Spin liquid phases of large-spin Mott insulating ultracold bosons

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Mott insulating ultracold gases possess a unique whole-atom exchange interaction which enables large quantum fluctuations between the Zeeman sublevels of each atom. By strengthening this interaction—either through the use of large-spin atoms or by tuning the particle-particle interactions via optical Feshbach resonance—one may enhance fluctuations and facilitate the appearance of the long-sought-after quantum spin liquid phase—all in the highly tunable environment of cold atoms. To illustrate the relationship between the spin magnitude, interaction strength, and resulting magnetic phases, we present and solve a mean-field theory for bosons optically confined to the one-particle-per-site Mott state, using both analytic and numerical methods. We find on square and triangular lattices for bosons of hyperfine spin $f > 2$ that making the repulsive s-wave scattering length through the singlet channel small—relative to the higher-order scattering channels—accesses a short-range resonating valence bond (s-RVB) spin liquid phase.

I. INTRODUCTION

Quantum spin liquids—insulating magnetic phases which remain disordered down to absolute zero temperature—have attracted great interest in the nearly 30 years since Anderson suggested an intimate relation between the cuprate superconductors and the resonating valence bond state [1,2]. While theorists now have a good understanding of the topological orders and fractionalized excitations that characterize these systems [3,4], the experimental realization of the spin liquid phase has remained a challenge, and despite success in recent years [5–7] the pool of spin liquid candidate materials remains small. The primary difficulty lies in finding systems with sufficiently large spin fluctuations, and to achieve this in the solid state—where electron exchange mediates the spin-spin interactions—one must restrict the search to low-dimensional, geometrically frustrated, spin-1/2 antiferromagnets [8–12]. We believe, however, that by fundamentally broadening our search to include other novel systems, we may bypass these restrictions and expedite the study of this long-sought-after phase.

In particular, Mott insulating ultracold atoms may provide an alternate route to the experimental realization of spin liquids. The spin degree of freedom remains unfrozen in these optically confined systems [13], and the virtual exchange of a whole atom mediates the low-energy spin-spin interaction, as illustrated by Fig. 1. Counter-intuitively, whole-atom exchange produces fluctuations that increase with the atomic hyperfine spin f [14], in dramatic contrast to the solid state, where large spins actually suppress the effect of fluctuations. This peculiar behavior may cause large-f Mott insulators to exhibit many exotic phases [15–20], including various atomic spin liquids. While efforts have focused thus far on SU(N)-symmetric alkaline-earth-metal atoms [21,22], whole-atom exchange should induce large fluctuations generically, potentially bringing the spin liquid phase to life in a wide variety of cold atomic systems. Additionally, the recently developed spin gradient

demagnetization technique [23] provides access to the entropy scales where magnetic ordering is expected, making us hopeful that in the near future experimentalists may directly probe these strongly correlated magnetic phases.

In this paper, we perform a mean-field theory for spin-f bosons on a square optical lattice, tightly confined to the Mott insulating state. We choose to include only the fundamental low-energy scattering interactions, described by a set of s-wave scattering lengths a_F [24] and tuned via microwave and optical Feshbach resonances [25–28]. We then show that our model supports the existence of a short-range resonating valence bond ground state, for certain values of a_F and f. At present, we omit both anisotropic dipole-dipole interactions and gauge fluctuations of the mean field, though we briefly discuss their effects in later sections of this paper. Despite these omissions in our model, the results indicate that whole-atom exchange may melt magnetic order and stabilize spin liquid phases in a much broader class of systems than currently under investigation.

II. GENERAL SPIN-f MODEL

We first write a model that captures the physics of whole-atom exchange, while fluidly describing bosons of different spin. To do this we begin with the spin-f Bose-Hubbard model [29,30],

$$\hat{H} = -t \sum_{\langle i,j \rangle} \sum_{m=-f}^{f} (\hat{b}_{i,m}^{\dagger} \hat{b}_{j,m} + \text{H.c.})$$
$$+ U \sum_{i} \sum_{F=0,2,\ldots}^{2f} a_F \hat{P}_i^F, \qquad (1)$$

