

Purely electronic model for exciton-polaron formation in moiré heterostructures

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Understanding interactions between excitons and correlated electronic states presents a fundamental challenge in quantum many-body physics. Here, we introduce a purely electronic model for the formation of exciton polarons in moiré lattices. Unlike conventional approaches that treat excitons as tightly bound bosonic particles, our model considers only electronic degrees of freedom, describing excitons as electron-hole bound states. Our findings reveal a pronounced renormalization of the polaron mass as a function of electron density, particularly near correlated insulators, consistent with recent transport experiments. Additionally, we predict an observable sign change in the effective polaron mass when increasing the electron density that can be measured in Hall-type experiments. Our purely electronic model provides a unified framework to investigate the formation and renormalization of exciton polarons in correlated states.

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I. INTRODUCTION

Excitons have proven to be a valuable tool for probing strongly correlated states in transition metal dichalcogenide (TMD) heterostructures, including periodic charge modulation [1–5], spin ordering [6,7], and dipolar excitonic insulators [8–10]. An increasing number of experiments focus on the physics of excitons in moiré heterostructures [11–21], with a particular focus on transport and diffusion experiments [22–27], yet our theoretical understanding of exciton dynamics in lattice systems and strongly correlated environments remains vastly open. When immersed in a strongly correlated bath, excitons become dressed by their environment, forming a quasiparticle known as polaron. While some recent studies have highlighted key aspects of exciton-polaron formation in moiré lattices [28–38] and other correlated systems [39–44], many open questions remain. In particular, the simplification of excitons as tightly bound bosonic particles without internal structure is insufficient to capture the behavior of interlayer excitons in TMD heterostructures [32]. Moreover, it remains unclear which role different stacking configurations have on the dressed exciton polaron. Developing effective theoretical models suited to describe the exciton-polaron dynamics in moiré lattices is therefore pertinent to help interpret already existing data and guide future experimental investigations.

In this work, we develop a simple yet powerful, purely electronic model to describe the dynamics of an interlayer

exciton in a moiré lattice. Our model only contains electronic degrees of freedom and fully accounts for the internal structure of the exciton as a bound state of an electron and hole. Employing the Chevy approximation [45], we calculate the exciton propagator in the presence of a Fermi liquid and correlated insulators. At low electron densities, the model reproduces the characteristic formation of attractive polaron (AP) and repulsive polaron (RP) branches, consistent with the continuum [46,47]. Beyond this, our model provides insight into the strong renormalization of exciton mobility as a function of electron filling, particularly near Mott insulating states, as observed in recent diffusion experiments [26,27]. We predict a sign reversal of the effective polaron mass at a critical filling and propose a Hall-type measurement for its experimental verification. Our purely electronic model, therefore, provides a unified framework for the exciton-polaron formation in correlated states that is beyond the reach of conventional Bose-Fermi models.

II. MODEL

Heterostructures of TMDs have been shown to host long-lived interlayer excitons where the electron and hole reside in different layers [11,48,49]. To describe the dressing of excitons in correlated environments, we propose a purely electronic model that incorporates the internal structure of an exciton as a bound state of an electron and a hole. Specifically, we consider a heterobilayer system in which, at charge neutrality, the moiré lattice of the bottom layer is fully filled while the top layer is empty [17,50,51]. In the deep-moiré regime, each layer is modeled as a triangular lattice, with holes $h^\dagger$ in the bottom layer and electrons $c^\dagger$ in the top layer. The Hamiltonian of the system is

$$H = H_e + H_h + H_{e-h}. \quad (1)$$

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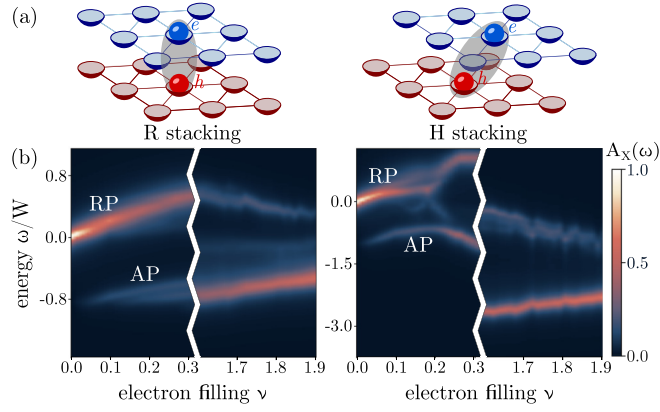


FIG. 1. Exciton polaron in low- and high-density regimes. (a) Schematics of interlayer-exciton formation in moiré lattices. The top layer is populated by electrons (blue sphere), and the bottom layer by holes (red sphere). For R stacking (left), the electron and hole sites lie on top of each other, while they are shifted for H stacking. (b) Zero-momentum exciton spectral function, showing the RP and AP, which appears at finite electron filling ν . The energy is given in units of the bare electron bandwidth $W = 9t$, with t the electron hopping. We use the same local electron-hole interactions ($U_{e-h} = -15t$) for both stackings; all other interactions are set to zero. System size $N = 12 \times 12$.

The electrons in the top layer are governed by

$$H_e = \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}}^e c_{\sigma\mathbf{k}}^\dagger c_{\sigma\mathbf{k}} + \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} V_{\mathbf{q}}^{e-e} c_{\sigma,\mathbf{k}+\mathbf{q}}^\dagger c_{\sigma',\mathbf{k}'-\mathbf{q}}^\dagger c_{\sigma'\mathbf{k}'} c_{\sigma\mathbf{k}}, \quad (2)$$

including a repulsive electron-electron interaction $V_{\mathbf{q}}^{e-e}$. We assume electron doping and work in the exciton impurity limit, corresponding to a low pump intensity in experiments (see, e.g., Ref. [26]). Thus, there is only a single hole in the system, such that for the hole we only need to consider the kinetic term:

$$H_h = \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}}^h h_{\sigma\mathbf{k}}^\dagger h_{\sigma\mathbf{k}}, \quad (3)$$

where $\varepsilon_{\mathbf{k}}^{e/h}$ is the dispersion of a particle hopping on a triangular lattice. We assume the nearest-neighbor hopping as $t_e = t_h \equiv t$. We also include attractive electron-hole interactions $V_{\mathbf{q}}$ between the two layers, responsible for the formation of interlayer excitons:

$$H_{e-h} = \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} \sum_{\sigma\sigma'} V_{\mathbf{q}} c_{\sigma,\mathbf{k}+\mathbf{q}}^\dagger h_{\sigma',\mathbf{k}'-\mathbf{q}}^\dagger h_{\sigma'\mathbf{k}'} c_{\sigma\mathbf{k}}. \quad (4)$$

In practice, we restrict all interactions to short-range on-site U , nearest-neighbor density-density V , and nearest-neighbor direct-exchange X interactions. Electron-electron interactions are repulsive $U_{e-e}, V_{e-e} > 0$, while electron-hole interactions are attractive $U_{e-h}, V_{e-h} < 0$. Our purely electronic model allows us to study the exciton formation in different stacking configurations of the heterobilayer. We consider both R and H stacking. In the former, the lattice sites for the electrons and holes are on top of each other, while in the latter, they are shifted [51] [see Fig. 1(a)]. In our model, the two distinct stacking configurations manifest in different interlayer interactions $V_{\mathbf{q}}$, given explicitly in Appendix B.

We work in the low-exciton-density regime, where a single exciton $x^\dagger$ is added to the electronic ground state. The exciton is treated as a composite particle. Its interlayer wavefunction for total momentum $\mathbf{p}$ is

$$|X_{\mathbf{p}}\rangle \equiv x_{\mathbf{p}}^\dagger |\text{GS}\rangle = \sum_{\mathbf{k}} \psi_{\mathbf{k}}(\mathbf{p}) c_{\uparrow\mathbf{p}-\mathbf{k}}^\dagger h_{\uparrow\mathbf{k}}^\dagger |\text{GS}\rangle. \quad (5)$$

We determine the wavefunction $\psi_{\mathbf{k}}(\mathbf{p})$ by solving the two-particle Schrödinger equation in the presence of the electron ground state $|\text{GS}\rangle$, which we for now choose to be a Fermi sea. We later relax this assumption, discussing correlated insulators as electronic ground states, which modifies the decomposition of the exciton (see Appendix D). The attractive electron-hole interaction is chosen to be sufficiently large, such that $\psi_{\mathbf{k}}(\mathbf{p})$ describes a well-defined, localized bound state. Our approach fully accounts for the composite nature of the exciton and relies exclusively on microscopic interactions without introducing an effective exciton-electron coupling. To study the exciton dynamics, we compute its propagator:

$$\mathcal{G}_X(\mathbf{k}, \omega) = \langle \text{GS} | x_{\mathbf{k}} \frac{1}{\omega + i\eta - H} x_{\mathbf{k}}^\dagger | \text{GS} \rangle, \quad (6)$$

by projecting the Hamiltonian [Eq. (1)] to a subspace that includes states with a single exciton as well as states containing a single exciton accompanied by a particle-hole excitation of the electronic ground state [45]. This procedure defines the following basis states:

$$|n=0\rangle = |X_{\mathbf{p}}\rangle \equiv x_{\mathbf{p}}^\dagger |\text{GS}\rangle, \quad (7a)$$

$$|n>0\rangle = |C_{\mathbf{k}\alpha\mathbf{q}\beta}^{\mathbf{p}}\rangle \equiv x_{\mathbf{p}+\mathbf{q}-\mathbf{k}}^\dagger c_{\alpha\mathbf{k}}^\dagger c_{\beta\mathbf{q}} |\text{GS}\rangle. \quad (7b)$$

We compute the matrix elements of the Hamiltonian in this basis (see Appendix F). This approximation [45] has proven to be a reliable framework for capturing the many-body physics of an impurity immersed in a bath [43,44,52].

III. LOW- AND HIGH-DENSITY REGIME

To connect our results with the established theory on exciton-polaron physics in the continuum [46,47], we first consider the low- and high-filling regimes of the electron band, close to $\nu = 0$ and $\nu = 2$, where ν corresponds to the average number of electrons per site. In this regime, the electronic ground state $|\text{GS}\rangle$ is a Fermi liquid, for which we treat the repulsive electron-electron interactions on a mean-field level, such that their only effect is to renormalize the electron dispersion $\varepsilon_{\mathbf{k}}^e$. Our model correctly captures the formation of an attractive and repulsive polaron upon electron doping, as seen in the zero-momentum exciton spectral function $\mathcal{A}_X(\omega) = -\frac{1}{\pi} \text{Im} \mathcal{G}_X(\mathbf{k} = 0, \omega)$ [Fig. 1(b)]. We found that including spin is essential for accurately describing the formation of the attractive polaron in the low-density regime. This is because the formation of the trion, a bound state of two electrons and a hole, requires the electrons to form a singlet. By increasing the electron density, the spectral weight shifts from the repulsive to the attractive polaron. Our purely electronic model enables the study of how the energies of the two resonances shift with electron filling as a function of the microscopic interactions. Generically, stronger repulsive electron-electron interactions cause a blue shift,

while enhanced attractive electron-hole interactions result in a red shift of the resonances. We also explore the impact of different stacking configurations in heterobilayers. For an H-stacked bilayer, the moiré sites of the electrons are shifted relative to the hole site. Consequently, the electron component of the exciton wavefunction is predominantly distributed over three sites, all equidistant from the hole site [51]. The more delocalized nature of the exciton wavefunction favors the formation of interlayer attractive polarons, as observed in photoluminescence experiments [17,26,50,51]. For similar attractive interactions, we find that both AP and RP resonances experience a stronger redshift as a function of filling for H stacking compared to R stacking, especially in the high-density regime [see Fig. 1(b)].

