Spin-quadrupole ordering of spin-3/2 ultracold fermionic atoms in optical lattices in the one-band Hubbard model

Hong-Hao Tu,1 Guang-Ming Zhang,1 and Lu Yu2
1Department of Physics, Tsinghua University, Beijing 100084, China
2Institute of Theoretical Physics, Chinese Academy of Sciences, Beijing 100080, China

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Based on a generalized one-band Hubbard model, we study magnetic properties of Mott insulating states for ultracold spin-$\frac{3}{2}$ fermionic atoms in optical lattices. When the $s$-wave scattering lengths for the total spin $S = 2, 0$ satisfy conditions $a_2 > a_0 > 0$, we apply a functional integral approach to the half filled case, where the spin-quadrupole fluctuations dominate. On a 2D square lattice, the saddle point solution yields a staggered spin-quadrupole ordering at zero temperature with symmetry breaking from SO(5) to SO(4). Both spin and spin-quadrupole static structure factors are calculated, displaying highly anisotropic spin antiferromagnetic fluctuations and antiferroquadrupole long-range correlations, respectively. When Gaussian fluctuations around the saddle point are taken into account, spin-quadrupole density waves with a linear dispersion are derived. Compared with the spin density waves in the half filled spin-$\frac{1}{2}$ Hubbard model, the quadrupole density wave velocity is saturated in the strong-coupling limit, and there are no transverse spin-quadrupole mode couplings, as required by the SO(4) invariance of the effective action. Finally, in the strong-coupling limit of the model Hamiltonian, we derive the effective hyperfine spin-exchange interactions for the Mott insulating phases in the quarter filled and half filled cases, respectively.

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I. INTRODUCTION

Ultracold atoms in optical lattices provide us with an ideal playground to study various interesting quantum phenomena and to explore novel many-body states with no counterparts in traditional solid state systems [1]. For alkali atoms, their hyperfine spin $F$ is given by a combination of the nuclear spin $I$ and the electron spin $S = \frac{1}{2}$, which has $2F + 1$ manifold magnetic states. In magnetic traps, these $2F + 1$ components are split, while in optical traps these hyperfine spin degrees of freedom are degenerate and strong quantum fluctuations are displayed. Depending on the value of the hyperfine spin $F$, one classifies ultracold atoms as bosons with integers and fermions with half integers.

It has been well-established that ultracold bosonic atoms in optical lattices can exhibit a variety of novel phenomena, such as spinor Bose condensation [2, 3, 4, 5], coherent spin dynamics [6, 7] and superfluid-Mott insulator quantum phase transition [8, 9].

Meanwhile a degenerate $F = \frac{3}{2}$ Fermi gas can be obtained by cooling alkali atoms $^{133}$Cs, as well as alkaline-earth atoms $^{9}$Be, $^{135}$Ba, and $^{137}$Ba. These high spin fermionic atoms in optical traps may lead to peculiar many-body ground states and exotic collective excitations, rarely appearing in interacting electron systems [10, 11]. For example, the Cooper pair structures in such high spin fermions are enriched: a kind of quintet pairing can be formed as an SO(5) polar condensate [12]. In one dimension, the competing SO(5) superfluid order has been investigated based on the bosonization technique [13, 14], and an exact Bethe-ansatz method [15] has also been used to describe the corresponding low-energy states. Furthermore, a spin-3/2 ladder model has been proposed with spontaneous plaquette ground state [16].

Out of these intensive studies, Mott insulating states of ultracold atoms in optical lattices are of particular interest. With a fixed number of atoms on each lattice site, the quantum fluctuations of the hyperfine spin degrees of freedom may lead to magnetic multipolar long-range order. Most previous studies focused on ultracold bosons with hyperfine spin $F = 1, 2$. Adiabatically increasing optical lattice trap depth, these systems go through a quantum superfluid-Mott insulator phase transition. The atomic virtual tunneling processes can result in effective hyperfine spin-exchange interactions between the nearest neighbor sites. In particular, an insulating spin-quadrupole or spin nematic ordering has been proposed for spin-1 bosons in optical lattices [5, 17, 18]. This is a quantum analogue of liquid crystal states, where the spin SU(2) rotational symmetry is broken while the time reversal symmetry is preserved [19, 20]. However, direct experimental observation of such a spin nematic ordering in correlated electron systems by conventional probes is rather difficult [21]. In this paper, we will demonstrate that such a novel quantum magnetic state also exists in the ultracold spin-$\frac{3}{2}$ fermionic atoms in optical lattices, which may provide new opportunities for experimental investigations.