where i ranges over all N lattice sites, and $\langle i,j \rangle$ denotes a sum over all distinct nearest-neighbor pairs. We have written the kinetic contribution—parametrized by t—in terms of $\hat{b}_{i,m}^{\dagger}$ ($\hat{b}_{i,m}$) operators, which create (annihilate) a spin-f boson at site i with magnetic quantum number m, while we have expressed the on-site interaction—parametrized by U—in terms of projection operators $\hat{P}_i^F = \sum_{M=-F,\ldots,F} |F,M\rangle \langle F,M|$, which
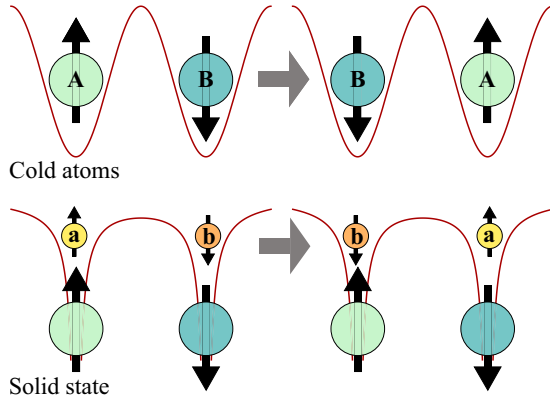
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FIG. 1. Comparison of superexchange mechanisms. For atomic gases optically confined to the Mott state (top), superexchange of whole spin-f atoms leads to magnetic fluctuations on the order of $2f$. In the solid state by contrast (bottom), electron superexchange restricts fluctuations to order 1, which may be small compared to the total spin on each site. Therefore, large-spin cold atoms could potentially realize exotic fluctuation-driven states not accessible with solid state systems. Figure adapted from Ref. [14].

project the two-particle states of site i into the subspace with total angular momentum F. As mentioned previously, a_F denotes the s-wave scattering length through the total angular momentum channel F. Throughout this paper sums over F and M imply a range of $F = 0, 2, \ldots, 2f$ and $M = -F, \ldots, F$, where the absence of the odd-F scattering lengths ensures proper particle statistics [31]. Furthermore, we only consider monotonically increasing repulsive interactions, such that $a_F > 0$ for all F, and $a_{F'} \geqslant a_F$ for $F' > F$. In this regime, antiferromagnetic interactions dominate—a necessary condition for the nontrivial magnetic ordering that we seek.

To move to the deep Mott limit, one quenches the kinetic energy relative to the on-site repulsion [32] ($t \ll U$), allowing a perturbative expansion of Eq. (1) to second order in t/U. By doing so, we obtain a spin-spin interaction in the one-particle-per-site Hilbert space, which in agreement with Refs. [33,34] yields

$$\hat{H} = -J \sum_{\langle i,j \rangle} \sum_{F} \frac{1}{a_F} \hat{P}_{i,j}^{F}, \qquad (2)$$

where the exchange energy is set by $J = 4t^2/U$, and the projection operator $\hat{P}_{i,j}^{F}$ now projects two sites i and j into total angular momentum state F. The natural decoupling of the interaction into total angular momentum channels, each parametrized by a scattering length a_F, arises from the rotational symmetry of the low-energy interaction, which conserves the total angular momentum of two bosons during a collision [24].

At this point, one commonly reexpresses the $\hat{P}_{i,j}^{F}$ operators of Eq. (2) in terms of operators which possess a more direct physical interpretation, such as a polynomial in the Heisenberg coupling $\hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j$, or with tensor operators of increasing rank. These methods do not move fluidly from one spin f to another, however, as one must continually define new operators upon increasing the spin. Although schemes have been developed to simplify such descriptions [35,36], we instead elect to

return our Hamiltonian to second quantization. While doing so simplifies study for generic f, it implicitly enlarges our Hilbert space to include multiply occupied sites. Therefore, to maintain equivalence between the two Hilbert spaces we must impose a one-particle-per-site constraint on average [37]. With these considerations in mind, we write our Hamiltonian in second quantization as

$$\hat{H} = -J \sum_{\langle i,j \rangle} \sum_{F,M} \frac{1}{a_F} \hat{A}_{i,j}^{FM\dagger} \hat{A}_{i,j}^{FM} + \sum_{i} \lambda_i (\hat{n}_i - 1), \quad (3)$$