We give an order-of-magnitude estimate for the interaction strengths based on WSe₂/WS₂ in Appendix C. Due to screening, the actual interaction strength in experiments will depend sensitively on the filling. Such a quantitative computation is beyond the scope of our work. Instead, our qualitative predictions are valid over a wide parameter regime, as exemplified by parameter scans in Appendix C.

IV. CORRELATED INSULATORS

So far, we have assumed the charge carriers to be in a metallic state. However, in intermediate density regimes, correlated insulators are observed at commensurate fillings [1–3,7,53–55]. We first focus on the generalized Wigner crystal at $\nu = 2/3$. The exciton-polaron spectral function of spinless generalized Wigner crystals was discussed in Refs. [43,44] using conventional Bose-Fermi models. We treat the repulsive electron-electron interactions on a mean-field level, allowing for the spontaneous breaking of the discrete translation symmetry. Concretely, we increase the unit cell to include three sites, allowing for the expected spin and charge order at $\nu = 2/3$. After mean-field decoupling of the electron-electron interaction, the electron Hamiltonian [Eq. (2)] is

$$H_e = \sum_{\mathbf{k}} \sum_{\lambda=1}^6 \epsilon_{\mathbf{k}}^{\lambda} \gamma_{\lambda\mathbf{k}}^{\dagger} \gamma_{\lambda\mathbf{k}}. \quad (8)$$

The Hartree-Fock quasiparticles $\gamma_{\lambda\mathbf{k}}^{\dagger}$ with dispersion $\epsilon_{\mathbf{k}}^{\lambda}$ are obtained by self-consistently solving the electronic mean-field problem (see Appendix D). To define the momenta of the top and bottom layers with respect to the same translations, we also enlarge the unit cell for the holes. After expressing Eq. (7), in this basis, we determine the exciton wavefunction by solving the two-particle Schrödinger equation in the presence of the correlated-insulator ground state |GS⟩. The resulting wavefunction describes the long-lived, lowest-energy exciton state to which optically created excitons relax. However, our approach also allows us to target exciton bound states at higher energies.

For the generalized Wigner crystal at $\nu = 2/3$, both the RP and AP branches blueshift with increasing electron-electron interaction U , which scales with the charge gap [Fig. 2(a)]. As the charge gap increases and the crystal order becomes more established, the spectral weight of the AP increases [see Fig. 2(b)]. Hence, the AP spectral weight peaks when the electrons are fully localized.

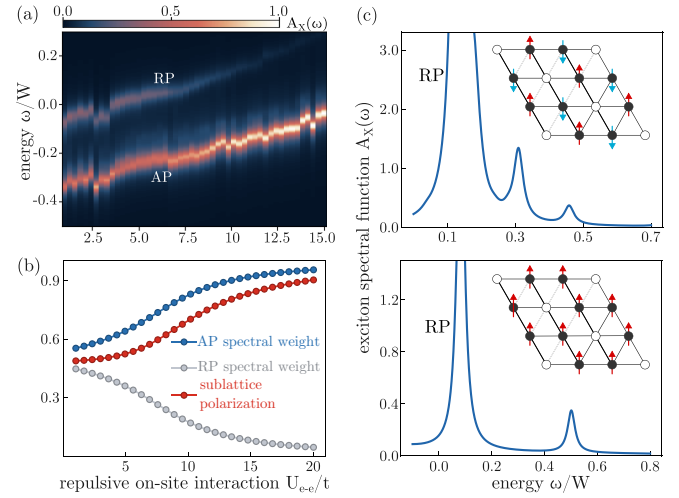


FIG. 2. Polaron formation near correlated insulators. (a) Zero-momentum exciton spectral function for the $\nu = 2/3$ generalized Wigner crystal ($U_{e-h} = -35t$, $V_{e-e}/U_{e-e} = 1/6$, R stacking). (b) The spectral weight of the AP increases with sublattice polarization $\Delta n = (n_A + n_B - n_C)/2$, an order parameter for the crystal. Here, $n_{A/B}$ is the average occupation of sublattice A/B, forming the crystal, and n_C is the occupation of the third sublattice, which is empty for an ideal crystal. (c) We observe two Umklapp peaks for the antiferromagnet, while there is only one Umklapp peak for the ferromagnet ($U_{e-e} = 12t$, the ferromagnet is stabilized by a direct exchange $X_{e-e} = 0.8t$).

Optical measurements provide the exciton spectral function at zero momentum, as momentum transfer by optical light is negligible. States with a periodic charge distribution coupled to excitons, however, give rise to additional optically active Umklapp resonances [4,5], at which a finite momentum state at a reciprocal lattice vector is backfolded to zero quasi-momentum. Due to the valley-Zeeman effect, the Umklapp peaks were also predicted to exhibit distinct signatures for antiferromagnetic and ferromagnetic spin order, offering a potential method to differentiate between the two [56]. The spin order of experimentally observed generalized Wigner crystals is still unclear, as definitive experimental probes are lacking [57]. Both antiferromagnetic and ferromagnetic couplings are possible, depending on the strength of direct exchange interactions, which favor ferromagnetism [58].

For the generalized Wigner crystal at $\nu = 2/3$, which forms a honeycomb charge order, we observe Umklapp peaks near the RP resonance. The number of Umklapp peaks depends on the symmetry of the state [56]. A generalized Wigner crystal with ferromagnetic spin order has C_6 symmetry, allowing only a single Umklapp peak. On the other hand, an antiferromagnet has a lower C_3 symmetry and more Umklapp peaks are allowed by symmetry. These symmetry arguments perfectly agree with our results, which consist of only a single peak for ferromagnetic spin order and further splitting of the peaks for antiferromagnetic spin order [see Fig. 2(c)]. We tune from an antiferromagnetic to a ferromagnetic crystal by introducing a finite nearest-neighbor direct exchange interaction (see Appendix D and Ref. [58]). Our model also predicts Umklapp peaks for the AP; however, with much weaker spectral weight than for the RP.

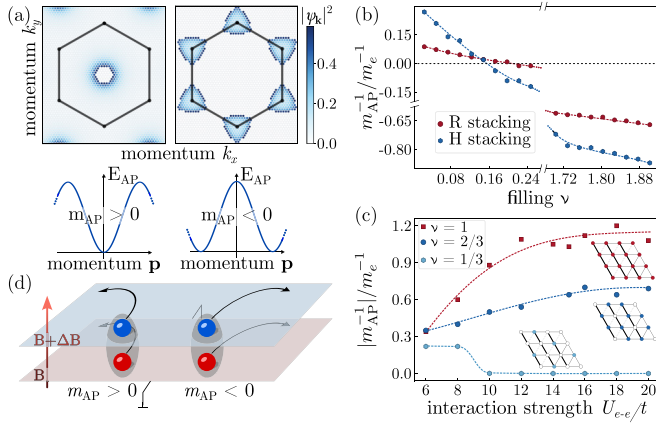


FIG. 3. Mass renormalization of attractive polaron. (a) Exciton wavefunction $\psi_{\mathbf{k}}(\mathbf{p}=0)$ at low (left) and high (right) densities ($\nu=0.06$ and $\nu=1.67$, respectively). In the high-density regime, the exciton wavefunction is localized near the band maxima at the Brillouin zone corners, leading to a negative exciton-polaron mass. (b) Inverse of the AP mass m_{AP}^{-1} relative to the electron mass $m_e^{-1}=3t$, in the low- and high-filling regime, with a Fermi sea as the ground state. (c) Inverse mass as a function of the interaction strength U for correlated insulators at $\nu=1$, $\nu=2/3$, and $\nu=1/3$ in R stacking. (d) The sign change of the polaron mass can be detected by a Hall-type measurement, where a magnetic field with an out-of-plane gradient is applied. The resulting Hall current is sensitive to the sign of the polaron mass. Details of the parameters can be found in Appendix E.

V. LARGE MASS RENORMALIZATION

Immersing an exciton in a correlated electronic bath can lead to a strong renormalization of its properties. We now theoretically investigate the mass renormalization of the AP as a function of the electron filling from the momentum-dependent exciton spectral function. We define the mass m of a particle with dispersion $\varepsilon_{\mathbf{k}}$ as $m^{-1} = \partial^2 \varepsilon_{\mathbf{k}} / \partial \mathbf{k}^2|_{\mathbf{k}=0}$. For a particle hopping on a triangular lattice, the mass is given by $m = 3/W$, where W is the bandwidth of $\varepsilon_{\mathbf{k}}$. We use this relation to determine the mass of the AP by calculating its bandwidth. We find a very strong mass renormalization of the polaron, consistent with recent diffusion experiments [26,27]. We emphasize that this mass renormalization for interlayer excitons in moiré lattices is orders of magnitude larger than the typical mass renormalization for polarons in the continuum [59–61]. In a system with continuous Galilean invariance, the effective mass of a bound state is independent of its binding energy. In contrast, in a lattice, where center-of-mass motion does not decouple, the effective mass depends on the binding energy, and second-order perturbation theory predicts that the exciton mass is proportional to the binding energy.

At low electron densities, increasing the filling provides more electronic excitations for the exciton to bind with, reducing its mobility [Fig. 3(b)]. In the high-density regime where the electron band is nearly full, available excitations decrease again, leading to weaker mass renormalization. Additionally, at sufficiently high densities, hopping channels emerge, allowing the hole to hop independently and rebind with a distant electron, enhancing mobility, referred to as nonmonogamous

hopping in Ref. [26]. This trend resembles exciton mobility measured in recent experiments [26,27].

The structure of the exciton wavefunction $\psi_{\mathbf{k}}(\mathbf{p})$ also greatly affects the exciton-polaron mobility, leading to a higher mobility in H-stacking than in R stacking due to the less-localized exciton wavefunction in H stacking [see Fig. 3(b)]. Relatively, the difference in mobility between R and H stacking is more substantial in the low-density regime. Another striking effect stemming from the nontrivial structure of the exciton wavefunction is a sign change in the effective polaron mass at a critical filling ν_0 . At low densities, the Fermi surface is close to the band minimum with positive curvature, resulting in a positive exciton mass. At high densities, however, available states for exciton formation lie near the band maxima at the Brillouin zone corners [see Fig. 3(a)]. There, the negative band curvature leads to a negative exciton mass. The critical density ν_0 at which this curvature effect becomes significant is quite low, with the precise value depending on the microscopic interactions. We propose to detect this sign change in the effective polaron mass in a Hall-type experiment. Since the charge carriers in the bottom layer are bound within excitons, the current in the bottom layer is directly proportional to the exciton-polaron transport. Applying a magnetic field with a spatial out-of-plane gradient induces a finite Lorentz force on the exciton, generating a measurable Hall current. Crucially, the Hall current is sensitive to the sign of the polaron mass (see, e.g., Ref. [62]), providing an experimental signature of the sign change [see Fig. 3(d)]. While a magnetic field gradient in the out-of-plane direction is necessary for the bare exciton to experience a finite Lorentz force, the AP is a charged quasiparticle and thus couples directly to the magnetic field and no gradient is needed.