In Sec. II, we will introduce the general spin-$\frac{3}{2}$ Hubbard model based on the microscopic $s$-wave atom-atom interactions. In Sec. III a functional integral approach is applied to the half filled generalized Hubbard model when the $s$-wave scattering lengths for the total spin $S = 2, 0$ satisfy conditions $a_2 > a_0 > 0$. On a square lattice, the saddle-point solution gives rise to a staggered spin-quadrupole (nematic) ordered state with sym-
metry breaking from SO(5) to SO(4), and the corresponding spin and spin-quadrupole correlation functions will be evaluated. Moreover, by taking into account the Gaussian fluctuations around the saddle point, the spin-quadrupole (nematic) density waves are derived and the corresponding density wave velocity and stiffness are calculated. In Sec. IV, using the second-order perturbation theory in the strong-coupling limit of the model Hamiltonian, we will derive the effective hyperfine spin-exchange interactions for the Mott insulating phases in the quarter filled and half filled cases, respectively. Finally, a summary is presented in Sec. V.

II. FORMULATION OF SPIN-$\frac{3}{2}$ ONE-BAND HUBBARD MODEL

In order to present our results in a self-contained way and to introduce notations, we first rewrite the fundamental interactions between two hyperfine spin $F = \frac{3}{2}$ fermion atoms with a contact potential [10, 11]. For the low energy states of fermions, it is convenient to consider only the s-wave scattering,

$$V(r_1 - r_2) = \frac{4\pi\hbar^2}{m} \delta(r_1 - r_2) (a_0 P_0 + a_2 P_2),$$

where $P_S$ projects the pair of atoms into states with total spin $S = 0, 2$ and $a_S$ is the s-wave scattering length in the spin-$S$ channel. Due to antisymmetry of the wave function, the s-wave scattering of identical fermions in channels $S = 1, 3$ is not allowed. Using the relations

$$P_0 + P_2 = 1, \quad S_1 \cdot S_2 = \lambda_0 P_0 + \lambda_2 P_2,$$

with $\lambda_0 = -15/4$ and $\lambda_2 = -3/4$, the interaction can be written in terms of spin operators

$$V(r_1 - r_2) = \delta(r_1 - r_2) (g_0 + g_2 S_1 \cdot S_2),$$

where $g_0 = \pi\hbar^2(5a_2 - a_0)/m$ and $g_2 = 4\pi\hbar^2(a_2 - a_0)/(3m)$.

By introducing fermionic creation operators $\psi_{i\alpha}^\dagger$ for states in the lowest Bloch band on site $i$ with spin components $\alpha = 3/2, 1/2, -1/2, -3/2$, a generalized one-band Hubbard model can be written as:

$$H = -t \sum_{<ij>,\alpha} \langle \psi_{i\alpha}^\dagger \psi_{j\alpha} + \text{H.c.} \rangle + \frac{c_0}{2} \sum_i N_i (N_i - 1) + \frac{c_2}{2} \sum_i \left( S_i^2 - \frac{15}{4} N_i \right) - \mu \sum_i N_i,$$

where the coupling parameters $c_0$ and $c_2$ are proportional to $g_0$ and $g_2$, respectively, while their explicit expressions depend on the potential of optical traps and the atomic recoil energy. The total number of atoms on site $i$ is

$$N_i = \sum_\alpha \psi_{i\alpha}^\dagger \psi_{i\alpha},$$

and the total spin on site $i$ is defined by

$$S_i = \sum_{\alpha\beta} \psi_{i\alpha}^\dagger S_{\alpha\beta} \psi_{i\beta},$$