where we enforce the constraint with a site-dependent Lagrange multiplier λ_i and the number operator for site i, given by $\hat{n}_i = \sum_m \hat{b}_{i,m}^{\dagger} \hat{b}_{i,m}$. The $\hat{A}_{i,j}^{FM\dagger}$ ($\hat{A}_{i,j}^{FM}$) pair operators create (annihilate) a pair of bosons on sites i and j in total angular momentum state $|F,M\rangle$, and relate to the projection operators via $\hat{P}_{i,j}^{F} = \sum_M \hat{A}_{i,j}^{FM\dagger} \hat{A}_{i,j}^{FM}$. Writing these pair operators in terms of the boson operators yields the relation $\hat{A}_{i,j}^{FM} = \sum_{m,n} C_{m,n}^{FM} \hat{b}_{i,m} \hat{b}_{j,n}$, where the presence of the Clebsch-Gordan coefficients $C_{m,n}^{FM} = \langle f,m; f,n|F,M\rangle$ ensures that the pair operators rotate irreducibly as an object with angular momentum F. In the form of Eq. (3), we can write the Hamiltonian for a given atomic hyperfine spin f by simply including the Clebsch-Gordan coefficients through the even-F pairing channels, up to $2f$. The straightforward calculation of these coefficients then provides for a much simpler study at large f.

Next, we mean-field-decouple the pairing operators $\hat{A}_{i,j}^{FM}$, first by expanding about the ground state expectation values $Q_{i,j}^{FM} = \langle \hat{A}_{i,j}^{FM} \rangle$, and then dropping terms of second order in the fluctuations $\delta \hat{A}_{i,j}^{FM}$. This reduces the Hamiltonian to a quadratic form, given by

$$\hat{H} = -\sum_{\langle i,j \rangle} \sum_{F,M} \frac{1}{\bar{a}_F} \left(Q_{i,j}^{FM} \hat{A}_{i,j}^{FM\dagger} + Q_{i,j}^{FM*} \hat{A}_{i,j}^{FM} \right.$$
$$\left. - |Q_{i,j}^{FM}|^2 \right) + \sum_{i} \lambda_i (\hat{n}_i - 1), \qquad (4)$$

where henceforth $J = 1$, and $\bar{a}_F = a_F/a_0$ denotes the scattering length of the F channel relative to the singlet channel, $F = 0$. As our interest lies in translationally invariant states, we demand bond-independent mean fields, such that $Q_{i,j}^{FM} = Q^{FM}$. The phase of the complex Q^{FM} fields remains a $U(1)$ gauge freedom of the problem, and while gauge fluctuations may have important effects on spin liquid mean-field theories [37], we do not consider them in the present approach.

For the investigation of spin liquid phases, as well as the study of atomic superconductors with nontrivial Cooper pairing [38], we note that decoupling the $\hat{A}_{i,j}^{FM}$ operators of Eq. (3) proves more convenient than the single-mode approximation used in the study of spinor Bose-Einstein condensates [31]. In fact, our mean-field Hamiltonian (4) allows direct competition between exotic paired states described by the $Q_{i,j}^{FM}$ fields and the spinor BEC phases described by the boson field $\langle \hat{b}_{i,m} \rangle$. In this way, our mean-field theory may reproduce the results of the well studied spinor BEC mean-field theories while also allowing for spin liquid ground states. The outcome of this competition depends fundamentally on the strength of magnetic fluctuations, as a spin liquid state will only

appear when sufficiently large spin fluctuations have melted the magnetic order of the spinor condensate phase.

III. s-RVB ANSATZ

To directly uncover a spin liquid phase in our model, Eq. (4), we retain only the order parameter of the short-range resonating valence bond (s-RVB) state—namely, an isotropic nearest-neighbor pairing amplitude through the singlet channel. In a pure s-RVB spin liquid state, all $F > 0$ pairing channels have zero amplitude, and the spins exist in an equal superposition of nearest-neighbor singlets. A ground state of this type preserves spin rotational and translational symmetry, making it one of the simplest spin liquid mean-field theories possible for this model. Furthermore, we may reach this s-RVB limit by taking $a_0 \to 0$ with $a_{F>0}$ fixed, since we then have $\bar{a}_F^{-1} \to 0$ for all $F > 0$, and we see from Eq. (4) that only the Q^{00} pairing contributes. The ability to access this limit by tuning a single parameter (a_0) may prove crucial to the eventual realization of this phase experimentally.