We now study the mass renormalization in the vicinity of correlated insulators. Both for $\nu=2/3$ and $\nu=1$, we observe a very small effective mass, corresponding to a very large mobility [see Fig. 3(c)]. Assuming that the attractive electron-hole interaction dominates over the repulsive electron-electron interaction $|U_{e-h}| > U_{e-e}$, the exciton forms with an electron on top of the crystal, resulting in a weakly bound state. This results in a high mobility for $\nu=2/3$ and $\nu=1$, as the exciton hopping scales with the inverse of the binding energy. Since the exciton can only hop on top of the crystal, the mobility is smaller in the $\nu=2/3$ than the $\nu=1$ state, as there are fewer available hopping sites. Polaron mobility increases with repulsive electron-electron interaction until charge and spin order are well established; at this point, the mobility starts to saturate. In contrast, for $\nu=1/3$ generalized Wigner crystals, the exciton is effectively localized as there are no nearest-neighbor sites for it to hop to while remaining on the crystal.

Depending on the energy scales, for the generalized Wigner crystals, it is possible to obtain excitons that occupy an empty site. Such a configuration is favored for weaker on-site $|U_{e-h}| \lesssim U_{e-e}$ and stronger long-range V_{e-h} attractive interactions. This enhances mobility for the $\nu=1/3$ state but drastically reduces it for the $\nu=2/3$ state, demonstrating how different microscopic interactions can dramatically alter the exciton mobility near correlated insulators. Further details are provided in Appendix E.

We stress that having a purely electronic model is essential for capturing the strong mass renormalization for excitons in moiré lattices. Treating excitons as tightly bound bosons fails to account for the diverse processes arising from their nontrivial wavefunction. In contrast, our model [Eq. (1)] inherently incorporates these effects.

VI. CONCLUSION AND OUTLOOK

We have developed an effective, purely electronic model to describe exciton-polaron formation in moiré heterostructures. Our model highlights key mechanisms of stacking dependence and mass renormalization of the exciton polarons, consistent with recent experimental observations of giant exciton mobilities [26,27]. While our work was inspired by and is directly relevant to experiments in WSe₂/WS₂ heterobilayers, our model is not tailored to any specific material. Beyond TMDs, the model may also be relevant for exciton formation in other strongly correlated Mott insulators, such as Sr₂IrO₄ [63]. An exciting direction for future research is to extend our approach to explore polaron formation in slightly doped Mott insulators, where the exciton is dressed by collective spin modes. Furthermore, our framework could be applied to other exotic states, such as fractional quantum anomalous Hall states [64–67] or anomalous Hall crystals [68,69], using a parton mean-field state as the electronic ground state.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [70], embargo periods may apply.

APPENDIX A: EXCITON WAVEFUNCTION

Before tackling the many-body problem of inserting a single exciton into some electronic ground state $|\text{GS}\rangle$, we solve the two-body problem of an electron-hole pair. We define the exciton wavefunction with total momentum $\mathbf{p}$ as

$$|X_{\mathbf{p}}\rangle = x_{\mathbf{p}}^{\dagger}|\text{GS}\rangle = \sum_{\mathbf{k}} \psi_{\mathbf{k}}(\mathbf{p}) c_{\uparrow\mathbf{p}-\mathbf{k}}^{\dagger} h_{\uparrow\mathbf{k}}^{\dagger} |\text{GS}\rangle, \quad (\text{A1})$$

where the ground state

$$|\text{GS}\rangle = \prod_{|\mathbf{k}| < k_F, \sigma} c_{\sigma\mathbf{k}}^{\dagger} |0\rangle \quad (\text{A2})$$

is assumed to be a Fermi sea with the Fermi momentum k_F fixing the filling ν . Here, $|0\rangle$ is the vacuum state, corresponding to charge neutrality. Note that we chose to fix the spin of the electron and hole forming the exciton when working with a Fermi sea as the electronic ground state. We derive an equation for $\psi_{\mathbf{k}}(\mathbf{p})$ in Eq. (A1), by projecting the Schrödinger equation $H|X_{\mathbf{p}}\rangle = E_{\mathbf{p}}^X |X_{\mathbf{p}}\rangle$ onto $\langle 0|h_{\mathbf{k}}c_{\mathbf{p}-\mathbf{k}}$, leading to

$$(\varepsilon_{\mathbf{p}-\mathbf{k}}^e + \varepsilon_{\mathbf{k}}^h + \nu V_{\mathbf{q}=0}) \theta_{\uparrow\mathbf{p}-\mathbf{k}} \psi_{\mathbf{k}}(\mathbf{p}) + \sum_{\mathbf{k}'} V_{\mathbf{k}'-\mathbf{k}} \theta_{\uparrow\mathbf{p}-\mathbf{k}} \theta_{\uparrow\mathbf{p}-\mathbf{k}'} \psi_{\mathbf{k}'}(\mathbf{p}) = E_{\mathbf{p}}^X \psi_{\mathbf{k}}(\mathbf{p}), \quad (\text{A3})$$

with $\theta_{\sigma\mathbf{k}} = 1 - n_{\sigma\mathbf{k}} = 1 - \langle c_{\sigma\mathbf{k}}^{\dagger} c_{\sigma\mathbf{k}} \rangle$. We treat the repulsive electron-electron interaction purely on a mean-field level, renormalizing the electron dispersion $\varepsilon_{\mathbf{k}}^e$. The free electron and hole dispersions are taken as

$$\varepsilon_{\mathbf{k}}^{e/h} = -2t[\cos(\mathbf{k} \cdot \mathbf{a}_1) + \cos(\mathbf{k} \cdot \mathbf{a}_2) + \cos(\mathbf{k} \cdot \mathbf{a}_3)], \quad (\text{A4})$$

with lattice translation vectors $\mathbf{a}_{1/2} = (\pm\sqrt{3}/2, 1/2)^T$ and $\mathbf{a}_3 = (0, -1)^T$, corresponding to a particle hopping on a triangular lattice with hopping parameter t . We use the lowest-energy solution of Eq. (A3) to define the exciton operator $x_{\mathbf{p}}^{\dagger}$ through Eq. (A1). For R stacking, the exciton wavefunction is strongly localized in real space, while for H stacking, it is spread over three sites (see Fig. 4). At very high electron densities ν , most states are already occupied, such that $\psi_{\mathbf{k}}(\mathbf{p})$ is only nonzero for $\mathbf{k}$ close to the corners of the Brillouin zone, leading to a less localized exciton wavefunction in real space.

APPENDIX B: ATTRACTIVE ELECTRON-HOLE INTERACTIONS AND STACKING DEPENDENCE

The top layer hosting electrons and the bottom layer hosting holes are coupled through attractive electron-hole interactions [Eq. (4)], responsible for forming the exciton as an electron-hole bound state. We are working with on-site and nearest-neighbor density-density interactions:

$$H_{e-h} = U \sum_{i, \sigma, \sigma'} c_{\sigma i}^{\dagger} h_{\sigma' i}^{\dagger} h_{\sigma' i} c_{\sigma i} + V \sum_{\langle i, j \rangle} c_{\sigma i}^{\dagger} h_{\sigma' j}^{\dagger} h_{\sigma' j} c_{\sigma i}, \quad (\text{B1})$$

with $U, V < 0$. Note that for H stacking, the electron and hole sites are shifted with respect to each other, such that for each hole site, there are three equally distant electron sites, which we consider to be “local.” After taking a Fourier transform, we obtain

$$H_{e-h} = \sum_{\mathbf{k}\mathbf{k}'} \sum_{\sigma\sigma'} V_{\mathbf{q}} c_{\sigma, \mathbf{k}+\mathbf{q}}^{\dagger} h_{\sigma', \mathbf{k}'}^{\dagger} h_{\sigma', \mathbf{k}'} c_{\sigma, \mathbf{k}}, \quad (\text{B2})$$

where the shape of $V_{\mathbf{q}}$ depends on the stacking. For R stacking, one has

$$V_{\mathbf{q}} = \frac{U}{N} + \frac{V}{N} (e^{-i\mathbf{a}_1 \cdot \mathbf{q}} + e^{-i\mathbf{a}_2 \cdot \mathbf{q}} + e^{-i\mathbf{a}_3 \cdot \mathbf{q}}), \quad (\text{B3})$$

with lattice translation vectors $\mathbf{a}_{1/2} = (\pm\sqrt{3}/2, 1/2)^T$ and $\mathbf{a}_3 = (0, -1)^T$ and N is the number of unit cells. On the other

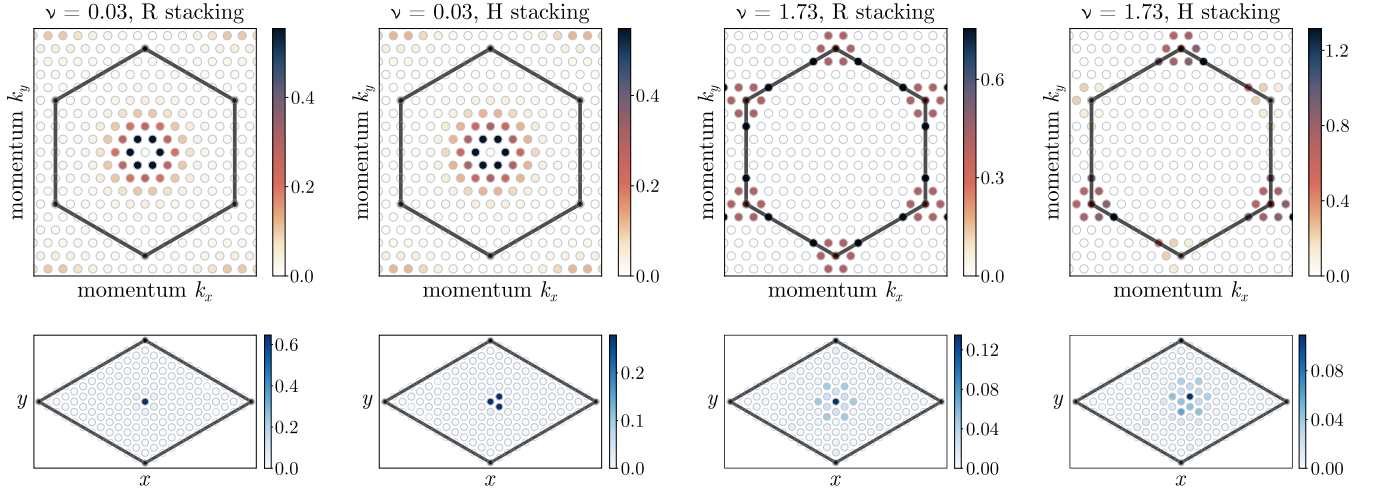


FIG. 4. Exciton wavefunction. Top: Zero total momentum exciton wavefunction $\psi_{\mathbf{k}}(\mathbf{p} = 0)$ for different stackings and electron densities ν . The boundaries of the Brillouin zone are highlighted. Bottom: Fourier transform of $\psi_{\mathbf{k}}(\mathbf{p} = 0)$, showing localization in real space. The exciton wavefunction is most strongly localized for R stacking at low densities. $U_{e-h} = -15t$ with all other interactions set to zero. System size $N = 12 \times 12$.

hand, for H stacking, we find

$$V_{\mathbf{q}} = \frac{U}{N} (1 + e^{-i\mathbf{a}_2 \cdot \mathbf{q}} + e^{+i\mathbf{a}_1 \cdot \mathbf{q}}) + \frac{V}{N} (e^{+i(\mathbf{a}_1 - \mathbf{a}_2) \cdot \mathbf{q}} + e^{-i\mathbf{a}_3 \cdot \mathbf{q}} + e^{+i\mathbf{a}_3 \cdot \mathbf{q}}). \quad (\text{B4})$$

