Spin-mediated Mott excitons

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I. INTRODUCTION

The physics of excitons in semiconductors, i.e., bound states of electrons and holes, is by now well established [1,2]. Excitons play an essential role in technologies such as light-emitting diodes [3], organic solar cells [4], and photodetectors [5], among others. Furthermore, there has recently been tremendous interest in hybridizing exciton states with photon modes in optical cavities [6]. Such exciton polaritons can form (nonequilibrium) Bose-Einstein condensates at remarkably high temperatures, even room temperature [7–11].

Given these applications, it is important to study the properties of excitons in systems other than conventional semiconductors. It has been convincingly established that excitonic states do exist in strongly correlated materials such as Mott insulators [12–28], yet essential aspects of Mott excitons remain poorly understood.

For example, it is known that Mott insulators are often antiferromagnetic at low temperature, but very little work has been performed to understand how and to what extent the presence of such order affects exciton properties. The qualitative role of magnetization is sketched in Fig. 1—in particular, “charges” remain bound so as to minimize the number of spins disrupted by their motion—but a quantitative description has been lacking.

Recent experiments have begun to investigate this question. In Refs. [29,30], pump-probe experiments were performed on the Mott insulator Na2IrO3 both with and without magnetic order (controlled by varying temperature or applying an intermediate pulse). The authors concluded that the binding energy and exciton mass are both enhanced by the presence of magnetization. Reference [31] similarly observed that the binding energy increases with the spin-spin interaction strength in cuprates. See also Ref. [32], which found that the relaxation time in Mott insulators decreases with increasing spin correlations.

The same question can apply to Mott insulators in synthetic quantum systems, such as ultracold gases. By loading fermionic atoms into an optical lattice and tuning their interactions, the Fermi-Hubbard model can be synthesized experimentally [33–37]. Unlike condensed-matter systems such as the iridates and cuprates, for which the Hubbard model is a significant idealization, neutral fermionic atoms in an optical lattice are genuinely described by the Hubbard Hamiltonian without any additional effects arising from longer-range Coulomb interactions, phonons, etc. Researchers have quite recently begun investigating the interplay of spin and charge degrees of freedom in this setting [38–44] (note that here the “charge” excitations are not actually charged).

In this paper, we perform a theoretical study of the role of magnetic order in Mott excitons. As depicted in Fig. 1, in an antiferromagnetic background, a hole and doubly occupied site can bind through a string of flipped spins. Such Mott excitons differ from conventional excitons formed by Coulomb interaction in two aspects. First, the spin-mediated interaction is far from instantaneous, and second, the individual charges are themselves renormalized by spin fluctuations. We will demonstrate that both effects are necessary ingredients in the trends reported here.

Given the complexity of the problem, our analysis requires multiple stages. We first use slave particles to isolate spin and charge degrees of freedom, then describe the spin dynamics...
to the Cooper pairing of holes in high-
and mass are proportional to each other. This
observation is in some tension with interpreta-
tions of recent experiments [30]. It also stands in
contrast to conventional Coulomb-mediated
excitons where the binding energy
is calculated by mean-field theory, calculate the
dispersion of charges self-
consistently, and finally characterize excitonic states via the
Bethe-Salpeter equation. Many of the steps in this program
are analogous to those in Ref. [45], which studied charge dy-
namics in the Hubbard model. The good agreement between
the results of Ref. [45] and alternate numerical methods lends
support to the present approach.

Our starting point is the two-dimensional (2D) Fermi-
Hubbard model, which by now needs no introduction,

\[ H_{\text{Hub}} = -t \sum_{\langle i,j \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}, \]

where \( \sigma \in \{\uparrow, \downarrow\} \) and \( \langle ij \rangle \) denotes nearest-neighbor sites on a square lattice. \( c_{i\sigma} \) is the usual electron annihilation operator and \( n_{i\sigma} \equiv c_{i\sigma}^\dagger c_{i\sigma} \). We will consider the system at half-filling in the \( U \gg t \) limit. Although admittedly an idealization, especially for strongly correlated materials, Eq. (1) already exhibits a wide range of interesting phenomena.