We employ the s-RVB ansatz explicitly in our formalism by substituting $\langle \hat{A}_{i,j}^{FM} \rangle = Q^{00}\delta_{F,0}$ in Eq. (4), where the bond-independent complex number Q^{00} represents the s-RVB order parameter. Due to the equivalence of each site by symmetry we require a translationally invariant constraint, so that $\lambda_i = \lambda$. Assuming periodic boundary conditions, we then exploit the lattice translational symmetry by Fourier-transforming the bosons via $\hat{b}_{i,m} = \sum_k \hat{b}_{k,m} e^{ik \cdot R_i}/\sqrt{N}$, where the sum runs over all wave vectors k in the first Brillouin zone, R_i denotes the Bravais lattice vector of site i, and N gives the total number of sites in the lattice.

Introducing the spinor $\Psi_{k,m}^T = (\hat{b}_{k,m}, \hat{b}_{-k,-m}^\dagger)$ allows us to compactly write the Fourier-transformed Hamiltonian as

$$\hat{H} = \sum_{k,m} \Psi_{k,m}^\dagger h_{k,m} \Psi_{k,m}$$
$$- \lambda N \frac{(2f+1)}{2} + \frac{ZN|Q^{00}|^2}{2}, \quad (5)$$

where we have defined the 2×2 matrix

$$h_{k,m} = \begin{pmatrix} \lambda/2 & -\epsilon_k Q^{00} C_{m,-m}^{00} \\ -\epsilon_k Q^{00} C_{m,-m}^{00} & \lambda/2 \end{pmatrix}, \quad (6)$$

where we have fixed the gauge such that Q^{00} is real. The lattice contribution to the Fourier transform yields $\epsilon_k = \sum_{\langle (i)j \rangle} e^{ik \cdot (R_j - R_i)}$, while $\langle (i)j \rangle$ denotes a sum over the Z nearest neighbors j of an arbitrary site i. Throughout this paper we explicitly consider a 2-dimensional square lattice with a lattice spacing of unity, for which $Z = 4$ and $\epsilon_k = 2(\cos[k_x] + \cos[k_y])$. However, we mention that the approach for triangular lattices follows similarly and produces qualitatively similar results to the square in all instances which follow.

The Clebsch-Gordan coefficients in Eq. (6) play a crucial role in writing a 2×2-dimensional Hamiltonian for all spin f. Primarily, the condition of the Clebsch-Gordan coefficients, that $C_{m,n}^{FM} = 0$ unless $m + n = M$, implies that the s-RVB state, which requires $M = 0$, retains only terms in which $n = -m$. In other words, the matrix elements of our s-RVB Hamiltonian only ever connect a spin state m with the corresponding state $-m$, a fact which leads to the chosen form

of the spinor $\Psi_{k,m}^T$. Compared to the general case of Eq. (4), where we need a $2(2f+1) \times 2(2f+1)$-dimensional matrix at each f, the s-RVB ansatz produces a dramatic mathematical simplification.

We now seek the ground state of the s-RVB Hamiltonian (5) in the presence of the one-particle-per-site constraint. Following the methods of Ref. [39], we move to the basis of collective excitations $\hat{\gamma}_{k,\mu}$ by finding a linear transformation $M_{k,m}$ which diagonalizes the matrix $h_{k,m}$ while preserving the bosonic commutation relations, $[\hat{\gamma}_{k,\mu}, \hat{\gamma}_{k',\mu'}^\dagger] = \delta_{k,k'}\delta_{\mu,\mu'}$ and $[\hat{\gamma}_{k,\mu}, \hat{\gamma}_{k',\mu'}] = 0$. After diagonalizing in this way, we find that the collective excitations possess a dispersion given by $\omega_{k,m} = \sqrt{(\lambda/2)^2 - |\epsilon_k Q^{00} C_{m,-m}^{00}|^2}$ for the band corresponding to magnetic sublevel m, and we note that the m-independence of $|C_{m,-m}^{00}| = 1/\sqrt{2f+1}$ forces complete degeneracy among these $2f+1$ bands. Additionally, on a square lattice the dispersion takes a minimum value at $k = (0,0)$ and $k = (\pi,\pi)$, and the value of $\omega_{k,m}$ at these points defines the energy gap $\Delta = \sqrt{(\lambda/2)^2 - 16|Q^{00}|^2/(2f+1)}$. This gap will play a crucial role in the thermodynamic ground state analysis to come.