APPENDIX C: ESTIMATE FOR THE INTERACTION STRENGTH

Our model and the predictions we derive from it are primarily qualitative, with the main goal being to provide a simple model capturing the essential mechanism underlying exciton-polaron formation in moiré heterostructures. Nevertheless, we connect to experiments by giving a rough estimate for the interaction parameters used in the main text. We assume a WSe_2/WS_2 heterostructure, as used in Ref. [26]. At zero twist angle, the moiré lattice constant is given by $a_M = a/\delta \sim 7$ nm, where we take $a = 0.3297$ nm as the atomic lattice constant for WSe_2 and δ is its lattice mismatch with WS_2 . Taking an effective electron mass $m^* = 0.5m_e$, we obtain a rough estimate for the hopping parameter of the electrons on a triangular lattice

$$t \sim \frac{\hbar^2}{3a_M^2 m^*} \sim 1 \text{ meV}. \quad (\text{C1})$$

We want to estimate the effective interaction strengths in the low-density regime, where we assume to have a Fermi liquid as the electronic ground state. In that case, the only effect of electron-electron interactions is to renormalize the bare parameters. We capture this by treating the electron-electron interactions on a mean-field level. Within this approximation, on-site interactions just introduce a density-dependent on-site potential

$$H_U = U_{e-e} \sum_i c_{\uparrow i}^\dagger c_{\downarrow i}^\dagger c_{\downarrow i} c_{\uparrow i} \approx U_{e-e} n_e \sum_{i,\sigma} c_{\sigma i}^\dagger c_{\sigma i}, \quad (\text{C2})$$

with $n_e = \langle c_{\sigma i}^\dagger c_{\sigma i} \rangle$ the average density. Similarly, nearest-neighbor density-density interactions result in an additional shift of the on-site potential by $4zn_e V_{e-e}$, where $z = 6$ is the number of nearest neighbors on the triangular lattice. We note that we always work with a fixed number of particles. Consequently, a shift in the on-site potential should be understood as a shift of the electron band relative to the hole band, influencing the exciton energy. Additionally, a finite nearest-neighbor interaction renormalizes the electron hopping $t \rightarrow t + \bar{t}$, with

$$\bar{t} = \frac{V_{e-e}}{N} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{a}} \langle c_{\sigma \mathbf{k}}^\dagger c_{\sigma \mathbf{k}} \rangle, \quad (\text{C3})$$

where $\mathbf{a}$ is a lattice vector, connecting nearest-neighbor sites of the triangular lattice. For low densities, we now take the repulsive interactions to be zero, which can be equivalently understood as working with the Fermi-liquid quasiparticles, instead of the bare electrons. Hence, also the value of the attractive interactions U_{e-h} , which we now estimate from experiments, should be thought of as the effective renormalized interaction strength. We fix U_{e-h} through the trion binding energy, which was measured in Ref. [26] as a redshift of the order of $E_T \sim 10$ meV in the photoluminescence spectrum, when tuning away from charge neutrality $\nu = 0$. In our calculation, this redshift corresponds to the difference in energy between the AP and RP resonances at low densities (see Fig. 5). Assuming, for simplicity, vanishing nearest-neighbor attraction, we find $U_{e-h} = 15t$ is consistent with $E_T \sim 10$ meV.

We use this value of U_{e-h} in Fig. 1. Note that the actual interaction strength will be strongly renormalized as a function of density due to screening. Accurately capturing this is beyond the scope of this work. Instead, we assume a constant interaction strength in the low- and high-density regimes and show parameter scans over different interaction strengths to argue that our results are qualitatively stable (see Figs. 6 and 7). In the Fermi-liquid regime, increasing repulsive interactions U_{e-e} and V_{e-e} leads to a linear blueshift,

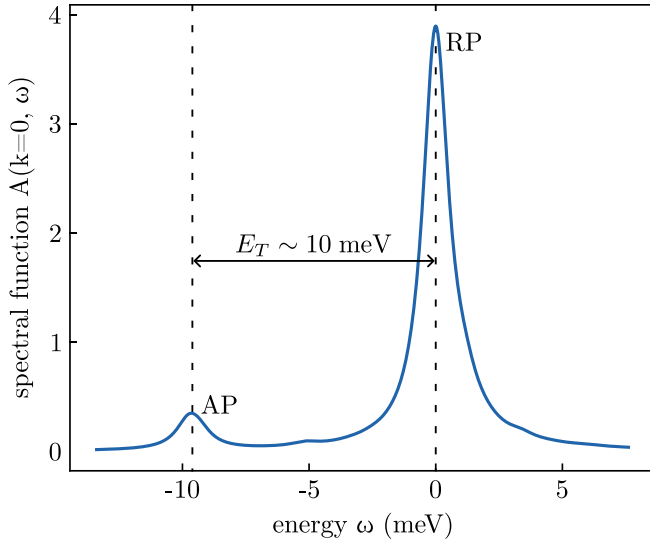


FIG. 5. Trion binding energy. We fix the interaction strength in the main text by calculating the binding energy of the trion, which corresponds to the energy difference between the AP and RP in the low-density regime $\nu \rightarrow 0$. Here, we use H stacking, system size $N = 12 \times 12$, $U_{e-h} = 15t$, and density $\nu = 1/36$, resulting in a trion energy of $E_T \sim 10$ meV, consistent with experiments [26]. The hopping parameter is estimated to be $t \sim 1$ meV. All other interaction strengths are set to zero.

as a consequence of the density-dependent on-site potential [Eq. (C2)]. At low densities, the effect of increasing U_{e-e} and V_{e-e} is marginal, which is why we have neglected it in the discussion of Fig. 1 in the main text.

Increasing the attractive electron-hole interaction U_{e-h} induces a redshift of both the RP and AP resonances. We find that the rate at which the AP redshifts is approximately twice as fast as the redshift of the RP. This is consistent with the interpretation of the AP as a charged exciton. Neglecting kinetic energy, we expect a naive binding energy of $\omega_{AP} \sim 2U_{e-h}$, while the energy of the RP, which is continuously connected to the bare exciton, should scale as $\omega_{RP} \sim U_{e-h}$. This is consistent with what we find (Figs. 6 and 7).

APPENDIX D: CORRELATED INSULATORS

We enlarge the unit cell to include three sublattices to describe the correlated insulators at $\nu = 1/3$, $\nu = 2/3$, and $\nu = 1$. The lattice translation vectors with respect to the unit cell are $\mathbf{b}_1 = \mathbf{a}_1 - \mathbf{a}_3$ and $\mathbf{b}_2 = \mathbf{a}_2 - \mathbf{a}_3$. Furthermore, we define $\mathbf{b}_0 = \mathbf{a}_1 - \mathbf{a}_2$, with the translation vectors $\mathbf{a}_i$ of the original lattice. We treat the repulsive electron-electron interactions on a mean-field level, defining $\chi_{\sigma\sigma'}^{ij} := \langle c_{\sigma i}^\dagger c_{\sigma' j} \rangle$. The electron-electron interaction Hamiltonian reads

$$H_{e-e} = U_{e-e} \sum_{i,\sigma,\sigma'} c_{\sigma i}^\dagger c_{\sigma' i}^\dagger c_{\sigma' i} c_{\sigma i} + V_{e-e} \sum_{\substack{(i,j) \\ \sigma,\sigma'}} c_{\sigma i}^\dagger c_{\sigma' j}^\dagger c_{\sigma' j} c_{\sigma i} + X_{e-e} \sum_{\substack{(i,j) \\ \sigma,\sigma'}} c_{\sigma i}^\dagger c_{\sigma' j}^\dagger c_{\sigma' i} c_{\sigma j}. \quad (D1)$$

After mean-field decoupling, it becomes

$$H_U = U_{e-e} \sum_{i,\sigma} (\chi_{\sigma\sigma}^{ii} c_{\sigma i}^\dagger c_{\sigma i} - \chi_{\sigma\bar{\sigma}}^{ii} c_{\sigma i}^\dagger c_{\bar{\sigma} i}) - U_{e-e} \sum_i (\chi_{\uparrow\uparrow}^{ii} \chi_{\downarrow\downarrow}^{ii} - \chi_{\uparrow\downarrow}^{ii} \chi_{\downarrow\uparrow}^{ii}), \quad (D2a)$$

$$H_V = V_{e-e} \sum_{\langle ij \rangle} \sum_{\sigma\sigma'} (\chi_{\sigma\sigma}^{ii} c_{\sigma' j}^\dagger c_{\sigma' j} + \chi_{\sigma'\sigma'}^{jj} c_{\sigma i}^\dagger c_{\sigma i} - \chi_{\sigma'\sigma}^{ji} c_{\sigma i}^\dagger c_{\sigma' j} - \chi_{\sigma\sigma'}^{ij} c_{\sigma' j}^\dagger c_{\sigma i} - \chi_{\sigma\sigma}^{jj} \chi_{\sigma'\sigma'}^{ij} + \chi_{\sigma'\sigma}^{ji} \chi_{\sigma\sigma'}^{ij}), \quad (D2b)$$

$$H_X = X_{e-e} \sum_{\langle ij \rangle} \sum_{\sigma\sigma'} (\chi_{\sigma\sigma}^{ii} c_{\sigma' j}^\dagger c_{\sigma' j} + \chi_{\sigma'\sigma'}^{jj} c_{\sigma i}^\dagger c_{\sigma i} - \chi_{\sigma\sigma'}^{ij} c_{\sigma' j}^\dagger c_{\sigma i} - \chi_{\sigma'\sigma}^{ji} c_{\sigma i}^\dagger c_{\sigma' j} - \chi_{\sigma\sigma}^{jj} \chi_{\sigma'\sigma'}^{ii} + \chi_{\sigma'\sigma}^{ji} \chi_{\sigma\sigma'}^{ii}). \quad (D2c)$$

where the nearest-neighbor direct exchange H_X favors ferromagnetism [58]. The notation $\bar{\sigma}$ implies opposite spin relative to σ , i.e., $\bar{\uparrow} = \downarrow$ and $\bar{\downarrow} = \uparrow$. In the enlarged unit cell, we bring the mean-field Hamiltonian into bilinear form $H_e = \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^\dagger H_{\mathbf{k}} \Psi_{\mathbf{k}}$, with the sublattice spinor $\Psi_{\mathbf{k}} = (c_{A\uparrow\mathbf{k}}, c_{A\downarrow\mathbf{k}}, c_{B\uparrow\mathbf{k}}, c_{B\downarrow\mathbf{k}}, c_{C\uparrow\mathbf{k}}, c_{C\downarrow\mathbf{k}})^T$. The matrix $H_{\mathbf{k}}$ depends on the mean-field parameters $\chi_{\sigma\sigma'}^{ab}$, which we determine self-consistently. To define the momenta of the top and bottom layers with respect to the same translations, we also need to enlarge the unit cell for the holes, but since we neglect hole-hole interactions, this only leads to a folding of the hole bands. Concretely, we write the electron and hole operators in the enlarged unit cell in terms of the eigenbases

$$c_{\sigma a \mathbf{k}} = \sum_{\lambda=1}^6 \mathcal{U}_{\sigma a}^\lambda(\mathbf{k}) \gamma_{\lambda \mathbf{k}} \quad \text{and} \quad h_{\sigma a \mathbf{k}} = \sum_{\lambda=1}^6 \mathcal{W}_{\sigma a}^\lambda(\mathbf{k}) h_{\lambda \mathbf{k}}, \quad (D3)$$

where $a \in \{A, B, C\}$ is the sublattice index. Expressed in these bases, the electron-hole interaction becomes

$$H_{e-h} = \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} \sum_{\substack{\mu, \mu' \\ \nu, \nu'}} V_{\mu'\mu}^{\nu\nu}(\mathbf{k}, \mathbf{k}', \mathbf{q}) \gamma_{\mu', \mathbf{k}+\mathbf{q}}^\dagger h_{\nu', \mathbf{k}'-\mathbf{q}}^\dagger h_{\nu, \mathbf{k}'} \gamma_{\mu, \mathbf{k}}, \quad (D4)$$

with

$$V_{\mu'\mu}^{\nu\nu}(\mathbf{k}, \mathbf{k}', \mathbf{q}) = \sum_{\substack{ab \\ \sigma\sigma'}} V_{\mathbf{q}}^{ab} \bar{\mathcal{U}}_{\sigma a}^{\mu'}(\mathbf{k} + \mathbf{q}) \mathcal{U}_{\sigma a}^\mu(\mathbf{k}) \times \bar{\mathcal{W}}_{\sigma' b}^{\nu'}(\mathbf{k}' - \mathbf{q}) \mathcal{W}_{\sigma' b}^\nu(\mathbf{k}'). \quad (D5)$$

