Tunable Feshbach Resonances and Their Spectral Signatures in Bilayer Semiconductors

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Feshbach resonances provide an invaluable tool in atomic physics, enabling precise control of interactions and the preparation of complex quantum phases of matter. Here, we theoretically analyze a solid-state analog of a Feshbach resonance in two dimensional semiconductor heterostructures. In the presence of interlayer electron tunneling, the scattering of excitons and electrons occupying different layers can be resonantly enhanced by tuning an applied electric field. The emergence of an interlayer Feshbach molecule modifies the optical excitation spectrum, and can be understood in terms of Fermi polaron formation. We discuss potential implications for the realization of correlated Bose-Fermi mixtures in bilayer semiconductors.

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Recently, bilayer structures of two-dimensional materials have emerged as fascinating platforms for realizing exotic phases of electronic matter [1,2]. Much of their success is driven by a new level of control, arising from twisting the two layers with respect to each other during stacking. Such twisted bilayers generate a moiré potential for electrons or holes, which quenches the kinetic energy and therefore enhances correlations. Most notably this has lead to the discovery of unconventional superconductivity [3,4], correlated insulators, and generalized Wigner crystals [5–7] in bilayer graphene and transition metal dichalcogenides (TMDs). In addition to electronic phases, semiconductors such as TMDs can host excitons, which are strongly bound electron-hole pairs. They act as mobile composite bosons and remain rigid due to their large binding energies. Moreover, excitons interact with free electrons or holes and can form charged molecules, termed trions. This renders bilayer TMDs promising candidates to study complex Bose-Fermi mixtures. Such mixtures have been recently investigated in dilute quantum gases [8–10], where Feshbach resonances are routinely used to control interactions between the atomic species [11–15]. By contrast, in solid state structures the molecular binding energies, and correspondingly the interaction strength among particles, are generically fixed by material properties, limiting the experimentally accessible regimes.

Here, we address this challenge by introducing a solid-state analog of a Feshbach resonance. Using the layer degree of freedom as a pseudospin, we demonstrate that the energy of a closed-channel bound state can be tuned with respect to scattering states in an open channel, simply by applying an external electric field E_z. The counterpart of hyperfine interactions in atomic systems, is provided by coherent interlayer electron or hole tunneling. The emerging Feshbach molecule controls the interlayer scattering and originates from the hybridization of exciton-electron scattering states with the intralayer (closed channel) trion state [16]. As such, it is fundamentally distinct from the formation of interlayer trions due to interactions determined by the material properties that are not tunable [17]. We demonstrate the impact of such Feshbach resonances on the spectrum of a single optically injected exciton immersed in a Fermi sea of charge carriers, taking into account the radiative exciton decay. Close to the Feshbach resonance we find a striking modification of the exciton spectrum. In particular, we show that the spectral shape is sensitive to the finite range of the effective interactions relative to the Fermi wavelength.

Our Letter is motivated by a recent experimental observation of an electrically tunable Feshbach resonance in a twisted bilayer TMD heterostructure [18]. We theoretically analyze a more generic scenario with vanishing twist angles and discuss how resonantly enhanced polaron formation can be observed in reflection measurements. Our findings demonstrate the potential for bilayer TMDs to control valley-selective interactions between itinerant carriers and establish a novel platform for exploring correlated quantum dynamics of degenerate Bose-Fermi mixtures.

Effective bilayer Hamiltonian.—We consider a bilayer semiconductor setup as depicted in Fig. 1. As we are

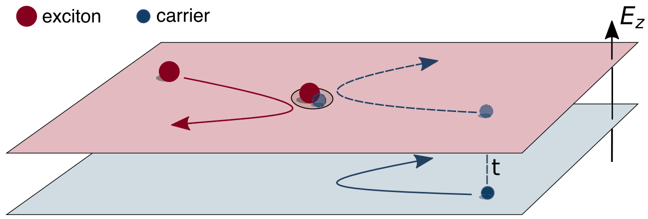

FIG. 1. Tunable Feshbach resonances in bilayer heterostructures. Illustration of exciton-carrier scattering in a bilayer TMD. The electrostatic potential energy is different in the two layers and can be tuned by a perpendicular electric field E_z. Scattering between excitons and electrons is enhanced when the intralayer trion energy is tuned into resonance with the energy of an electron and an exciton in separate layers.