IV. RESULTS OF THE s-RVB ANSATZ

To determine the ground state of the s-RVB Hamiltonian in the thermodynamic limit ($N \to \infty$ with N/V fixed) we solve the self-consistent equation, $Q^{00} = \langle \hat{A}_{i,j} \rangle$, in the presence of the constraint, $n_i = \langle \hat{b}_{i,m}^\dagger \hat{b}_{i,m} \rangle = 1$. Writing the constraint in terms of the dispersion $\omega_{k,m}$ yields

$$n_\gamma + (2f+1) \int \frac{d^2k}{(2\pi)^2} \frac{1 - \bar{\omega}_k^2}{2(\bar{\omega}_k^2 + \bar{\omega}_k)} = 1, \quad (7)$$

where we define $\bar{\omega}_k = 2\omega_{k,m}/\lambda$ to clean up the notation a bit, and n_γ denotes the condensate fraction of collective excitations, created by the $\hat{\gamma}_{k,\mu}$ operators, in states with energy Δ. With gapped excitations ($\Delta > 0$), it costs finite energy to occupy these minimum energy states, and so the system prefers $n_\gamma = 0$.

By numerically solving the constraint (7) we find two scenarios shown in Fig. 2. The first occurs for $f \geqslant 3$, where one may satisfy the constraint with $\Delta > 0$. The gapped excitations imply that the condensate fraction is zero, and so the Q^{00} field characterizes the state completely, making it a realization of a pure s-RVB spin liquid. On the other hand, for $f \leqslant 2$ one cannot satisfy the constraint with a gap, implying $\Delta = 0$ at $k = (0,0)$ and $k = (\pi,\pi)$. The collective excitations condense at these points, and so $n_\gamma \neq 0$. This restores the
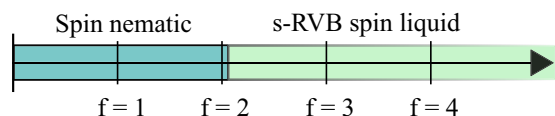

FIG. 2. Phase diagram of the s-RVB ansatz ($\alpha_{F>0} \to \infty$) as a function of f. For $f \leqslant 2$ the ground state is a spin nematic with $\langle \mathbf{S} \rangle = 0$ and $\langle S_x^2 \rangle = \langle S_y^2 \rangle \neq \langle S_z^2 \rangle$ on each site. For $f > 2$ however, degeneracy of the magnetic sublevels enhances fluctuations, and the ground state becomes a short-range resonating valence bond (s-RVB) spin liquid.

validity of the constraint, but one must now describe the state with a spinor of condensate parameters $\langle \hat{b}_{k,m} \rangle$, in addition to the Q^{00} field. A spinor of this type breaks spin rotational symmetry, implying a magnetically ordered ground state for $f \leqslant 2$. At present, the spinor contains equally weighted m and $-m$ pairs, and so possesses nematic symmetry with $\langle \hat{\mathbf{S}}_i \rangle = 0$ on each site.

The fact that for small spin we have a magnetically ordered spin nematic ground state, while for large spin we have a disordered spin liquid phase, results directly from the increasing number of magnetic sublevels as one moves to large f. We understand this by noting that the integral in Eq. (7),

$$\langle \hat{n}_{i,m} \rangle = \int \frac{d^2 k}{(2\pi)^2} \frac{1 - \bar{\omega}_k^2}{2(\bar{\omega}_k^2 + \bar{\omega}_k)} \leqslant 0.19 \ldots,$$

corresponds to the contribution from the noncondensed bosons of the m band, $\langle n_{i,m} \rangle$, and has a maximum value $\simeq 0.19$ when $\Delta = 0$. Degeneracy of the $2f + 1$ bands then implies that for $\Delta > 0$ we can write $\langle \hat{n}_i \rangle < 0.19(2f + 1)$, and so to satisfy $\langle \hat{n}_i \rangle = 1$ for finite Δ, we must have $f > 2$.