with the spin-$3/2$ matrices

$$S^x = \begin{pmatrix} 0 & \sqrt{3} & 0 & 0 \\ \sqrt{3} & 0 & 1 & 0 \\ 0 & 1 & 0 & \sqrt{3} \\ 0 & 0 & \sqrt{3} & 0 \end{pmatrix},$$

$$S^y = \begin{pmatrix} 0 & -i\sqrt{3} & 0 & 0 \\ i\sqrt{3} & 0 & -i & 0 \\ 0 & i & 0 & -i\sqrt{3} \\ 0 & 0 & i\sqrt{3} & 0 \end{pmatrix},$$

$$S^z = \begin{pmatrix} \frac{2}{3} & 0 & 0 & 0 \\ 0 & \frac{1}{3} & 0 & 0 \\ 0 & 0 & -\frac{1}{3} & 0 \\ 0 & 0 & 0 & \frac{2}{3} \end{pmatrix}. \quad (4)$$

These spin operators form an SU(2) Lie algebra $[S^a, S^b] = i\epsilon_{abc} S^c$. The first term in Eq. (3) describes the nearest-neighbor hopping of fermionic atoms, the second term denotes the on-site Hubbard repulsion between atoms, while the third term represents the spin-dependent energy of the individual sites. When the s-wave scattering lengths for $S = 0$ and $S = 2$ are equal, the third term vanishes and the generalized one-band Hubbard model displays an SU(4) symmetry [11, 22]. In optical lattices, the atomic tunneling amplitude $t$ can be easily tuned experimentally by varying the depth of optical traps. Using the Feshbach resonance, the scattering lengths $a_0$ and $a_2$ can be varied, and the coupling parameters $c_0$ and $c_2$ can change over a wide parameter range.

Moreover, for the spin-$\frac{3}{2}$ fermion systems, the spin-quadrupole operators can be introduced as

$$Q^{xy} = \frac{1}{\sqrt{3}} (S^{x} S^{y} + S^{y} S^{x}) = \Gamma^1,$$

$$Q^{xx} = \frac{1}{\sqrt{3}} (S^{x} S^{x} + S^{y} S^{y}) = \Gamma^2,$$

$$Q^{yy} = \frac{1}{\sqrt{3}} (S^{x} S^{y} + S^{y} S^{x}) = \Gamma^3,$$

$$Q^{(0)} = (S^{x})^2 - \frac{5}{4} = \Gamma^4,$$

$$Q^{(2)} = \frac{1}{\sqrt{3}} [(S^{x})^2 - (S^{y})^2] = \Gamma^5,$$

where

$$\Gamma^1 = \begin{pmatrix} 0 & -iI \\ iI & 0 \end{pmatrix}, \quad \Gamma^{2,3,4} = \begin{pmatrix} \vec{\sigma} & 0 \\ 0 & -\vec{\sigma} \end{pmatrix}, \quad \Gamma^5 = \begin{pmatrix} 0 & I \\ I & 0 \end{pmatrix},$$

correspond to the five Dirac matrices, $I$ is a $2 \times 2$ unit matrix, and $\vec{\sigma}(\alpha = x, y, z)$ are Pauli matrices.
The corresponding dipole and octupole operators can be expressed in terms of generators of the SO(5) Lie group as $\Gamma^{ab} = -\frac{i}{2}[\Gamma^a, \Gamma^b]$. Thus, three spin operators, five spin-quadrupole operators, together with seven spin-octupole operators, form the fifteen generators of the SU(4) Lie group. In terms of the Dirac matrices, the spin-quadrupole density operator is represented by a five-component vector

$$n_i^\alpha = \frac{1}{2} \sum_{\alpha\beta} \bar{\psi}_{i\alpha}^\dagger \Gamma^a_{\alpha\beta} \psi_{i\beta} \quad (a = 1, 2, 3, 4, 5).$$

(6)

