1.4 \text{ V}$. The corresponding avoided crossing between the bottom-layer AP and the bottom-layer exciton transitions is observed at $V_{\text{E}} = -2.1 \text{ V}$. If this had been a simple zero-dimensional system, such as a quantum-dot molecule, we could have explained the avoided crossing as stemming from hybridization of a top-layer trion with a state consisting of a top-layer exciton and a bottom-layer hole ($25, 26$). The experiments depicted in Fig. 4C, by contrast, are carried out in a 2D system with a layer pseudospin degree of freedom ($27, 28$): The resonance responsible for the avoided crossing is between a bound molecular state (trion) and a continuum of interlayer exciton-hole states and is referred to as a Feshbach molecule ($4$). (i) The observed hole density dependence of the avoided crossing can be explained in terms of RP and AP formation associated with this interlayer molecular state ($29, 30$).

Figure 4D shows the derivative of $\Delta R/R_0$ with respect to energy $E$ in the spectral region highlighted by the green dashed box in (C) for $\nu = 3, \nu = 2.5,$ and $\nu = 2$, respectively. The spectrum around the $X_{\text{top}}$ resonance is visibly asymmetric and depends on the density of holes in the opposite (bottom) layer.

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Fig. 4. Feshbach resonance. (A) Illustration of resonantly enhanced scattering of an exciton and a hole in a bilayer TMD. The Feshbach resonance is achieved by tuning $E_z$ and setting the electrostatic potential difference between the layers to match the intralayer exciton-hole bound-state (trion) energy $E_T$. (B) Schematic depiction of the potential energy of an exciton and a hole in the open channel (red) and the closed channel (blue). (C) $V_{\text{E}}$-dependent differential reflectance spectra at a fixed chemical potential for $\nu = 3$. (D to F) $V_{\text{E}}$-dependent differential reflectance spectrum differentiated with respect to energy $E$, in the area marked with dashed green box in (C) for $\nu = 3, \nu = 2.5,$ and $\nu = 2$, respectively. The spectrum around the $X_{\text{top}}$ resonance is visibly asymmetric and depends on the density of holes in the opposite (bottom) layer.
Our observation of strongly asymmetric splitting between the AP and RP resonances as the system is tuned across a Feshbach resonance is typical for scattering processes with negligible finite-range corrections (4). These features are in contrast to experiments on polariton Feshbach resonances, where signatures of the characteristic scattering physics were not conclusively demonstrated (2D); there, the cavity-like dispersion of the polaritons was much smaller than the cavity-like dispersion of the polaritons. Furthermore, the reported experiments establish that the combination of applied electric field, interlayer hole tunneling, and layer-selective optical excitations can in principle allow for arbitrary rotation of the layer pseudospin on the Bloch sphere together with projective measurements in the up-down basis. The degree of achievable control could enable a new set of quantum optics experiments in 2D materials, including optical pumping of valley and layer pseudospin and electromagnetically induced transparency exploiting layer coherence.

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