Again, we emphasize that these fluctuation-driven states result directly from the increasing number of magnetic sublevels as one moves to larger spin. The enlarged space through which the spins may interact enhances the fluctuations, melting magnetic order and driving the system into an s-RVB spin liquid phase. Despite these positive results, to better describe the atomic species used in cold atom experiments we must study the more general problem, which allows scattering through the $F > 0$ angular momentum channels.

V. NEMATIC ANSATZ

The most general case of the mean-field Hamiltonian (4) allows scattering through all total angular momentum channels, $F = 0, 2, \ldots, 2f$. However, with all Q^{FM} fields allowed, the increasing size of the interaction space at large f makes the Hamiltonian increasingly cumbersome to solve numerically. So to efficiently probe the large-f behavior as a function of the scattering lengths a_F, we retain only the order parameter of the spin nematic state, given by $\langle \hat{A}_{i,j}^{FM} \rangle = Q^{F0} \delta_{M,0}$. With this set of mean fields each site will have $\langle \hat{\mathbf{S}} \rangle = 0$ and $\langle \hat{S}_x^2 \rangle = \langle \hat{S}_y^2 \rangle \neq \langle \hat{S}_z^2 \rangle$—the symmetry of a spin nematic. Fortunately, since this retains only the $M = 0$ pairing, we may again write the Hamiltonian with the 2-dimensional spinor $\Psi_{k,m}^T = (b_{k,m}, b_{-k,-m}^\dagger)$. This again allows us to diagonalize in a straightforward manner, but we omit the details due to their similarity with the s-RVB case covered previously.

We motivate this ansatz by extending our results from the s-RVB ansatz, as shown in Fig. 2, and by looking to the phase diagram for the spin 1, 2, and 3 spinor Bose-Einstein condensates [24,33,40–47]. We find that our region of interest, parametrized by $a_F > 0$ for all F and $a_{F'} \geqslant a_F$ for $F' > F$, lies entirely within the nematic sector of these phase diagrams. Additionally, our ansatz consists of a linear combination of the uniaxial and biaxial spin nematic states, which are known to possess an accidental degeneracy at mean-field level [48,49]. Thus, it provides a suitable trial state for our specific parameter regime, capable of describing both a nematic spinor condensate and a Q^{00}-only s-RVB spin liquid phase. We therefore proceed

with this ansatz with the belief that our results describe the physically accessible states of the general Q^{FM} model (4).

While we have simplified the determination of the ground state for a general set of scattering lengths, a difficulty remains in how to best present the results graphically. The number of scattering lengths grows as $f + 1$, which on a phase diagram would require the introduction of an additional axis at each f. To avoid this we seek an approximation which describes the various scattering lengths with a single parameter. Guided by the s-RVB case, where we found that $a_0 \to 0$ favors the singlet pairing and induces an s-RVB spin liquid, we shall use the following approximation for the relative scattering lengths $\bar{a}_F = a_F / a_0$,

$$\bar{a}_F = \begin{cases} \alpha & \text{for } F > 0, \\ 1 & \text{for } F = 0, \end{cases} \tag{8}$$

where scattering lengths through nonzero angular momentum channels have equal magnitude, and differ from a_0 through the proportionality factor α. Varying α from 1 to ∞ covers our original range of the scattering lengths—$a_F > 0$ and $\bar{a}_F \geqslant 1$ for all F—while the $\alpha \to \infty$ limit recovers Eq. (5) directly. While in real atomic systems the $\bar{a}_{F>0}$ are not generically equal, they effectively appear so when compared to a_0 in the $\alpha \to \infty$ limit, making this approximation especially useful for describing the spin liquid phase. Most importantly, we may now construct a phase diagram as a function of f and α, since α is a parameter common to all spin f.