Here, a barred variable denotes complex conjugation. The precise form of $V_{\mathbf{q}}^{ab}$ depends on the stacking configuration between the two layers. For R stacking, we find

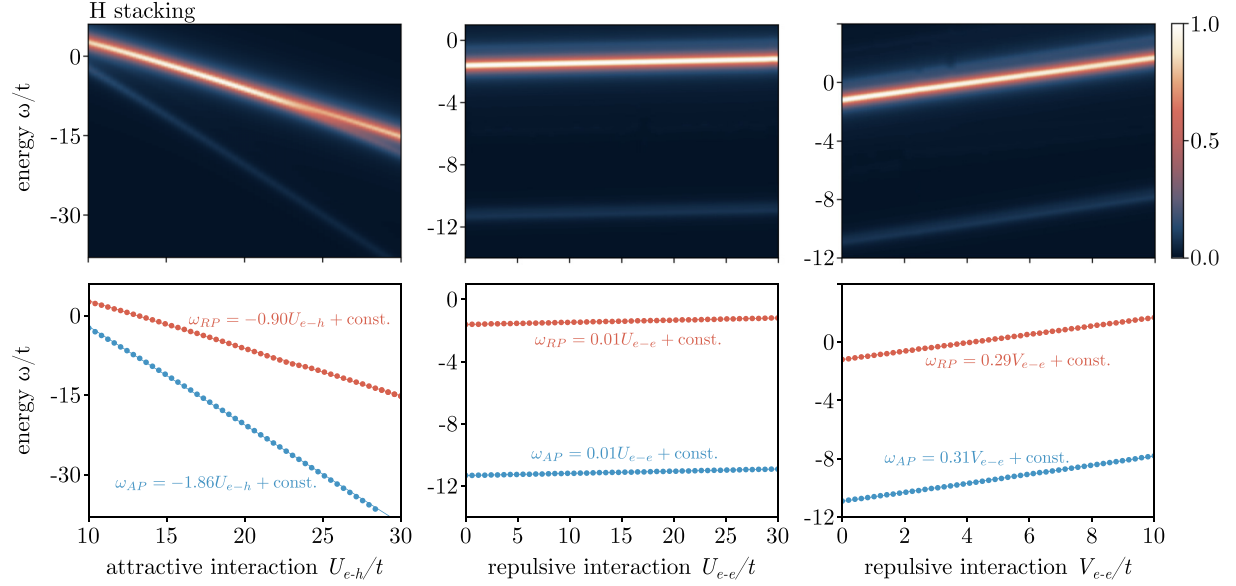


FIG. 6. Parameter scans for H stacking. Top panels show the zero-momentum spectral function as a function of various interaction strengths. Bottom panels show the extracted RP and AP resonances and the linear fit to them. Left: Tuning the attractive electron-hole interaction U_{e-h} , with all other interactions set to zero. The AP redshifts approximately twice as fast as the RP, consistent with the interpretation of the AP as a charged exciton. Center: Tuning the repulsive on-site interaction U_{e-e} , with $U_{e-h} = 15t$ fixed and all other interactions zero. On a mean-field level, the only effect of U_{e-e} is to provide a constant energy shift. Right: Tuning the repulsive nearest-neighbor interaction V_{e-e} , with $U_{e-h} = 15t$ and $U_{e-e} = 30t$ fixed. $N = 12 \times 12$, $\nu = 1/72 \approx 0.014$.

$$V_{\mathbf{q}}^{ab} = U\delta^{ab}/N + VA_{\mathbf{q}}^{ab}/N, \text{ with}$$

$$A_{\mathbf{k}}^{AB} = 1 + e^{i\mathbf{b}_1 \cdot \mathbf{k}} + e^{i(\mathbf{b}_1 - \mathbf{b}_2) \cdot \mathbf{k}}, \quad (\text{D6a})$$

$$A_{\mathbf{k}}^{AC} = 1 + e^{i\mathbf{b}_1 \cdot \mathbf{k}} + e^{i\mathbf{b}_2 \cdot \mathbf{k}}, \quad (\text{D6b})$$

$$A_{\mathbf{k}}^{BC} = 1 + e^{i\mathbf{b}_2 \cdot \mathbf{k}} + e^{i(\mathbf{b}_2 - \mathbf{b}_1) \cdot \mathbf{k}}, \quad (\text{D6c})$$

$$\text{and } A_{\mathbf{k}}^{aa} = 0, A_{\mathbf{k}}^{ba} = \bar{A}_{\mathbf{k}}^{ab}.$$

In the basis (D3), we write the exciton [Eq. (5)] as

$$|X_{\mathbf{p}}\rangle = x_{\mathbf{p}}^{\dagger}|\text{GS}\rangle = \sum_{\mathbf{k}} \sum_{\lambda, \rho=1}^6 \chi_{\mathbf{k}}^{\lambda\rho}(\mathbf{p}) \gamma_{\lambda\mathbf{p}-\mathbf{k}}^{\dagger} h_{\rho\mathbf{k}}^{\dagger} |\text{GS}\rangle. \quad (\text{D7})$$

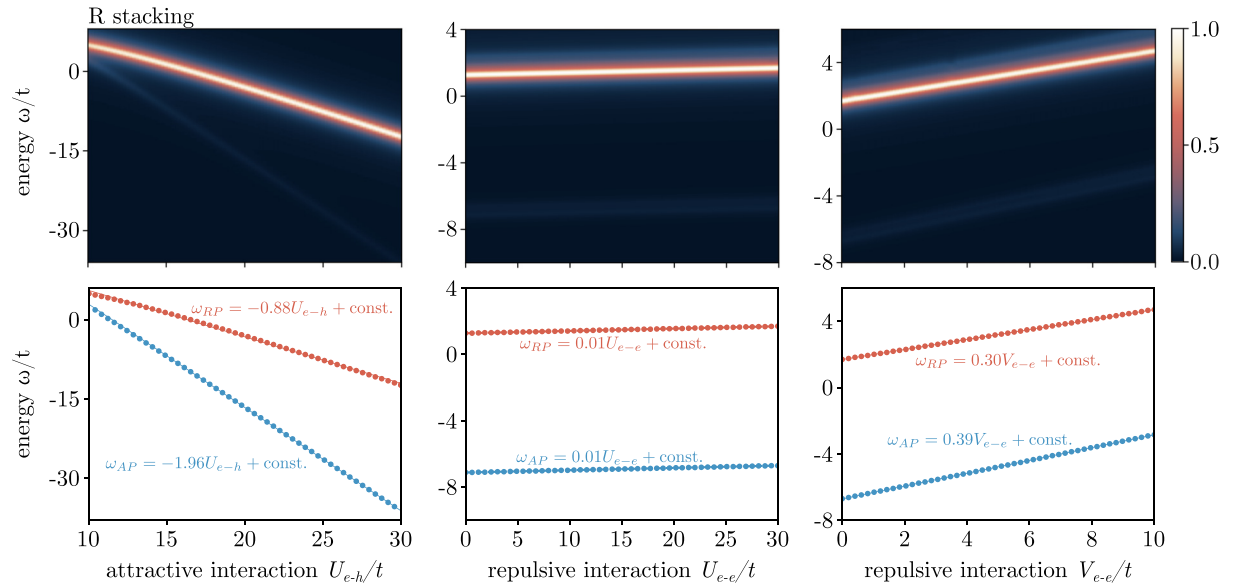


FIG. 7. Parameter scans for R stacking. Top panels show the zero-momentum spectral function as a function of various interaction strengths. Bottom panels show the extracted RP and AP resonances and the linear fit to them. Left: Tuning the attractive electron-hole interaction U_{e-h} , with all other interactions set to zero. The AP redshifts approximately twice as fast as the RP, consistent with the interpretation of the AP as a charged exciton. Center: Tuning the repulsive on-site interaction U_{e-e} , with $U_{e-h} = 15t$ fixed and all other interactions zero. On a mean-field level, the only effect of U_{e-e} is to provide a constant energy shift. Right: Tuning the repulsive nearest-neighbor interaction V_{e-e} , with $U_{e-h} = 15t$ and $U_{e-e} = 30t$ fixed. $N = 12 \times 12$, $\nu = 1/72 \approx 0.014$.

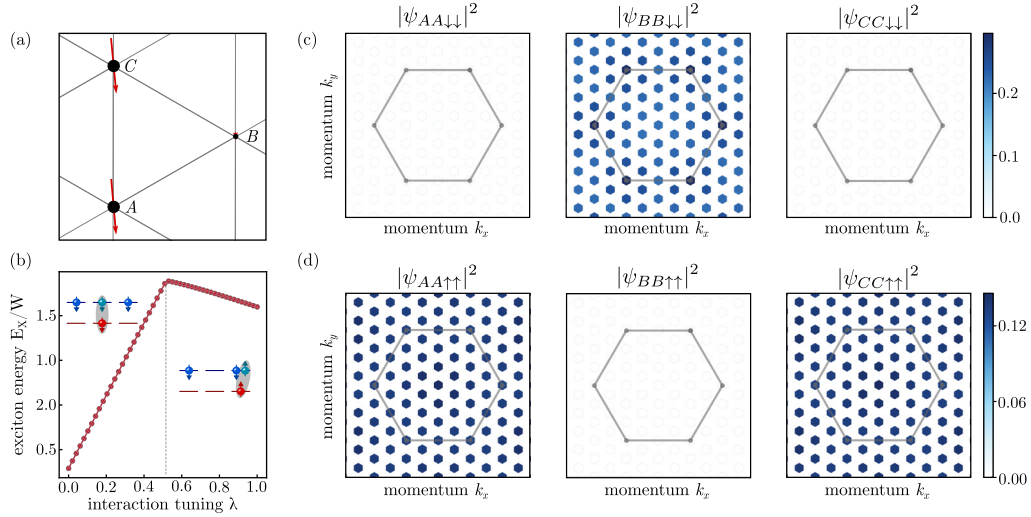


FIG. 8. Exciton wavefunction in generalized Wigner crystals. (a) Mean-field solution for ferromagnetic generalized Wigner crystal at $\nu = 2/3$, $U_{e-e} = 16t$, $V_{e-e} = U_{e-e}/6$, and $X_{e-e} = 0.8t$. The two sublattices A and C are occupied, while B is nearly empty. (b) Exciton energy as a function of a continuous tuning parameter λ , which linearly interpolates between $U_{e-h} = -15t$, $V_{e-h} = -10t$ ($\lambda = 0$) and $U_{e-h} = -35t$, $V_{e-h} = 0$ ($\lambda = 1$). For small λ , the exciton is formed on empty sites; for large λ , it forms on top of the crystal. (c) Sublattice and spin-resolved components of the exciton wavefunction $\psi_{ab,\mathbf{k}}^{\sigma\sigma'}(\mathbf{p} = 0)$, for $\lambda = 0$. Here, the exciton forms on an empty site, with the electron spin aligned with the spins of the crystal. (d) For $\lambda = 1$, where the on-site attraction is strong, the exciton forms on top of the crystal. All other components of the wavefunction, which are not shown, vanish. System size $N = 3 \times (6 \times 6)$ and R stacking.