It is well known that in the large-\( U \) limit, the Hubbard
model features two types of excitations, associated with the
transport of charge and spin, respectively [51–54]. Furthermore, the charge excitations can be either positive or negative, corresponding to sites with zero or two electrons, and their creation comes with a high energy cost of order \( U \). By analogy with conventional semiconductors, we thus expect this system to support well-defined excitons in the dilute-charge limit. However, long-wavelength spin excitations do not come with an energy cost, and their presence plays a significant role in determining the exciton properties.

There are many formalisms with which to study the Hub-
rard model [28,55–58]. Since our focus is on the motion of
only a few charges within a background of spin excita-
tions, the slave-particle formalism is particularly well suited
[45,59,60]. The steps of our calculation are as follows:

(i) Express the Hamiltonian in terms of slave particles—
doublons, holons, and spinons—and reduce to the \( t-J \) model
following the standard procedure [61].

(ii) Treat the Heisenberg interaction within semiclassical
and mean-field approximations, while neglecting the backac-
tion of doublons and holons on the magnetic order.

(iii) Calculate the dispersion of individual doublons and
holons in the magnetic background via the self-consistent
Born approximation.

(iv) Calculate exciton properties using the Bethe-Salpeter
equation.

The major limitation of this program is our approximate
description of the magnetic order. Thus, we do not claim to
have quantitatively accurate results, especially at small mag-
netization. That said, we do expect that the qualitative trends
seen here are accurate, including near the equilibrium value of
magnetization for which mean-field theory is known to work
reasonably well (see Ref. [45] and references therein).

A. Slave particles

In the slave-particle formalism, we express the electron
operator as (with \( \sigma = \pm 1 \))

\[ c_{i\sigma} = s_{i\sigma}^0 d_i + \sigma e_i^\dagger s_{i\sigma}, \]

where \( d_i \) and \( e_i \) are fermionic operators and \( s_{i\sigma} \) is bosonic. One can confirm that Eq. (2) is consistent with the commutation relations. A site with a \( d \) particle is to be interpreted as a site with two electrons (a “doublon”), a site with an \( e \) particle is to be interpreted as an empty site (a “holon”), and a site with an \( s \) particle is one with a single electron having spin \( \sigma \) (a “spinon”). See Fig. 2. The physical content of Eq. (2) is then clear: removing an electron of given spin is equivalent to replacing the doublon with the opposite spinon if the site is doubly occupied and replacing the spinon with a holon if the site is singly occupied (otherwise the state is annihilated).
We neglect one of the fictitious particles on each site,
\[ d_i^\dagger d_i + e_i^\dagger e_i + s_i^\dagger s_i + s_i^\dagger s_i = 1, \quad \forall i. \tag{3} \]

The original Hamiltonian clearly preserves this relationship.

Substituting Eq. (2) into Eq. (1), we have that

\[ H_{\text{Hub}} = -t \sum_{(ij),\sigma} (d_i^\dagger d_j - e_i^\dagger e_j)s_j^\dagger s_j + U \sum_i d_i^\dagger d_i \\
- J \sum_{(ij)} (s_i^\dagger s_j + e_j e_i s_j^\dagger s_i) - \sum_i s_i^\dagger s_i), \tag{4} \]

Note that the first line preserves the number of doublons and holons, whereas the second line does not.

At large \( U \), the second line of Eq. (4) can be treated by perturbation theory in \( t/U \) (starting from states with a definite number of double-occupancies). The method as applied here is standard, and can be found in, e.g., Ref. [61]. We obtain the \( t-J \) model,

\[ H_{tJ} = -t \sum_{(ij),\sigma} (d_i^\dagger d_j - e_i^\dagger e_j)s_j^\dagger s_j + U \sum_i d_i^\dagger d_i \\
- J \sum_{(ij)} (s_i^\dagger s_j + e_j e_i s_j^\dagger s_i) + \sum_i s_i^\dagger s_i), \tag{5} \]

where \( J \equiv 4t^2/U \). Strictly speaking, Eq. (5) should include additional next-nearest-neighbor terms as well as a direct interaction between nearest-neighbor doublons and holons, but these are commonly neglected.