We note that applying Eq. (8) takes us to an enhanced symmetry point of the original Hamiltonian (3)—namely, the bosonic analog to Wu's hidden symmetry found in large-spin Fermi gases [16,50]. Our results do not depend on this symmetry however, and we may show this by using the alternate approximation, $\bar{a}_F = \alpha' F + 1$, where the scattering lengths have a linear relationship with slope α'. This approximation does not generically possess symmetry higher than SU(2), yet the results obtained coincide qualitatively with the results outlined in the next section using Eq. (8). The qualitative similarity stems from the fact that in each case, increasing α or α' effectively takes $a_0 \to 0$, and so the singlet pairing dominates—the crucial condition for obtaining a spin liquid phase in this model.

VI. RESULTS OF THE NEMATIC ANSATZ

Figure 3 shows the phase diagram for $\alpha = [1, \infty)$ and $f = 1, 2, \ldots, 13$. For $f \leqslant 2$ the system always forms a nematic condensate, in agreement with our s-RVB solution in the $\alpha \to \infty$ limit. On the other hand, for $f > 2$ the system moves into the spin liquid phase for α greater than some critical value α_C. As we move to large f, we find that α_C decreases and the spin liquid region grows in size. Again, decreasing a_0 relative to the other scattering lengths increases α, and so by tuning a single parameter one may access the spin liquid phase for $f > 2$ atoms.

We describe the behavior of the spin liquid region as follows. Increasing α (or α') biases the system towards singlet pairing, which causes equal occupation of the Zeeman sublevels and maximizes magnetic fluctuations. Additionally, moving to large f increases the number of available magnetic sublevels, also enhancing fluctuations [14]. The shape of
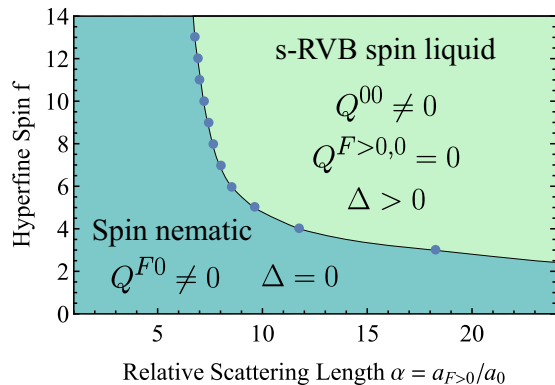
FIG. 3. Phase diagram on the square lattice of the nematic ansatz as a function of f and α, where $\alpha = a_{F>0}/a_0$ parametrizes the relative scattering lengths, as introduced by Eq. (8). The fluctuations responsible for the spin liquid state are enhanced by the increasing number of magnetic sublevels as one moves to large f, and by increased scattering through the singlet channel ($F = 0$) as one moves to large α. For $f \leqslant 2$ the system is nematic for all α, while for $\alpha = 1$ the spin liquid phase is not accessed at any f. The phase diagram for the triangular lattice is qualitatively similar in that the s-RVB region expands to smaller values of α, but condensate order still occurs for $f \leqslant 2$ at all interaction strengths.

the spin liquid region as shown in Fig. 3 results from the cumulative effect of these two scenarios. For $f \leqslant 2$, too few sublevels contribute to produce the necessary fluctuations, regardless of any biasing towards the singlet channel. With $f > 2$ but still small, the system requires strong biasing to reach the spin liquid phase, while at large f, the multitude of participating sublevels allows access to the spin liquid phase with small biasing. In light of this, large-spin atoms would require the least experimental tuning necessary to obtain the long-sought-after spin liquid phase.

VII. EXPERIMENTAL ACCESSIBILITY

Upon inspection of the "untuned" scattering lengths, as conveniently compiled in Ref. [31] for the commonly used atoms—^{87}Rb and ^{23}Na with $f = 1,2$, and ^{52}Cr with $f = 3$—we see that a_0 and the smallest $a_{F>0}$ have roughly the same order of magnitude. In each case, this places them near the $\alpha = 1$ region of Fig. 3, and predicts a spin nematic ground state in agreement with previous theoretical work [24,33,40–47,49]. However, upon tuning a_0 to small enough values via optical Feshbach resonance, a transition to the spin liquid phase may occur. We note that this transition may even occur for $f \leqslant 2$ atoms as well, since fluctuations beyond mean-field theory may actually enlarge the spin liquid region.