The exciton wavefunction $\chi_{\mathbf{k}}^{\lambda,\rho}(\mathbf{p})$ is obtained as the lowest-energy solution of the following equation:

$$\begin{aligned} & \sum_{\nu'} [(\epsilon_{\mathbf{p}-\mathbf{k}}^{\mu} + \tilde{\epsilon}_{\nu}^{\nu})\delta_{\nu\nu'} + \mathcal{V}_{\nu\nu'}(\mathbf{k})]\theta_{\mathbf{p}-\mathbf{k}}^{\mu}\chi_{\mathbf{k}}^{\mu\nu'}(\mathbf{p}) \\ & + \sum_{\substack{\mathbf{k}' \\ \mu'\nu'}} V_{\mu\mu'}^{\nu\nu'}(\mathbf{p}-\mathbf{k}', \mathbf{k}', \mathbf{k}-\mathbf{k}')\theta_{\mathbf{p}-\mathbf{k}'}^{\mu'}\theta_{\mathbf{p}-\mathbf{k}}^{\mu}\chi_{\mathbf{k}'}^{\mu'\nu'}(\mathbf{p}) \\ & = E_{\mathbf{p}}^X \chi_{\mathbf{k}}^{\mu\nu}(\mathbf{p}), \end{aligned} \quad (\text{D8})$$

with $\mathcal{V}_{\nu\nu'}(\mathbf{k}) = \sum_{\lambda\mathbf{k}'} n_{\mathbf{k}'}^{\lambda} V_{\lambda\lambda}^{\nu\nu'}(\mathbf{k}', \mathbf{k}, 0)$ and $\tilde{\epsilon}_{\mathbf{k}}^{\lambda}$ the hole dispersion in the basis. As remarked in the main text, the resulting exciton wavefunction does not necessarily represent an optically generated exciton. Instead, it describes the long-lived, lowest-energy exciton state to which optically created excitons relax. The relaxed, lowest-energy exciton state is relevant for photoluminescence and transport measurements, while an optical reflectance measurement would directly probe the optically excited exciton. Since reflectance measurements are difficult for interlayer excitons due to their small oscillator strength [48,49], we believe that the long-lived exciton states we focus on are experimentally more relevant. Nonetheless, assuming that one could selectively excite excitons at different energies, we could also target higher-energy exciton bound states in Eq. (D8) and use them to study the polaron formation in our model.

After solving Eq. (D8) in the Hartree-Fock basis, one can express the exciton wavefunction in the original basis again:

$$\psi_{ab,\mathbf{k}}^{\sigma\sigma'}(\mathbf{p}) = \sum_{\mu\nu} \chi_{\mathbf{k}}^{\mu\nu}(\mathbf{p}) \mathcal{U}_{\sigma a}^{\mu}(\mathbf{p}-\mathbf{k}) \mathcal{V}_{\sigma' b}^{\nu}(\mathbf{k}). \quad (\text{D9})$$

In the parameter regime discussed in the main text, with $|U_{e-h}| > U_{e-e}$, the exciton bound state is formed with an electron sitting atop the crystal (see Fig. 8 for $\nu = 2/3$). The

strength of the respective interactions will depend on material properties. For $|U_{e-h}| \lesssim U_{e-e}$, we find that a nearest-neighbor attractive interaction V_{e-h} is required to form an exciton bound state, which now occupies an empty site next to the crystal. Note that such a configuration is only possible for generalized Wigner crystals, not for Mott states.

In the main text, we discussed the appearance of Umklapp peaks and how they can be used to distinguish between AFM and FM spin order based on symmetry arguments. Here, we provide a simple complementary description. The Umklapp peaks can be understood as the exciton scattering off with a particle-hole excitation of the crystal. A generalized Wigner crystal at $\nu = 2/3$ filling has two of every three sites filled, while the third one is empty (see inset in Fig. 2). For the AFM, one can have both particle-hole excitations from a filled to an empty site, and a particle-hole excitation from a filled site to another filled site (creating a doublon). The two Umklapp peaks correspond to the exciton scattering with these two different particle-hole excitations, where the Umklapp peak at higher energy corresponds to the scattering with a particle-hole excitation involving the formation of a doublon. Conversely, for the FM, the Pauli principle forbids a particle-hole excitation from a filled to another filled site of the crystal. This leaves only the Umklapp peak, where the exciton scatters with a particle-hole excitation from a filled site to an empty site.

APPENDIX E: DETERMINING THE MASS OF EXCITON POLARONS

We extract the effective mass of the attractive polaron from the momentum-dependent exciton spectral function $\mathcal{A}_X(\mathbf{k}, \omega) = -\frac{1}{\pi} \text{Im} \mathcal{G}_X(\mathbf{k}, \omega)$.

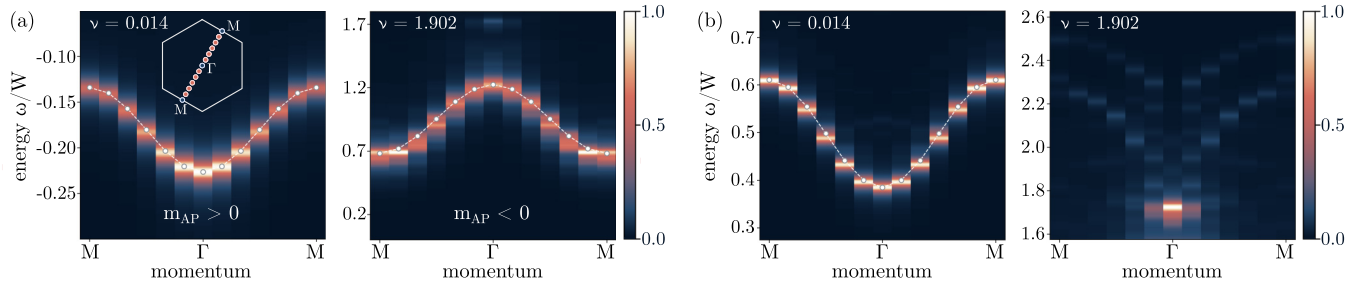


FIG. 9. Momentum-dependent exciton-polaron spectral function (R stacking). (a) Left: Dispersion of attractive polaron in the low-density regime with a positive mass. Right: In the high-density regime, the attractive polaron has a negative effective mass. Note the vastly different energy scales in the low- and high-density regimes, with a much larger mobility for high densities. The white dotted line is the fit to the dispersion of a particle on a triangular lattice with nearest-neighbor hopping. The cut through the Brillouin zone is shown as an inset. (b) Dispersion for the RP branch. In the high-density regime, the RP is no longer a well-defined quasiparticle. We used $U_{e-h} = -12t$ for the electron-hole interaction; all other interactions are set to zero. System size of $N = 12 \times 12$ and R stacking.

We define the mass m of a particle with dispersion $\varepsilon_{\mathbf{k}}$ as $m^{-1} = \partial^2 \varepsilon_{\mathbf{k}} / \partial \mathbf{k}^2|_{\mathbf{k}=0}$. For a particle hopping on a triangular lattice, the mass is given by $m = 3/W$, where W is the bandwidth of $\varepsilon_{\mathbf{k}}$. We use this relation to determine the mass of the AP by calculating its bandwidth.

Concretely, we calculate the bandwidth of the attractive polaron in units of the bare electron bandwidth $W = 9t$. For a particle hopping on a triangular lattice, its mass is related to the bandwidth through $m = 3/W$, which follows from $m^{-1} = \partial^2 \varepsilon_{\mathbf{k}} / \partial \mathbf{k}^2|_{\mathbf{k}=0}$ and $\varepsilon_{\mathbf{k}}$. The sign of t_{AP} is determined by the curvature of the dispersion at the Γ point. The mass of the attractive polaron is inversely proportional to the hopping t_{AP} . For low-electron densities, the mass is positive, while in the high-density regime, it is negative [see Fig. 9(a)]. In the low-density regime, the RP is also a quasiparticle with a well-defined dispersion. At the same electron densities, we find that the RP mass is smaller than the AP mass [compare Figs. 9(a) and 9(b)]. In the high-density regime, the RP no longer has a well-defined dispersion, but rather exhibits a spectral weight peak at zero momentum. This indicates that the RP is no longer a well-defined quasiparticle. Analogous results also hold for H stacking (see Fig. 10)

For Fig. 3(b), we used $U_{e-h} = -12t$ and $V_{e-h} = -3t$ with a system size of $N = 9 \times 9$ in the low- and high-density

regimes, while we used $U_{e-h} = -35t$ and a system size of $N = 3 \times (6 \times 6)$ for the correlated insulators at the fillings $\nu \in \{1/3, 2/3, 1\}$. For lower fillings, stronger long-range interactions V_{e-e} are required to stabilize the crystal order. For $\nu = 1$, $\nu = 2/3$, and $\nu = 1/3$, we use $V_{e-e} = 0$, $V_{e-e} = U_{e-h}/6$, and $V_{e-e} = U_{e-h}/3$, respectively. We find that exciton mobility is strongly affected by the different exciton wavefunctions. As discussed in the previous section, the exciton forms on top of the crystal for large $|U_{e-h}|$, while it sits on an empty site for smaller $|U_{e-h}|$ and larger $|V_{e-h}|$ [see Figs. 8 and 11(a)]. In the latter case, for $\nu = 1/3$, the exciton polaron can hop between empty sites, leading to enhanced mobility compared to the localized exciton atop the crystal. The situation is reversed for the $\nu = 2/3$ state since empty sites are not connected to each other [see Fig. 11(b)].

APPENDIX F: CHEVY APPROXIMATION

Using the Chevy approximation, we approach the many-body problem of a single exciton immersed in an electronic ground state. We calculate the exciton propagator $\mathcal{G}_X(\mathbf{p}, \omega)$, defined in Eq. (6), by rewriting it as

$$\mathcal{G}_X(\mathbf{k}, \omega) = \langle X_{\mathbf{k}} | \Psi_{\mathbf{k}}(\omega) \rangle, \quad (\text{F1})$$

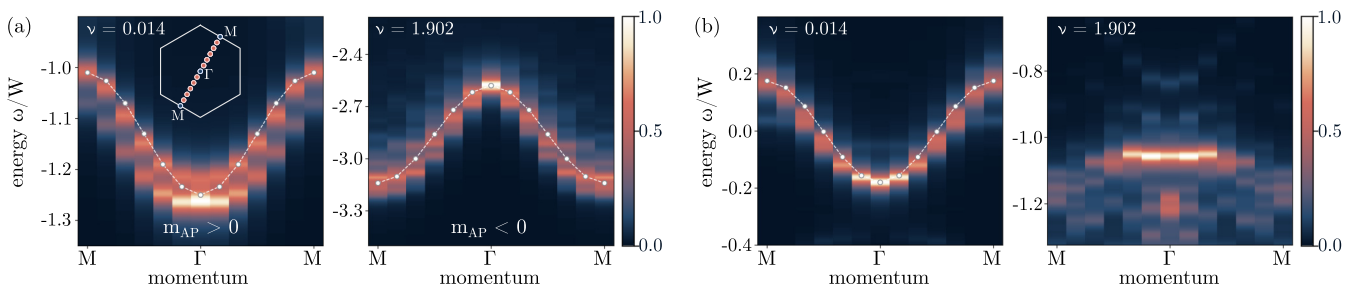


FIG. 10. Momentum-dependent exciton-polaron spectral function (H stacking). (a) Left: Dispersion of attractive polaron in the low-density regime with a positive mass. Right: In the high-density regime, the attractive polaron has a negative effective mass. Note the vastly different energy scales in the low- and high-density regimes, with a much larger mobility for high densities. The white dotted line is the fit to the dispersion of a particle on a triangular lattice with nearest-neighbor hopping. The cut through the Brillouin zone is shown as an inset. (b) Dispersion for the RP branch. In the high-density regime, the RP is no longer a well-defined quasiparticle. We used $U_{e-h} = -15t$ for the electron-hole interaction; all other interactions are set to zero. System size of $N = 12 \times 12$ and H stacking.