interested in low-energy scattering, details of the underlying atomic lattice are irrelevant due to the large separation of scales between the lattice momentum and the momenta of excitons and electrons. In this regime excitons and electrons have essentially parabolic dispersions. Tunneling of electrons (or holes) between the two layers can be described by an effective average coupling constant t, which can be adjusted by incorporating tunnel barriers [7,19]. For concreteness we focus on two identical TMD layers separated by a distance d. Generically, the exciton resonances in the top and bottom layers have different energies, either due to the difference in material properties or strain, enabling layer-selective exciton creation. Furthermore, for electric fields close to the Feshbach resonance, the hybridization of inter- and intralayer excitons is small due to their sizable energy difference. This allows us to focus only on intralayer excitons [20]. For simplicity we assume that excitons are injected optically and are present only in the top layer. The system is then described by the effective Hamiltonian

$$\hat{H} = \sum_{\mathbf{k}} x_{\mathbf{k}}^{\dagger} \frac{k^2}{2M} x_{\mathbf{k}} + \begin{pmatrix} c_{\mathbf{k},T}^{\dagger} \\ c_{\mathbf{k},B}^{\dagger} \end{pmatrix} \begin{pmatrix} \xi_{\mathbf{k}} + \Delta & t \\ t & \xi_{\mathbf{k}} \end{pmatrix} \begin{pmatrix} c_{\mathbf{k},T} \\ c_{\mathbf{k},B} \end{pmatrix} + \frac{U}{V} \sum_{kk'q} c_{\mathbf{k},T}^{\dagger} c_{\mathbf{k}+\mathbf{q},T} x_{\mathbf{k}'}^{\dagger} x_{\mathbf{k}'-\mathbf{q}}, \tag{1}$$

where $x_{\mathbf{k}}^{\dagger}$ creates an exciton of mass M in the top layer, and $c_{\mathbf{k},T}^{\dagger}$ and $c_{\mathbf{k},B}^{\dagger}$ create fermions of mass m in the top and bottom layer, respectively.

From now on we refer to itinerant charges as electrons, although all conclusions apply equally to holes. We omit the valley and spin degree of freedom and assume that electrons and excitons reside in different valleys, since only this scattering channel will be resonantly enhanced. As the exciton's Bohr radius is small, excitons and electrons experience sizable attractive contact interactions U, only when both particles are in the same layer and opposite valleys. We also neglect the composite nature of the exciton and treat it as a structureless boson [21]. The potential energy difference $\Delta = qdE_z$ between the two layers, can be tuned by changing E_z, as illustrated in Fig. 1. We consider the scenario where Δ is chosen such that electrons reside predominantly in the bottom layer.

Feshbach resonance in exciton-electron scattering.—To understand scattering properties in such a heterostructure, we focus on the two-particle subspace of the system. In the center of mass frame, Eq. (1) can then be expressed in first quantization as

$$\hat{H}_{2\,\text{body}} = \hat{H}_0 + \hat{U} = \begin{pmatrix} -\frac{\nabla_{\mathbf{R}}^2}{2m_{\text{tot}}} - \frac{\nabla_{\mathbf{r}}^2}{2\mu} + \Delta & t \\ t & -\frac{\nabla_{\mathbf{R}}^2}{2m_{\text{tot}}} - \frac{\nabla_{\mathbf{r}}^2}{2\mu} \end{pmatrix} + U \begin{pmatrix} \delta^2(\mathbf{r}) & 0 \\ 0 & 0 \end{pmatrix}, \tag{2}$$

where $\mu = 1/(m^{-1} + M^{-1})$ and $m_{\text{tot}} = m + M$ are the reduced mass and the total mass, respectively. The wave function carries the layer degree of freedom and the part describing the relative motion can be expressed as $\psi(\mathbf{r}) = [\psi_T(\mathbf{r}), \psi_B(\mathbf{r})]^T/\sqrt{2}$. Asymptotic eigenstates with large spatial separation between the two particles define the open and closed channel. We consider E_z for which $\Delta \simeq |E_B^0|$, where E_B^0 is the binding energy of the intralayer trion. Although both channels are hybridized between the layers, only the open channel is energetically accessible and electrons reside predominantly in the bottom layer (Fig. 2). The scattering threshold for the open (ε_O) and closed (ε_C) channel is $\varepsilon_{O,C} = \Delta/2 \mp \sqrt{t^2 + \Delta^2/4}$.