The relative contribution from the $F > 0$ pairing channels,

$$Q_R = \frac{\sum_{F>0} |Q^{F0}|^2}{\sum_{F \geqslant 0} |Q^{F0}|^2}, \tag{9}$$

represents a potential order parameter for the spin liquid–to–spin nematic phase transition. Figure 4 shows the behavior of this quantity for an $f = 3$ system when tuned across α. This quantity is similar to the singlet fraction measured in Ref. [51], and may allow the observation of a spin liquid
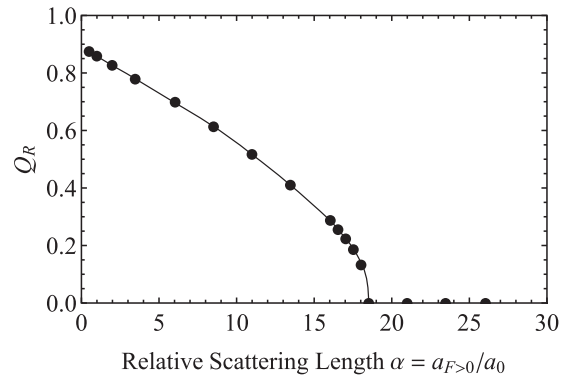

FIG. 4. Relative contribution from the higher-order scattering channels ($F > 0$) for an $f = 3$ system, as captured by the Q_R parameter defined in Eq. (9). In the spin liquid phase ($\alpha \gtrsim 18$) all pairings except the singlet pairing Q^{00} are negligible, while in the spin nematic phase ($\alpha \lesssim 18$) pairing through the nonzero angular momentum channels becomes relevant. Measurement of this parameter could distinguish between the phases.

phase experimentally. Additionally, by spatially resolving vortices in the $Q_{i,j}^{FM}$ fields via photoassociation intensity experiments [52–56] one may investigate vison excitations in the system, in a similar manner to the "vison experiment" conducted by Moler and collaborators for high-T_C cuprate superconductors [57]. Overall, the increasingly varied techniques used in the preparation and characterization of cold atomic systems may provide several avenues for the eventual observation of these novel states.

We expect that the primary challenge remaining in the quest to observe large-f spin liquids will be to overcome the anisotropic dipole-dipole interactions. These interactions grow with the spin as f^2, making them particularly problematic for large spin, where we expect spin liquid phases to be competitive ground states. For example, several recently trapped isotopes of dysprosium have an extremely large spin of $f = 7$ and 8, and the dipole-dipole interactions in these systems are believed to affect the ground state physics in a nonperturbative way [58]. Essentially, the dipole-dipole energy sets a lower bound on the allowed tuning of a_0, below which our approximation breaks down and we must account for these interactions explicitly [59]. Fortunately, for $f = 3$ chromium, the dipole-dipole interactions do not significantly affect the ground state physics [24], allowing use of our mean-field description (4).

VIII. CONCLUSIONS

In this paper we considered an isotropic mean field theory which captures the competition between an s-RVB spin liquid and magnetically ordered condensates. To conclude our discussion, we will now briefly comment on the existence of additional phases and the effect of gauge fluctuations and lattice frustration beyond mean-field theory. First, phases such as the dimer or plaquette phases are also possible, and in future studies non-mean-field calculations, such as quantum Monte Carlo or DMRG, may be used to determine the true ground state phase diagram for this model. Second, the interplay between gauge fluctuations and lattice frustration likely plays a key role beyond mean-field theory, as expected from studies

of similar large-*N* models on square, triangular, and kagome lattices [39,60]. Particularly, on the square lattice we expect that gauge fluctuations make the spin liquid phase unstable to forming a dimer phase, while on frustrated lattices, such as triangular and kagome, the spin liquid phase will be stable against these fluctuations. We therefore expect lattice frustration to play a crucial role in the experimental realization of these phases.

To conclude, Mott insulating ultracold bosonic systems may provide a rich environment in which to observe and study the long-sought-after quantum spin liquid phase. Given the richness of the large-*f* spin models, there may even be a whole class of atomic-spin-liquid-like phases, each with different spin, lattice geometry, dimensionality, and interaction range. Fortunately, as the experimental control and manipulation of large-spin atoms improves, we only edge closer to the exciting time when we may capture the elusive spin liquid phase in the novel environment of cold atoms.

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