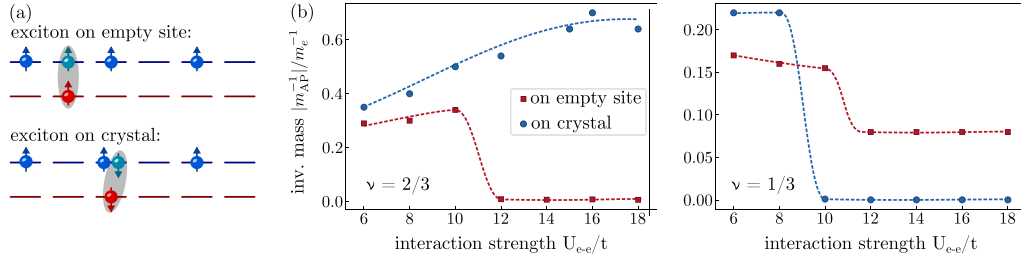


FIG. 11. Exciton mass renormalization in generalized Wigner crystals. (a) If the on-site attractive interaction is small, $|U_{e-h}| \lesssim U_{e-e}$, and V_{e-h} sufficiently large, the exciton forms on an empty site (top). In contrast, for large $|U_{e-h}| > U_{e-e}$, doubly occupancy is favored, leading to an exciton formed atop the crystal (bottom). (b) Exciton-polaron mobility in generalized Wigner crystals for $\nu = 2/3$ (left) and $\nu = 1/3$ (right) is strongly affected by whether the exciton is on an empty site or atop the crystal. For the empty site exciton, we use $U_{e-h} = -15t$ and $V_{e-h} = -10t$, for the exciton atop the crystal $U_{e-h} = -35t$ and $V_{e-h} = 0$. In the $\nu = 2/3$ crystal $V_{e-e} = U_{e-e}/6$ and in the $\nu = 1/3$ crystal $V_{e-e} = U_{e-e}/3$. System size $N = 3 \times (6 \times 6)$ and R stacking.

with

$$|\Psi_{\mathbf{k}}(\omega)\rangle = \frac{1}{\omega + i\eta - H} |X_{\mathbf{k}}\rangle. \quad (\text{F2})$$

We obtain the state $|\Psi_{\mathbf{k}}(\omega)\rangle$ by iteratively solving the linear equation

$$(\omega + i\eta - H)|\Psi_{\mathbf{k}}(\omega)\rangle = |X_{\mathbf{k}}\rangle, \quad (\text{F3})$$

for each ω and some finite regularization η . To make this computationally feasible, we use the Chevy approximation, which restricts the Hilbert space to states with a single exciton and an exciton with a particle-hole excitation of the electronic ground state. Concretely, we define the following basis states:

$$\begin{aligned} |n=0\rangle &= |X_{\mathbf{p}}\rangle \equiv x_{\mathbf{p}}^{\dagger} |\text{GS}\rangle, \\ |n>0\rangle &= |C_{\mathbf{k}\alpha\mathbf{q}\beta}^{\mathbf{p}}\rangle \equiv x_{\mathbf{p}+\mathbf{q}-\mathbf{k}}^{\dagger} c_{\alpha\mathbf{k}}^{\dagger} c_{\beta\mathbf{q}} |\text{GS}\rangle, \end{aligned} \quad (\text{F4})$$

and compute the matrix elements of the Hamiltonian in this basis:

$$H_{nm} = \langle n | H | m \rangle. \quad (\text{F5})$$

Note that we treat the electron-electron interaction only on a mean-field level. Consequently, its only effect is to renormalize the electron dispersion. In the conventional Chevy ansatz, where the exciton is treated as a bosonic pointlike particle, the Chevy basis [Eq. (F4)] is orthogonal. However, since we take the internal structure of the exciton into account, this is no longer the case,

$$b_{nm} := \langle n | m \rangle \neq \delta_{nm}. \quad (\text{F6})$$

Projected to the Chevy basis, Eq. (F3) reduces to the following matrix equation:

$$\sum_m [(\omega + i\eta)b_{nm} - H_{nm}] \Psi_{\mathbf{k}}^m(\omega) = X_{\mathbf{k}}^n \quad \text{with} \quad \Psi_{\mathbf{k}}^n(\omega) = \langle n | \Psi_{\mathbf{k}}(\omega) \rangle \quad \text{and} \quad X_{\mathbf{k}}^n = \langle n | X_{\mathbf{k}} \rangle. \quad (\text{F7})$$

The matrix elements of the Hamiltonian [Eq. (F5)] can be computed explicitly:

$$\langle X_{\mathbf{p}} | H | X_{\mathbf{p}} \rangle = \sum_{\mathbf{k}} |\psi_{\mathbf{p}-\mathbf{k}}(\mathbf{p})|^2 \theta_{\uparrow\mathbf{k}} (\varepsilon_{\mathbf{k}}^e + \varepsilon_{\mathbf{p}-\mathbf{k}}^h + \nu V_{\mathbf{p}=0}) + \frac{1}{N} \sum_{\mathbf{k}\mathbf{q}} V_{\mathbf{q}} \bar{\psi}_{\mathbf{p}-\mathbf{k}-\mathbf{q}}(\mathbf{p}) \psi_{\mathbf{p}-\mathbf{k}}(\mathbf{p}) \theta_{\uparrow\mathbf{k}} \theta_{\uparrow\mathbf{k}+\mathbf{q}}, \quad (\text{F8a})$$

$$\begin{aligned} \langle C_{\mathbf{k}\alpha\mathbf{q}\beta}^{\mathbf{p}} | H | X_{\mathbf{p}} \rangle &= -\bar{\psi}_{\mathbf{p}-\mathbf{k}}(\mathbf{p}) \psi_{\mathbf{p}-\mathbf{k}}(\mathbf{p} + \mathbf{q} - \mathbf{k}) (\varepsilon_{\mathbf{k}}^e + \varepsilon_{\mathbf{p}-\mathbf{k}}^h + \nu V_{\mathbf{p}=0}) \delta_{\alpha\uparrow} \delta_{\beta\uparrow} \\ &\quad - \frac{1}{N} \sum_{\mathbf{q}'} [V_{\mathbf{q}'} \bar{\psi}_{\mathbf{p}-\mathbf{k}-\mathbf{q}'}(\mathbf{p}) \psi_{\mathbf{p}-\mathbf{k}}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \theta_{\uparrow\mathbf{k}+\mathbf{q}'} + V_{\mathbf{q}'} \bar{\psi}_{\mathbf{p}-\mathbf{k}}(\mathbf{p}) \psi_{\mathbf{p}-\mathbf{k}+\mathbf{q}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \theta_{\uparrow\mathbf{q}-\mathbf{q}'} \\ &\quad - V_{\mathbf{q}-\mathbf{k}} \bar{\psi}_{\mathbf{q}'-\mathbf{q}+\mathbf{k}}(\mathbf{p}) \psi_{\mathbf{q}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \theta_{\uparrow\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{q}'}] \delta_{\alpha\uparrow} \delta_{\beta\uparrow}, \end{aligned} \quad (\text{F8b})$$

$$\begin{aligned} \langle C_{\mathbf{k}\alpha\mathbf{q}\beta}^{\mathbf{p}} | H | C_{\mathbf{k}'\alpha'\mathbf{q}'\beta'}^{\mathbf{p}} \rangle &= \Gamma_1^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta) \delta_{\alpha\alpha'} \delta_{\beta\beta'} \delta_{\mathbf{k}\mathbf{k}'} \delta_{\mathbf{q}\mathbf{q}'} + \Gamma_2^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta; \mathbf{q}', \beta') \delta_{\alpha\alpha'} \delta_{\mathbf{k}\mathbf{k}'} \\ &\quad + \Gamma_3^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta; \mathbf{k}', \alpha') \delta_{\beta\beta'} \delta_{\mathbf{q}\mathbf{q}'} + \Gamma_4^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta; \mathbf{k}', \alpha', \mathbf{q}', \beta'), \end{aligned} \quad (\text{F8c})$$

with

$$\begin{aligned} \Gamma_1^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta) &= \sum_{\mathbf{p}'} |\psi_{\mathbf{p}'}(\mathbf{p} + \mathbf{q} - \mathbf{k})|^2 \theta_{\uparrow\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{p}'} (\varepsilon_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{p}'}^e + \varepsilon_{\mathbf{k}}^e - \varepsilon_{\mathbf{q}}^e + \varepsilon_{\mathbf{p}'}^h + \nu V_{\mathbf{p}=0}) \\ &\quad + \frac{1}{N} \sum_{\mathbf{p}'\mathbf{q}'} V_{\mathbf{q}'} \bar{\psi}_{\mathbf{p}'-\mathbf{q}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \theta_{\uparrow\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{p}'} \theta_{\uparrow\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{p}'+\mathbf{q}'}, \end{aligned} \quad (\text{F9a})$$

$$\Gamma_2^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta; \mathbf{q}', \beta') = \bar{\psi}_{\mathbf{p}-\mathbf{k}}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}-\mathbf{k}}(\mathbf{p} + \mathbf{q}' - \mathbf{k}) (\varepsilon_{\mathbf{k}}^e + \varepsilon_{\mathbf{p}-\mathbf{k}}^h + \nu V_{\mathbf{p}=0}) \delta_{\beta\uparrow} \delta_{\beta'\uparrow}$$

$$\begin{aligned}
 & + \frac{1}{N} \sum_{\mathbf{p}'} [V_{\mathbf{p}'} \bar{\psi}_{\mathbf{p}-\mathbf{k}-\mathbf{p}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}-\mathbf{k}}(\mathbf{p} + \mathbf{q}' - \mathbf{k}) \theta_{\uparrow \mathbf{q}+\mathbf{p}'} \delta_{\beta \uparrow} \delta_{\beta' \uparrow} \\
 & + V_{\mathbf{p}'} \bar{\psi}_{\mathbf{p}-\mathbf{k}}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}-\mathbf{k}+\mathbf{p}'}(\mathbf{p} + \mathbf{q}' - \mathbf{k}) \theta_{\uparrow \mathbf{q}'-\mathbf{p}'} \delta_{\beta \uparrow} \delta_{\beta' \uparrow} \\
 & - V_{\mathbf{q}'-\mathbf{q}} \bar{\psi}_{\mathbf{p}'-\mathbf{q}'+\mathbf{q}}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}'}(\mathbf{p} + \mathbf{q}' - \mathbf{k}) \theta_{\uparrow \mathbf{p}-\mathbf{k}-\mathbf{p}'} \delta_{\beta \beta'}], \quad (\text{F9b})
 \end{aligned}$$