The outgoing scattering states $|\psi_\alpha^+\rangle$, in channel α with energy E can be found as solutions of the Lippmann-Schwinger equation:

$$|\psi_\alpha^+\rangle = |\phi_\alpha\rangle + \frac{1}{E - \hat{H}_0 + i0^+} \hat{U} |\psi_\alpha^+\rangle, \tag{3}$$

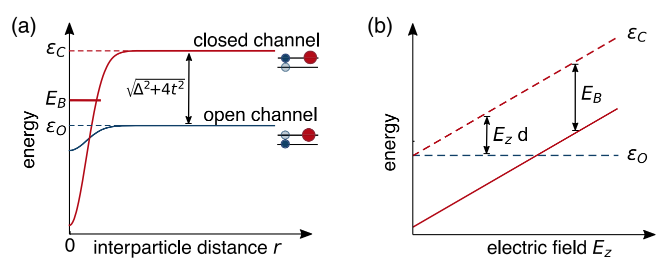

FIG. 2. Illustration of scattering channels. (a) Interparticle potential for an exciton and an electron prepared in the open (blue) or closed (red) channel. Tunnel coupling imprints the closed channel attraction also on the open channel. (b) Threshold energies of the open and closed channel ε_O and ε_C, as the electric field is varied. The bare closed channel bound-state energy is denoted as a red line. This bound state can be brought into resonance with ε_O for an appropriately chosen electric field.

where $\langle \mathbf{r}|\phi_\alpha\rangle \sim e^{ikx}$ is an incoming plane wave [15,22,23]. We can reformulate the problem by introducing the T matrix $\hat{T}^R|\phi_\alpha\rangle = \hat{U}|\psi_\alpha^+\rangle$, which connects the incoming plane waves with the full outgoing scattering state. Equation (3) translates to an equation for the off-shell T matrix $\hat{T}^R(E)$:

$$\hat{T}^R(E) = \hat{U} + \hat{U}(E - \hat{H}_0 + i0^+)^{-1}\hat{T}^R(E). \tag{4}$$

We solve Eq. (4) analytically in a plane-wave basis which diagonalizes $\hat{H}_0$:

$$\hat{T}^R(E,\mathbf{k}) = [\mathbb{1}_{2\times 2} - \hat{U}\cdot\Pi^R(E,\mathbf{k})]^{-1}\cdot\hat{U},$$
$$\Pi^R_{\alpha\beta}(E,\mathbf{k}) = \int \frac{d^2q}{(2\pi)^2}\frac{\delta_{\alpha\beta}}{E - \frac{\mathbf{q}^2}{2\mu} - \frac{\mathbf{k}^2}{2m_{\mathrm{tot}}} - \varepsilon_\alpha + i0^+}, \tag{5}$$

where E is the scattering energy, and $\mathbf{k}$ is the total incoming momentum. The 2×2 matrix structure of $\hat{T}^R(E,\mathbf{k})$, and $\hat{U}$, due to the two channels, is implicitly assumed.

Scattering can be resonantly enhanced if E_z is tuned such that the closed channel bound state is in proximity of the open channel threshold ε_O, see Fig. 2(b) for an illustration. Similar to cold atomic systems, we are interested in two-particle collisions with small incoming momenta. In this case, scattering is accurately described by a finite-range expansion, which is performed by expanding the denominator of the T matrix in powers of $E - \varepsilon_O$. In two dimensions the finite range expansion of the on-shell T matrix takes the universal form

$$T^R(\mathbf{q}^2/2\mu, \mathbf{0})^{-1} = \frac{\mu}{2\pi}\left(i\pi - \ln(\mathbf{q}^2a^2) + \frac{r_0\mathbf{q}^2}{2} + \mathcal{O}(\mathbf{q}^3)\right), \tag{6}$$

which is characterized by the scattering length a and effective range r_0 [24,25]. We relate this expansion to our effective description by integrating Eq. (5) and matching the open channel scattering amplitude $T^R_{OO}(\mathbf{q}^2/2\mu,\mathbf{0})$ to Eq. (6). In this way we obtain the open channel scattering length a_O and effective range r_0:

$$a_O = a\exp\left\{-\frac{1}{2}\left(\frac{\Delta}{2t} + \sqrt{1+\frac{\Delta^2}{4t^2}}\right)^2 \ln\frac{-E_B^0}{\sqrt{4t^2+\Delta^2}}\right\},$$
$$r_0 = \frac{1}{2\mu}\frac{(\Delta/2 + \sqrt{t^2+\Delta^2/4})^2}{t^2\sqrt{t^2+\Delta^2/4}}, \tag{7}$$