### B. Magnetic ordering

The second line of Eq. (5) is precisely the antiferromagnetic Heisenberg Hamiltonian, expressed in terms of Schwinger bosons (here the spinons \( s_{i\sigma} \)) [61]. We are specifically considering Néel-ordered ground states with positive magnetization on the A sublattice and negative on \( B \). It is, thus, convenient to express \( s_i^\dagger (s_i) \) in terms of \( s_i^\dagger (s_i) \) for \( i \in A (B) \) using Eq. (3)—this is equivalent to using Holstein-Primakoff rather than Schwinger bosons [61],

\[ s_i^\dagger = \sqrt{1 - s_i^\dagger s_i^\dagger} \]
\[ s_i = \sqrt{1 - s_i^\dagger s_i^\dagger} \]

We neglect \( d_i^\dagger d_i \) and \( e_i^\dagger e_i \) in using Eq. (3) because we are interested in the dilute-charge limit. From here on, we will simply write \( s_i \) in place of \( s_i^\dagger (s_i) \) for \( i \in A (B) \). On both sublattices, the \( s_i \) boson represents a fluctuation relative to perfect Néel order.

Note that Eq. (6) cannot be an exact equality because it does not respect the fact that \([s_{i\sigma}, s_{i\sigma}^\dagger] = 1\). It is more of a semiclassical approximation, valid in the limit of large spin \( S \).

Of course, the case \( S = 1/2 \) under consideration here is far from large, but all other analytical techniques of which we are aware for treating long-range order via slave particles (such as Bose-Einstein condensation of the Schwinger bosons [62,63]) have the same regime of validity. We refer to Ref. [61] for more details.

Inserting Eq. (6) into \( H_{tJ} \) does not yield a solvable Hamiltonian on its own. Thus, to progress further, we perform a mean-field approximation by expanding \( H_{tJ} \) to first order in \( s_i^\dagger s_i \) (this is also reasonable in the semiclassical limit). With \( (s_i^\dagger s_i) = 1/2 - m \), where \( m \) is the Néel magnetization, we find that

\[ H_{tJ} \approx -\sqrt{1 + 2m} \frac{t}{2} \sum_{kj} d_k^\dagger d_k (M_{kq} \beta_q + M_{kq}^\dagger \beta_{q}^-) \]
\[ + \sqrt{1 + 2m} \frac{t}{2} \sum_{kj} e_k^\dagger e_k (M_{kq} \beta_q + M_{kq}^\dagger \beta_{q}^-) \]
\[ + \frac{1}{2} \sum_{q} \omega_q \beta_q^\dagger \beta_q, \tag{7} \]

where the sum is over the 2D Brillouin zone (\( N \) is the number of lattice sites) and \( \beta_q \equiv u_q s_q + v_q s_{-q} \) is the transformed spinon operator. With

\[ u_q = \sqrt{\frac{1}{2} \left( 1 + \frac{1}{\sqrt{1 - \gamma_q^2}} \right)}, \tag{9} \]
\[ v_q = \text{sgn}[\gamma_q] \sqrt{\frac{1}{2} \left( 1 - \frac{1}{\sqrt{1 - \gamma_q^2}} \right)}, \tag{10} \]

where

\[ \gamma_q = \frac{1}{2} (\cos q_x + \cos q_y), \tag{11} \]

the frequencies \( \omega_q \) and vertices \( M_{kq} \) entering into Eq. (8) are given by

\[ \omega_q = 4(1 + 2m)J \sqrt{1 - \gamma_q^2}, \tag{12} \]
\[ M_{kq} = 4\gamma_k u_q - 4\gamma_{k+q} v_q. \tag{13} \]

Normally one would determine \( m \) self-consistently from the ground-state spinon occupation: \( 1/2 - m = N^{-1} \sum q \gamma_q^2 \). This is known to give \( m \approx 0.3 \) for a 2D square lattice [45]. We will instead treat \( m \) as an
independent parameter—this is an approximate (albeit crude) means of estimating exciton properties as a function of magnetization.

C. Self-consistent Born approximation

The self-consistent Born approximation (SCBA) gives the doublon-doublon and holon-holon propagators by the integral equation in Fig. 3. The same equation holds for each propagator separately. This approximation is expected to be accurate in the dilute-charge limit, where the charge dynamics is strongly affected by spinons but not vice versa.