$$\begin{aligned}
 \Gamma_3^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta; \mathbf{k}', \alpha') & = -\bar{\psi}_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{k}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{k}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}') (\epsilon_{\mathbf{k}}^e + \epsilon_{\mathbf{k}'}^e - \epsilon_{\mathbf{q}}^e + \epsilon_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{k}'}^h + \nu V_{\mathbf{p}=0}) \delta_{\alpha \uparrow} \delta_{\alpha' \uparrow} \\
 & - \frac{1}{N} \sum_{\mathbf{p}'} [V_{\mathbf{p}'} \bar{\psi}_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{k}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{k}'+\mathbf{p}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}') \theta_{\uparrow \mathbf{k}-\mathbf{p}'} \delta_{\alpha \uparrow} \delta_{\alpha' \uparrow} \\
 & + V_{\mathbf{p}'} \bar{\psi}_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{k}'-\mathbf{p}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{k}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}') \theta_{\uparrow \mathbf{k}'+\mathbf{p}'} \delta_{\alpha \uparrow} \delta_{\alpha' \uparrow} \\
 & - V_{\mathbf{k}-\mathbf{k}'} \bar{\psi}_{\mathbf{p}'-\mathbf{k}+\mathbf{k}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}') \theta_{\uparrow \mathbf{p}+\mathbf{q}-\mathbf{k}'-\mathbf{p}'} \delta_{\alpha \alpha'}], \quad (\text{F9c})
 \end{aligned}$$

$$\begin{aligned}
 \Gamma_4^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta; \mathbf{k}', \alpha', \mathbf{q}', \beta') & = -V_{\mathbf{k}-\mathbf{q}} \bar{\psi}_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{k}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}-\mathbf{k}'}(\mathbf{p} + \mathbf{q}' - \mathbf{k}') \delta_{\alpha \beta} \delta_{\alpha' \uparrow} \delta_{\beta' \uparrow} \\
 & - V_{\mathbf{q}'-\mathbf{k}'} \bar{\psi}_{\mathbf{p}-\mathbf{k}}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}-\mathbf{k}-\mathbf{q}'}(\mathbf{p} + \mathbf{q}' - \mathbf{k}') \delta_{\alpha' \beta'} \delta_{\alpha \uparrow} \delta_{\beta \uparrow} \\
 & + V_{\mathbf{k}-\mathbf{k}'} \bar{\psi}_{\mathbf{p}-\mathbf{k}}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}-\mathbf{k}'}(\mathbf{p} + \mathbf{q}' - \mathbf{k}') \delta_{\alpha \alpha'} \delta_{\beta \uparrow} \delta_{\beta' \uparrow} \\
 & + V_{\mathbf{q}'-\mathbf{q}} \bar{\psi}_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{k}'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \psi_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{k}'}(\mathbf{p} + \mathbf{q}' - \mathbf{k}') \delta_{\beta \beta'} \delta_{\alpha \uparrow} \delta_{\alpha' \uparrow}. \quad (\text{F9d})
 \end{aligned}$$

For correlated insulators, the Chevy calculation can be carried out analogously: Expressed in terms of the Hartree-Fock quasiparticles defined in Eq. (D3), the Chevy basis becomes

$$|n = 0\rangle = |X_{\mathbf{p}}\rangle \equiv x_{\mathbf{p}}^{\dagger} |\text{GS}\rangle, \quad (\text{F10a})$$

$$|n > 0\rangle = |C_{\mathbf{k}\mu\mathbf{q}\nu}^{\mathbf{p}}\rangle \equiv x_{\mathbf{p}+\mathbf{q}-\mathbf{k}}^{\dagger} \gamma_{\mu\mathbf{k}}^{\dagger} \gamma_{\nu\mathbf{q}} |\text{GS}\rangle, \quad (\text{F10b})$$

and the matrix elements of the Hamiltonian are given by

$$\begin{aligned}
 \langle X_{\mathbf{p}} | H | X_{\mathbf{p}} \rangle & = \sum_{\mathbf{k}} \bar{\chi}_{\mathbf{p}-\mathbf{k}}^{\mu\nu'}(\mathbf{p}) \chi_{\mathbf{p}-\mathbf{k}}^{\mu\nu}(\mathbf{p}) [(\epsilon_{\mathbf{k}}^{\mu} + \tilde{\epsilon}_{\mathbf{p}-\mathbf{k}}^{\nu}) \delta_{\nu\nu'} + \mathcal{V}_{\nu\nu'}(\mathbf{p} - \mathbf{k})] \theta_{\mathbf{k}}^{\mu} \\
 & + \frac{1}{N} \sum_{\mathbf{k}\mathbf{q}} \sum_{\mu\nu\mu'\nu'} V_{\mu'\mu}^{\nu'\nu}(\mathbf{k}, \mathbf{p} - \mathbf{k}, \mathbf{q}) \bar{\chi}_{\mathbf{p}-\mathbf{k}-\mathbf{q}}^{\mu'\nu'}(\mathbf{p}) \chi_{\mathbf{p}-\mathbf{k}}^{\mu\nu}(\mathbf{p}) \theta_{\mathbf{k}}^{\mu} \theta_{\uparrow\mathbf{k}+\mathbf{q}}^{\mu'}, \quad (\text{F11a})
 \end{aligned}$$

$$\begin{aligned}
 \langle C_{\mathbf{k}\alpha\mathbf{q}\beta}^{\mathbf{p}} | H | X_{\mathbf{p}} \rangle & = - \sum_{\nu\nu'} \bar{\chi}_{\mathbf{p}-\mathbf{k}}^{\alpha\nu'}(\mathbf{p}) \chi_{\mathbf{p}-\mathbf{k}}^{\beta\nu}(\mathbf{p} + \mathbf{q} - \mathbf{k}) [(\epsilon_{\mathbf{k}}^{\alpha} + \tilde{\epsilon}_{\mathbf{p}-\mathbf{k}}^{\nu}) \delta_{\nu\nu'} + \mathcal{V}_{\nu\nu'}(\mathbf{p} - \mathbf{k})] \\
 & - \frac{1}{N} \sum_{\substack{\mathbf{q}' \\ \mu\nu\nu'}} [V_{\mu\alpha}^{\nu'\nu}(\mathbf{k}, \mathbf{p} - \mathbf{k}, \mathbf{q}') \bar{\chi}_{\mathbf{p}-\mathbf{k}-\mathbf{q}'}^{\mu\nu'}(\mathbf{p}) \chi_{\mathbf{p}-\mathbf{k}}^{\beta\nu}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \theta_{\mathbf{k}+\mathbf{q}'}^{\mu} \\
 & + \bar{V}_{\mu\beta}^{\nu\nu'}(\mathbf{q}, \mathbf{p} - \mathbf{k}, -\mathbf{q}') \bar{\chi}_{\mathbf{p}-\mathbf{k}}^{\mu\nu}(\mathbf{p}) \chi_{\mathbf{p}-\mathbf{k}+\mathbf{q}'}^{\mu\nu}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \theta_{\mathbf{q}-\mathbf{q}'}^{\mu} \\
 & - V_{\beta\alpha}^{\nu\nu'}(\mathbf{k}, \mathbf{q}', \mathbf{q} - \mathbf{k}) \bar{\chi}_{\mathbf{q}'-\mathbf{q}+\mathbf{k}}^{\mu\nu'}(\mathbf{p}) \chi_{\mathbf{q}'}^{\mu\nu'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \theta_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{q}'}^{\mu}], \quad (\text{F11b})
 \end{aligned}$$

$$\begin{aligned}
 \langle C_{\mathbf{k}\alpha\mathbf{q}\beta}^{\mathbf{p}} | H | C_{\mathbf{k}'\alpha'\mathbf{q}'\beta'}^{\mathbf{p}} \rangle & = \Gamma_1^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta) \delta_{\alpha\alpha'} \delta_{\beta\beta'} \delta_{\mathbf{k}\mathbf{k}'} \delta_{\mathbf{q}\mathbf{q}'} + \Gamma_2^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta; \mathbf{q}', \beta') \delta_{\alpha\alpha'} \delta_{\mathbf{k}\mathbf{k}'} \\
 & + \Gamma_3^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta; \mathbf{k}', \alpha') \delta_{\beta\beta'} \delta_{\mathbf{q}\mathbf{q}'} + \Gamma_4^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta; \mathbf{k}', \alpha', \mathbf{q}', \beta'), \quad (\text{F11c})
 \end{aligned}$$

with

$$\begin{aligned}
 \Gamma_1^{\mathbf{p}}(\mathbf{k}, \alpha, \mathbf{q}, \beta) & = \sum_{\substack{\mathbf{p}' \\ \mu\nu\nu'}} \bar{\chi}_{\mathbf{p}'}^{\mu\nu'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \chi_{\mathbf{p}'}^{\mu\nu}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \theta_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{p}'}^{\mu} [(\epsilon_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{p}'}^{\mu} + \tilde{\epsilon}_{\mathbf{p}'}^{\nu} + \epsilon_{\mathbf{k}}^{\alpha} - \epsilon_{\mathbf{q}}^{\beta}) \delta_{\nu\nu'} + \mathcal{V}_{\nu\nu'}(\mathbf{p}')] \\
 & + \frac{1}{N} \sum_{\substack{\mathbf{p}'\mathbf{q}' \\ \mu\mu'\nu\nu'}} V_{\mu'\mu}^{\nu'\nu}(\mathbf{p} + \mathbf{q} - \mathbf{k} - \mathbf{p}', \mathbf{p}', \mathbf{q}') \bar{\chi}_{\mathbf{p}'-\mathbf{q}'}^{\mu'\nu'}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \chi_{\mathbf{p}'}^{\mu\nu}(\mathbf{p} + \mathbf{q} - \mathbf{k}) \theta_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{p}'}^{\mu} \theta_{\mathbf{p}+\mathbf{q}-\mathbf{k}-\mathbf{p}'+\mathbf{q}'}^{\mu'}, \quad (\text{F12a})
 \end{aligned}$$

For the remaining terms in Eqs. (F8) and (F9), we only show the diagrams with an interaction vertex. Note that additionally, there would also be the same diagram without interaction vertex, equivalent to the first diagram in Eq. (G3a), for all terms except for Γ_4 :

$$\langle C_{kq}^P | H | X_P \rangle \sim \text{[Diagram 1]} + \text{[Diagram 2]} + \text{[Diagram 3]} + \text{[Diagram 4]} \quad (\text{G3b})$$

$$\Gamma_1^P(k, q) \delta_{k,k'} \delta_{q,q'} \sim \left(\text{[Diagram 1]} + \text{[Diagram 2]} \right) \begin{matrix} q \rightarrow q' \\ k \rightarrow k' \end{matrix} \quad (\text{G4a})$$

$$\Gamma_2^P(k, q, q') \delta_{k,k'} \sim \left(\text{[Diagram 1]} + \text{[Diagram 2]} + \text{[Diagram 3]} + \text{[Diagram 4]} \right) k \rightarrow k' \quad (\text{G4b})$$

$$\Gamma_3^P(k, q, k') \delta_{q,q'} \sim \left(\text{[Diagram 1]} + \text{[Diagram 2]} + \text{[Diagram 3]} + \text{[Diagram 4]} \right) q \rightarrow q' \quad (\text{G4c})$$

$$\Gamma_4^P(k, q, k', q') \sim \text{[Diagram 1]} + \text{[Diagram 2]} + \text{[Diagram 3]} + \text{[Diagram 4]} \quad (\text{G4d})$$

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