where $a = 1/\sqrt{2\mu E_B^0}$ is the scattering length of the closed channel in the absence of tunnel coupling. Analyzing Eqs. (5)–(7), we find that the open channel T matrix has a pole at energies below the scattering threshold ε_O. This is the signature of a Feshbach molecule which forms in interlayer scattering [23]. Equation (6) demonstrates that the energy of the molecule depends on both the scattering length a_O and range r_0. We plot the energy of the Feshbach molecule as a function of detuning in Fig. 3 for three different t. As the detuning becomes large and positive, the scattering length starts to diverge while the molecular energy approaches the scattering threshold. For large detunings the binding energy is then approximately given by $1/2\mu a_O^2$.In the case $\Delta > E_B^0 \gg t$ we obtain simple expressions for the binding energy of the Feshbach molecule E_B and the effective range close to resonance, which reads

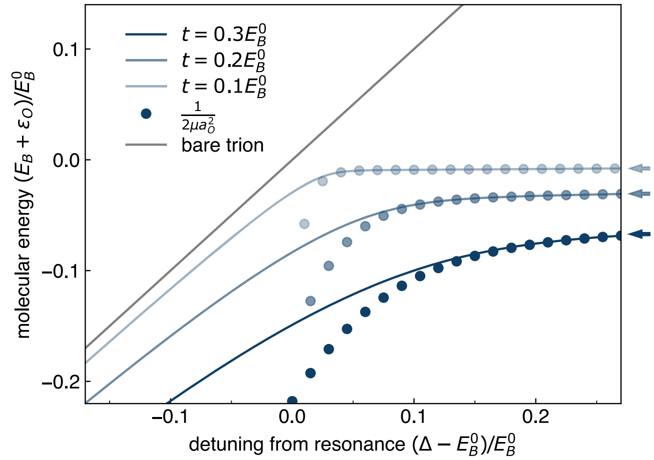

FIG. 3. Feshbach molecule binding energy. Molecular energy as a function of electric field (solid blue lines). We have assumed an exciton mass of $M = 2m$ and contact interactions between the exciton and electron. In two dimensions, and in the absence of interlayer repulsion, a bound state exists for all values of Δ. When the size of the molecule exceeds the range of the interactions, the scattering length alone determines the binding energy (blue dots). For large positive detunings the molecular energy approaches the open channel threshold ε_O (arrows), implying that the binding energy E_B goes asymptotically to zero. For large negative detunings the binding energy approaches the energy of the bare intralayer trion.

$$E_B \simeq E_B^0\frac{1}{e^{-2}}\left|\frac{\Delta}{E_B^0}\right|^{-\Delta^2/t^2}, \qquad r_0 \simeq \frac{1}{\mu}\frac{\Delta}{t^2}. \tag{8}$$

This demonstrates the power of a Feshbach resonance: complete control over the energy of the Feshbach molecule can be achieved simply by changing E_z. Thus the system can be electrically tuned to arbitrarily large scattering lengths [26,27]. While the binding energy of the Feshbach molecule changes exponentially, the effective range r_0 depends only linearly on E_z. We find that weakly coupled layers lead to large values of r_0, and the resulting physics is reminiscent of narrow Feshbach resonances in three dimensions.

In contrast to the three dimensional case, however, the bound state does not dissolve for any value of E_z, as long as there is no repulsive background scattering [28]. The purely two dimensional geometry of the system also distinguishes the proposed resonance from realizations in cold atom systems, where scattering remains effectively three dimensional due to the finite transverse confinement [14].

Optical impurities strongly coupled to a Fermi sea.—Resonantly enhanced two-particle scattering affects correlations in electron-exciton mixtures. We consider a low concentration of excitons injected into a Fermi sea of electrons in the open channel. The excitons in such a system are mobile impurities and form collective excitations known as Fermi polarons [30–34]. Here, we analyze the polaron spectrum as E_z is tuned over the Feshbach resonance.