In terms of the doublon/holon self-energy $\Sigma_k(\epsilon)$, Fig. 3 translates to (after a frequency integration)

$$\Sigma_k(\epsilon) = \frac{(1 + 2m) \tau^2}{2N} \sum_q \frac{M^2_{kq}}{\epsilon - \omega_q - \Sigma_{k-q}(\epsilon - \omega_q)}. \quad (14)$$

The quasiparticle spectrum $\epsilon_k$ is given by the solution to $\Sigma_k(\epsilon_k) = \epsilon_k$.

Equation (14) can be solved quite efficiently. Note that all $\omega_q$’s are positive [64], thus Eq. (14) in fact expresses $\Sigma_k(\epsilon)$ in terms of the self-energy at lower frequencies. We start at sufficiently negative $\epsilon$, below which we approximate $\Sigma_k(\epsilon) \approx (1 + 2m) \tau^2 / 2N \sum_q M^2_{kq} / (\epsilon - \omega_q)$, and then compute the self-energy at incrementally higher frequencies in terms of the previous values. To help avoid numerical errors, we add a small imaginary part [namely, $0.1(1 + 2m)\tau$] to $\epsilon$.

Although one could proceed using the full $\Sigma_k(\epsilon)$, it has been found that the quasiparticle dispersion can be well approximated by the form [65]

$$\epsilon_k = -2t_1[\cos (k_x + k_y) + \cos (k_x - k_y)]$$

$$- 2t_2[\cos (2k_x) + \cos (2k_y) + 2]. \quad (15)$$

This expression has a clear physical interpretation: $t_1$ is the amplitude for performing a two-step hop along the diagonals of the lattice, and $t_2$ is the amplitude for a two-step hop along the principal axes (see Fig. 4). Thus, in what follows, we will use for the single-particle propagators the simpler expression,

$$G_k(\epsilon) = \frac{1}{\epsilon - (1 - it)\epsilon_k}, \quad (16)$$

with $\epsilon_k$ given by Eq. (15).

D. Bethe-Salpeter equation

We next consider the two-particle Green’s function ($T$ denotes time ordering),

$$G_{j_0,k_0;\epsilon,j',k'(t_d,t_e;\epsilon';t'_d,t'_e)} \equiv \langle T d_{k_d}(t_d)e_{j_d}(t_e)e_{j'_d}(t'_e)^\dagger d_{k'_d}(t'_d) \rangle,$$  \quad (17)

and its Fourier transform $\hat{G}_{k_d,k'_d;j_d,j'_d}(\epsilon_d,\epsilon'_d;\epsilon,e';E)$. Due to translational invariance, $\hat{G}$ depends only on differences in position and time, which we choose to parametrize by the relative coordinates,

$$j \equiv j_d - j_e,$$

$$j' \equiv j'_d - j'_e,$$

$$r \equiv \frac{j_d + j_e}{2} - \frac{j'_d + j'_e}{2},$$ \quad (18)

with relative times defined analogously. The corresponding momenta are

$$k = \frac{k_d - k_e}{2},$$

$$k' = \frac{k'_d - k'_e}{2},$$ \quad (19)

$$K = k_d + k_e = k'_d + k'_e.$$

We will use absolute and relative momenta interchangeably, depending on notational convenience, with Eq. (19) always giving the relationship between the two. $\hat{G}_{k,k';\epsilon,\epsilon'}(\epsilon_d,\epsilon'_d;\epsilon,e';E)$ will often be written as $\hat{G}_{k,k';\epsilon,\epsilon'}(\epsilon,e';E)$.

We determine $\hat{G}$ within the ladder approximation as shown in Fig. 5. Although admittedly uncontrolled, this

FIG. 4. Illustration of hopping parameters $t_1$ and $t_2$ for the approximation to the quasiparticle dispersion in Eq. (15). Singly occupied sites show the background magnetic order in which the double occupancy hops.

FIG. 5. The integral equation which determines the two-particle Green’s function, within the ladder approximation. The hatched square is the Green’s function, and all other symbols are as in Fig. 3.
approximation does conveniently represent the physics of two-particle bound states being mediated by emission and absorption of spin waves. Written out, the integral equation of Fig. 5 is

\[
G_{kk';k'}(\epsilon, \epsilon'; E) = G_k(\epsilon) G_{k'}(\epsilon') \left[ \delta_{kk'} + \frac{(1 + 2m)^2}{2N} \int \frac{d\omega}{2\pi i} \left( \frac{M_{k-q, q} M_{k-q}}{\omega - (1 - i0)\omega_q} - \frac{M_{k-q} M_{k+q, q}}{\omega + (1 - i0)\omega_q} \right) \times G_{k-q, k'}(\epsilon - \omega, \epsilon'; E) \right],
\]

where \(G_k(\epsilon)\) is given by Eq. (16) and the vertices \(M_{kq}\) are as in Eq. (8).

Since our goal is to identify bound states, we reduce Eq. (20) to the Bethe-Salpeter equation. The details of this approach can be found in Ref. [66]. We assume that \(G\) has an isolated pole in the total energy \(E\), near which it has the form

\[
G_{kk';k'}(\epsilon, \epsilon'; E) \sim -i \frac{\psi_k(\epsilon) \overline{\psi}_{k'}(\epsilon')}{E - (1 - i0)E_b},
\]

where the “wavefunction” \(\psi_k(\epsilon)\), its time-reversed partner \(\overline{\psi}_{k'}(\epsilon')\), and the bound-state energy \(E_b\) remain to be determined. Inserting this ansatz into both sides of Eq. (20) and equating the residues at \(E_b\) on each side, we obtain a nonlinear eigenvalue problem (the Bethe-Salpeter equation),

\[
\psi_k(\epsilon) = G_k \left( \frac{E_b}{2} + \epsilon \right) G_k \left( \frac{E_b}{2} - \epsilon \right) \left( 1 + 2m \right)^2 \int \frac{d\omega}{2\pi i} \left( \frac{M_{k-q, q} M_{k-q}}{\omega - (1 - i0)\omega_q} - \frac{M_{k-q} M_{k+q, q}}{\omega + (1 - i0)\omega_q} \right) \psi_{k-q}(\epsilon - \omega).
\]

Here and \(\psi_k(\epsilon)\) is the bound-state wavefunction in a quite literal sense: It is the Fourier transform of \(\langle 0 | d_j(t) \psi_0(0) | b \rangle\), where \(|b\rangle\) denotes the bound state and \(|0\rangle\) denotes the ground state. Note that \(t = 0\) is of particular interest since it gives the amplitude for simultaneously observing the holon at site 0 and the doublon at site \(j\). Thus, to simplify the problem, we integrate Eq. (22) over \(\epsilon\), and furthermore, make the ansatz,

\[
\psi_k(\epsilon) = -G_k \left( \frac{E_b}{2} + \epsilon \right) G_k \left( \frac{E_b}{2} - \epsilon \right) (E_b - \epsilon_{k_d} - \epsilon_k) \Psi_k,
\]

with \(\Psi_k\) independent of \(\epsilon\). The explicit factor of \(E_b - \epsilon_{k_d} - \epsilon_k\) is included so that \(\Psi_k\) is the equal-time wavefunction, i.e., \(\Psi_k = \psi_k(t = 0)\). This ansatz allows us to perform the \(\epsilon\) integral straightforwardly, giving a closed equation for \(E_b\) and \(\Psi_k\),

\[
(E_b - \epsilon_{k_d} - \epsilon_k) \Psi_k = -\frac{(1 + 2m)^2}{2N} \sum_q \left( \frac{M_{k-d-q, q} M_{k-q}}{E_b - \epsilon_{k_d} - \omega_q - \epsilon_k} + \frac{M_{k-d-q} M_{k+q, q}}{E_b - \epsilon_{k_d} - \epsilon_k + \omega_q} \right) \Psi_{k-q}.
\]

Equation (25) is the two-particle Schrödinger equation albeit with an energy-dependent potential. We find the values of \(E_b\) at which it has a nonzero solution and record the corresponding eigenvector.

Strictly speaking, Eq. (24) is not a valid ansatz for \(\psi_k(\epsilon)\), i.e., it does not solve the frequency-dependent Eq. (22). However, it has a clear physical interpretation. The Fourier transform \(\psi_k(t)\) gives the wavefunction for inserting the doublon and holon separated by time \(t\) [see Eq. (23)]. The poles coming from the single-particle propagators in Eq. (24) correspond to the phase factor acquired by the remaining particle during that interval, and our ansatz amounts to neglecting any other time dependence. This approximation has been applied previously to study holon-holon binding [49], and we expect it to be qualitatively accurate for our purposes.