Our previous discussions focused on two-body scattering with small but finite momentum, for which the exciton is long-lived and the scattering matrix is essentially unitary [35]. Here, we focus on optically excited $\mathbf{k}=0$ excitons. In this regime excitons couple to the radiation field, which allows them to decay via electron-hole recombination via the emission of an optical photon. As this decay process is essentially memoryless, it can be described by a Lindblad master equation

$$\dot{\rho}(t) = -i[\hat{H},\rho] + \sum_{\mathbf{k}} L_{\mathbf{k}}\rho L_{\mathbf{k}}^{\dagger} - \frac{1}{2}\{L_{\mathbf{k}}^{\dagger}L_{\mathbf{k}},\rho\},$$
$$L_{\mathbf{k}} = \sqrt{2\Gamma(\mathbf{k})}x_{\mathbf{k}}, \tag{9}$$

where $\Gamma(\mathbf{k})$ is the decay rate of the exciton, which we approximate to be finite only for $\mathbf{k}=0$, due to the steep light cone of the photons. In the presence of a Fermi sea Eq. (9) constitutes a complex many-body system, which can not be solved exactly. However, it was found that key properties can already be inferred purely from the scattering properties of the system [36,37] and that T-matrix approximations provide an accurate description of the ground and excited states of mobile impurities [25,38–45].

For our heterostructure setting we develop a T-matrix approximation to include dissipation as well as finite-range corrections from the Feshbach resonance:

$$\hat{T}^R(E,\mathbf{k}) = [\mathbb{1}_{2\times 2} - \hat{U}\cdot\Pi^R(E,\mathbf{k})]^{-1}\cdot\hat{U}$$
$$\Pi^R_{\alpha\beta}(E,\mathbf{k}) = \int_{|\mathbf{q}'|>k_F}\frac{d^2q'}{(2\pi)^2}\frac{\delta_{\alpha\beta}}{E-\xi_{\mathbf{q}'}-\varepsilon_\alpha-\frac{(\mathbf{k}-\mathbf{q}')^2}{2M}+i\Gamma(\mathbf{k}-\mathbf{q}')}. \tag{10}$$

Details on the calculation can be found in the Supplemental Material [28]. Compared to Eq. (5), the momentum of the electron in the open channel is now restricted to lie above the Fermi surface due to Pauli blocking by the Fermi sea. Exciton recombination results in an imaginary part $i\Gamma(\mathbf{k})$ of the exciton energy [46,47]. Using this T matrix, we then determine the self-energy of the exciton as a function of frequency ω:

$$\Sigma^R(\omega,\mathbf{k}) = \int_{|\mathbf{q}|<k_F}\frac{d^2q}{(2\pi)^2}T^R_{OO}(\omega+\xi_{\mathbf{q}},\mathbf{k}+\mathbf{q}). \tag{11}$$

This equation originates from the creation of a particle-hole pair in the open channel, with hole momentum $\mathbf{q}<k_F$. The spectral function of the exciton then reads

$$A_x(\omega,\mathbf{k}) = -2\mathrm{Im}\left[\frac{1}{\omega-\mathbf{k}^2/2M-\Sigma^R(\omega,\mathbf{k})+i\Gamma(\mathbf{k})}\right]. \tag{12}$$

As the master equation fulfills fluctuation-dissipation relations and we have treated dissipation exactly, the resulting spectral function respects the sum rule $\int(d\omega/2\pi)A_x(\omega,\mathbf{k}) = \langle[x_{\mathbf{k}},x_{\mathbf{k}}^{\dagger}]\rangle = 1$.

We compute the spectrum as a function of detuning, by integrating Eq. (12) numerically. We show the resulting