Equation (25) and those preceding it differ from the equations for holon-holon binding in two respects. First, the holon-holon equations must include exchange terms not found here. Second, due to the relative phase between the doublon-spinon and holon-spinon vertices, the effective potential in Eq. (25) would have the opposite sign for the holon-holon problem.

III. RESULTS

A. Single-particle properties

We first review the behavior of individual quasiparticles, determined within the SCBA as described above. Although these calculations have been reported previously, e.g., in Refs. [39,65], it will be useful to reproduce them here.

Figure 6 shows the quasiparticle dispersion throughout the Brillouin zone with the magnetization set to the equilibrium value for concreteness. As noted above, it can be well approximated by a next-nearest-neighbor hopping model with amplitude \(t_1\) for moving along the diagonals of the lattice and amplitude \(t_2\) for moving along the principal axes (see Fig. 4).
The form of the dispersion is not sensitive to the value of magnetization. However, the effective hopping amplitudes, which we determine empirically by fitting the computed spectrum to Eq. (15), do depend on $m$ as shown in Fig. 7.

Some features of the dispersion can be explained by a simple Hartree-Fock (HF) approximation to the original Hamiltonian in which the Hubbard interaction is replaced by $n_i \sigma \langle n_i \bar{\sigma} \rangle + \langle n_i \bar{\sigma} \rangle \sigma$. Assuming Néel order for $\langle n_i \bar{\sigma} \rangle$, the Hamiltonian becomes a tight-binding model on a bipartite lattice with dispersion,

$$\epsilon_k^{(HF)} = \sqrt{U^2 + 4t^2(\cos k_x + \cos k_y)^2} \sim U + \frac{4t^2}{U} (\cos k_x + \cos k_y)^2,$$

using that $t \ll U$. Up to a constant shift, the second line is equivalent to Eq. (15) for the special case $t_1 = 2t_2$. Note, in particular, that $t_1, t_2 < 0$. Thus, Hartree-Fock correctly predicts that the band minimum is within the lines $k_x + k_y = \pm \pi$. It also correctly suggests that the bandwidth should be significantly reduced to $O(J)$ instead of $O(t)$. However, it incorrectly claims that the dispersion is degenerate along the entire magnetic Brillouin-zone boundary. The more sophisticated SCBA resolves this degeneracy, identifying four minima at $(k_x, k_y) = (\pm \pi/2, \pm \pi/2)$.

Recent work on magnetic polarons in the $t$-$J$ model [67,68] has made clear that this behavior can be understood by the charge excitations forming spinon-charge bound states (the “polarons”), held together by strings of displaced spins (much as we have sketched in Fig. 1 but with individual charges). The charges are forced to move on the timescale set by their slower spinon partners, namely, $O(1/J)$, and are subject to the bipartite lattice felt by the spinons. The dispersion results shown above confirm this string picture quite nicely if one interprets them as being for the polaron as a whole. With that in mind, it is rather striking that these three approaches—SCBA, Hartree-Fock, and the string picture—all lead to consistent conclusions.

Returning to Fig. 7, we see that the bandwidth $W$ increases noticeably as the magnetization increases. Equivalently, the single-particle mass decreases. Within the framework of our calculation, the explanation is clear: a doublon/holon can move only if a spinon takes its place [see Eq. (5)], and since one spinon factor is always in the direction of the Néel magnetization, the doublon/holon hopping term is proportional to $\sqrt{1 + 2m}/2$.

**B. Exciton properties**

We now turn to the exciton properties as functions of magnetization, using the Bethe-Salpeter equation. All of the quantities presented here are straightforward to compute from the energy $E_k$ and wavefunction $\psi_k$ given by Eq. (22).

Figure 8 shows the energy of the lowest internal state as a function of the center-of-mass momentum $K$. As was the case for the single-particle dispersion, the shape of the exciton dispersion is not particularly sensitive to the magnetization. Note that the bottom of the band is not at the origin but rather
at \((K_x, K_y) = (\pi, \pi)\). The wavefunction of the \((\pi, \pi)\) state is shown in Fig. 9. We leave for future work a full group-theoretic classification of the exciton wavefunctions based on square-lattice symmetry, including the selection rules for creating Mott excitons, but we expect that it can be carried out straightforwardly—for example, the wavefunction in Fig. 9 has \(s\)-wave symmetry about the point \((\pi/2, \pi/2)\) [which is the high-symmetry point of Eq. (25)].

The binding energy, mass, and radius of the exciton are plotted versus magnetization in Fig. 10. We see that as one increases the magnetization \(m\), the mass decreases whereas the binding energy and size increase. It is interesting to compare these trends with what one would expect for a conventional exciton formed via Coulomb attraction. In that situation, a decrease in mass is associated with an increase in radius and a decrease in binding energy. Here, we find a similar relationship between radius and mass, but the binding energy instead scales inversely with mass.

Equation (25) can be simplified further in the large-\(t/J\) limit. We will see that \(E_b\) scales as \(t\), whereas \(\epsilon_k\) and \(\omega_q\) are asymptotically smaller [65]. Thus, we can neglect the single-particle and spinon dispersions, leaving the equation,

\[
E_b^2 \Psi_k = -\frac{(1 + 2m)t^2}{2N} \sum_q (M_{ki,q}M_{k,q} + M_{ki,q}M_{k,q} - M_{ki,q}M_{k+q,q}) \Psi_k \Psi_q.
\]

(27)

Although still not of the Schrödinger form, Eq. (27) is much simpler to solve than Eq. (25): The kernel on the right-hand side no longer depends self-consistently on the energy (and as claimed, \(E_b \sim t\)). The results obtained from the large-\(t/J\) equation are plotted alongside the others in Fig. 10.

As is clear from Eq. (25), the spinon-mediated interaction between charges is not instantaneous. To assess the importance of this retardation, we have compared the results in Fig. 10 to what would be obtained through the static approximation [setting \(\omega = 0\) in the kernel of Eq. (22)]. The static approximation would predict significantly different results as
FIG. 11. Comparison between the full results and the two approximations considered in the text. Data shown are for $t/J = 2$ and a lattice of size $32 \times 32$. Vertical dashed lines indicate the equilibrium value of $m$. For the fixed-mass points, the values of $t_1$ and $t_2$ are set to their values at $m = 0.5$ (see Fig. 7).

seen in Fig. 11: The binding energy would instead decrease slightly with magnetization and the mass would increase slightly. Thus, the retardation of the effective interaction is an essential ingredient to the behavior seen here.

Similarly, one can ask whether the trends observed in Fig. 10 are due primarily to changes in the spinon behavior or rather due to the single-particle mass, which itself decreases with magnetization. We have repeated the above calculations under “fixed-mass” conditions in which the single-particle parameters $t_1$ and $t_2$ are kept fixed (to their values at $m = 0.5$) as we vary the magnetization. Figure 11 shows that each of the three observables responds differently. The binding energy becomes more sensitive to magnetization, indicating that the quasiparticle and spinon properties play antagonistic roles. On the other hand, the exciton mass becomes less sensitive—the change to the effective interaction suppresses the mass by itself. Finally, the exciton radius shows the reverse behavior to before, instead decreasing with magnetization (although the size remains quite small in absolute terms).

Certain trends seen here are somewhat surprising. For example, we find that the exciton radius increases with increasing magnetization, but one might have expected it to decrease. The fixed-mass calculation provides a partial explanation: the spin waves on their own do, in fact, force the charges closer together at larger magnetization, but this effect is overcompensated by each individual charge becoming lighter. Whether the charges becoming lighter is itself a real effect or rather an artifact of our approximations is less clear, and something that warrants further investigation.

It is also surprising that the exciton binding energy does not vanish as $J/t \to 0$ since the binding is mediated by spin waves in the first place. Although more systematic studies are clearly needed, this does not strike us as necessarily contradictory. Keep in mind that we are considering the behavior of a single doublon-holon pair on top of an antiferromagnetically ordered background. Setting $J = 0$ does not change the fact that our initial state is ordered, nor that the motion of charges disturbs that order—in the slave-particle language, the spinons are inert at $J = 0$, but the hopping of a charge does still leave a string of spin excitations in its wake [see Eq. (7)]. It is conceivable that interference effects analogous to Nagaoka ferromagnetism [61,69] [whereby background ferromagnetic order lowers the energy of a single hole by $O(t)$ even at $J = 0$] could bind pairs of charges together in an antiferromagnetic background as well, implying $E_b = O(t)$. Further investigation is certainly warranted, however.