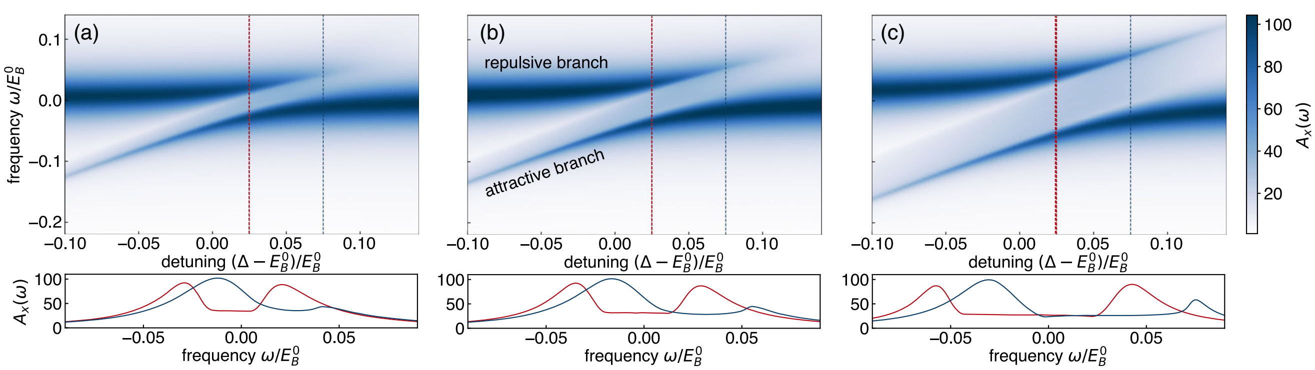

FIG. 4. Exciton spectra across the Feshbach resonance. The zero-momentum spectral function $A_x(\omega)$ of a dissipative exciton as a function of the bias Δ, computed within a T-matrix approximation. The Fermi energy E_F is increasing from left to right: (a) $E_F = E_B^0/30$, (b) $E_F = E_B^0/20$, (c) $E_F = E_B^0/10$. All spectra are computed for weak channel coupling $t = 0.15E_B^0$. The splitting of the repulsive and attractive branch depends on E_F, as highlighted in the line cuts of the spectra for two different Δ in the lower panels. For large E_F, finite range corrections become increasingly important and the repulsive branch is stabilized and regains oscillator strength. Motivated by recent experiments, the exciton is assumed to have a radiative lifetime of $\Gamma = E_B^0/30$ [49,50].

exciton spectra in Fig. 4 for three different Fermi energies (a)–(c). They are characterized by the formation of an attractive branch, with maxima at negative frequencies; and a repulsive branch, with maxima at positive frequencies. For small Fermi energies [Fig. 4(a)] the two resonances approach the Feshbach molecule and bare exciton energy, respectively: the spectrum can be understood in terms of the formation of a Fermi polaron and is highly asymmetric. We observe that the repulsive polaron abruptly transfers spectral weight to the attractive branch as the Feshbach molecule becomes weakly bound and blueshifts in energy.

With increasing carrier density, the maximal splitting between the repulsive and attractive branch grows [Fig. 4(b)]. Surprisingly, we find that the repulsive polaron branch is stabilized with growing electron densities, as seen in Fig. 4(c), despite the possible relaxation channel via excitations in the Fermi sea. This change in spectral shape cannot be explained assuming contact interactions, but rather arises from significant finite range corrections [48,51]. Since the average scattering process involves momenta on the order of k_F, the nonlogarithmic terms in Eq. (6) become successively more important at high densities and strongly renormalize the spectrum. In our setup Feshbach resonances are rather broad, which leads to characteristic spectral asymmetries due to the strong coupling to a continuum of scattering states. For Feshbach resonances based on polaritons on the other hand, this coupling is typically very weak due to the steep polariton dispersion, which can obscure the relevant scattering physics [52]. As the spectral function of the exciton is directly accessible in reflection measurements, the features we identified provide particularly clear experimental signatures, which result from many-body effects.

Conclusions and outlook.—We have investigated an electrically tunable solid-state Feshbach resonance, using the layer pseudospin degree of freedom of semiconductor bilayers. Our scheme allows for a controlled enhancement of electron-exciton scattering in experiments. We find that much of the resulting Feshbach physics, such as Fermi-Polaron formation for a dilute concentration of excitons, may be observed in experiments as a highly asymmetric and density-dependent reflection spectrum. This makes TMD bilayers ideal systems to study two dimensional Fermi polarons in parameter regimes that have so far been inaccessible in cold atomic gases and monolayer semiconductors.

By extending our setup to finite exciton densities, Feshbach resonances could enable precise control of degenerate Bose-Fermi mixtures in solid state systems. This is particularly appealing as excitations of an excitonic Bose gas can mediate superconductivity in a Fermi sea [53,54]. Since the bound state exists only for excitons and electrons with a different spin-valley degree of freedom, the Feshbach resonance could allow for spin selective interaction control and may induce instabilities in exotic pairing channels.

Feshbach resonances can also form in different scattering channels than the one considered here, i.e., an electron and an interlayer exciton in resonance with an intralayer bound state, which could prove to be useful in the context of long-lived indirect exciton condensates [55]. This is possible as interlayer and intralayer excitons have been shown to hybridize due to tunneling, and the energy of the former can be tuned electrically [7,56]. We further remark that while we discuss structures consisting of two identical TMD layers, the emerging Feshbach physics is universal. By choosing different TMDs and spacer materials one can vary the tunnel coupling and therefore the resonance width r_0.

Furthermore, our results generate the opportunity to study few-body physics in two dimensional semiconductors. The tunable scattering length can be used to explore exotic multiparticle bound states, where a single electron binds multiple excitons [57,58]. While we specifically considered resonant scattering between excitons and electrons, Feshbach physics in 2D materials could be a generic phenomenon that may also be relevant for understanding purely electronic processes [59].

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