One final surprise is that the binding energy does not vanish at $m = 0$ either, but this we fully expect to be an artifact of our approximations. We have treated the magnetic order along the lines of spin-wave theory—this is most justified at full magnetization and seems to work well at the equilibrium magnetization [45] but fails at $m = 0$. We do not see the failure at $m = 0$ as invalidating the trends found for larger magnetization, however.

The recent pump-probe experiments in Refs. [29,30] have investigated how excitons are influenced by magnetic order in the Mott insulator Na$_2$IrO$_3$. Our results support their interpretation in some aspects but not in others. In Ref. [29], the authors observe an increase in the fraction of bound excitations when below the Néel temperature, which they attribute to an increase in the exciton binding energy. Figure 10 shows that magnetic order does indeed increase the binding energy. On the other hand, Ref. [30] demonstrates that the relaxation dynamics following a pump are slower in the presence of magnetic order. This is attributed to the mass increasing with magnetization, yet we have observed the opposite (consistent with past works calculating the dependence on $J/t$ [60,65,70]). Given the highly nonequilibrium nature of the experiments as well as the approximations inherent in an analytical approach, further investigation is clearly needed.

Lastly, let us compare the present calculation of doublon-holon binding to that of holon-holon binding, which is obviously of significant interest in its own right [58,71]. Clearly the two have much in common, yet there are two important differences. First, the integral equation which determines the two-particle Green’s function (Fig. 5) has an additional exchange term due to the indistinguishability of the holons. Second, even the direct term comes with an extra minus sign, i.e., the effective interaction is of opposite sign. The sign can be removed by redefining the hole operator on one sublattice, but the additional phase may modify further
results depending on the application. It is important to keep these distinctions in mind when relating the present results to the high-$T_c$ literature.

IV. CONCLUSION

We have studied the role that magnetic order plays in the formation of excitons within Mott insulators, using the Hubbard model as a concrete (albeit simplified) Hamiltonian. The binding energy increases in the presence of antiferromagnetic magnetization, whereas the exciton mass decreases. The size of the exciton increases slightly, yet the radius is never more than a lattice spacing. Using the standard classification, these are Frenkel excitons regardless of magnetic order.

In addition, we have established that the trends observed here require a detailed understanding of the many-body dynamics in these systems. Retardation effects in the effective spinon-mediated interaction are essential. Furthermore, the constituent charge and spin excitations are each affected separately by the background magnetic order in ways cooperative for some exciton properties but antagonistic for others.

It must be noted that despite the complexity, there are significant limitations to our approach. In particular, we have made approximations in the spirit of linear spin-wave theory, which is only justified at large spin $S$ and full magnetization (neither of which we assume here). Thus, we do not expect these results to be quantitatively accurate—we instead view this analysis as expressing our physical intuition regarding Mott excitons in the language of slave particles, from which we can make sharp predictions to be verified or falsified by more systematic investigations.

As an outlook, the predictions made here will be important when analyzing recent and future experiments on the optical properties of strongly correlated electronic materials. The existing experiments are quite complex and require interpretations of their own. Our results agree with those interpretations in some respects but disagree in others. A complete understanding of the systems will require many approaches both experimental and theoretical, including but not limited to the one described here.

Particularly promising are the recent experiments on fermionic atoms in optical lattices [41,42,44]. Since ultracold gases do not have many of the complicating features found in condensed-matter systems, we expect that this will be a valuable direction to explore further. It is also likely that our conclusions, being based on the single-band nearest-neighbor Hubbard model, are more applicable to those systems than to materials such as the iridates. Importantly, current quantum gas microscopes allow one to directly create localized doublons and holons via optical tweezers and reliably measure the spin correlation functions [43]. Such an unprecedented direct access to the system microscopics will provide a powerful way of investigating many-body excitons.

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