Therefore, the generalized one-band Hubbard model for interacting spin-$\frac{3}{2}$ fermions in optical lattices can be rewritten in an SO(5) invariant form [11]

$$H = -t \sum_{\langle ij \rangle, \alpha} (\psi_{i\alpha}^\dagger \psi_{j\alpha} + \text{h.c.}) - \frac{3c_2}{4} \sum_i (\bar{n}_i)^2 + \frac{8c_0 - 15c_2}{16} \sum_i (N_i - 2)^2 - \mu \sum_i N_i.$$  

(7)

At half-filling, the chemical potential $\mu$ should be set to zero to ensure the particle-hole (p-h) symmetry: $\psi_{i\alpha} \rightarrow (-1)^i \psi_{i\alpha}^\dagger$, and the average number of fermions per site should be $\langle N_i \rangle = 2$. Thus, as far as the Mott insulating state is concerned, we can safely neglect the particle number fluctuations focusing on the quantum spin fluctuations. In particular, for $c_2 > 0$, the quantum spin-quadrupole fluctuations are an interesting feature of the spin-$\frac{3}{2}$ half filled Hubbard model in the Mott insulating phase.

III. FUNCTIONAL INTEGRAL APPROACH TO HALF FILLED HUBBARD MODEL WITH $c_2 > 0$

A. Saddle point solution

It is known that the functional integral approach provides a powerful tool for studying the antiferromagnetic ground state and spin density wave excitations of half filled spin-$\frac{3}{2}$ Hubbard model [23, 24, 25, 26]. Following the same route, we will apply the functional integral approach to the generalized spin-$\frac{3}{2}$ Hubbard model at half filling.

The partition function is written in an imaginary time functional path-integral form

$$Z = \int D\psi^\dagger D\psi \exp \left[ - \int_0^\beta L (\tau) d\tau \right],$$

(8)

where $\beta = 1/T$ and the Lagrangian $L$ is given by $L = \sum_{i,\alpha} \bar{\psi}_{i\alpha}^\dagger \partial_\tau \psi_{i\alpha} + H$. In the following we denote $U \equiv 3c_2/4$ for simplicity and then an SO(5) invariant Hubbard-Stratonovich transformation can be performed,

$$Z = \int \mathcal{D}\psi^\dagger \mathcal{D}\psi \mathcal{D}\tilde{\phi} \exp \left[ - \int_0^\beta d\tau L' (\tau) \right],$$

with

$$L' = \sum_{i\alpha} \bar{\psi}_{i\alpha}^\dagger \partial_\tau \psi_{i\alpha} - t \sum_{ij, \alpha} (\psi_{i\alpha}^\dagger \psi_{j\alpha} + \text{h.c.})$$

$$+ \sum_i \frac{1}{2} \bar{\phi}_i^\alpha + \sqrt{\frac{U}{2}} \sum_{ij, \alpha\beta} \bar{\phi}_i^\alpha \Gamma_{\alpha\beta} \phi_{j\beta},$$

(9)

where a five-component real bosonic field $\phi_i^\alpha (\tau)$ has been introduced. By integrating out the fermion fields $\psi^\dagger$ and $\psi$, we obtain an effective action

$$S_{\text{eff}} = \int_0^\beta d\tau \sum_i \frac{1}{2} \bar{\phi}_i^\alpha (\tau) - \text{Tr} \ln \left[ \partial_\tau + M \right].$$

(10)

Here the trace is taken over the Nambu space, the spatial and imaginary time coordinates. The matrix element of $M$ and fermionic Green’s function (GF) are given by

$$G_{\alpha\beta}(r_i, \tau; r_j, \tau') = -\langle r_i, \tau, \alpha | \frac{1}{\partial_\tau + M} | r_j, \tau', \beta \rangle,$$

$$\langle r_i, \tau, \alpha | M | r_j, \tau', \beta \rangle = -2t \delta_{\alpha\beta} \delta_{\tau, \tau'} \delta_{i, j + \delta} + \sqrt{\frac{U}{2}} \delta_{\tau, \tau'} \delta_{i, j} \bar{\phi}_i^\alpha (\tau) \cdot \Gamma_{\alpha\beta}.$$  

(11)

So far no approximations have been made.

In order to reveal the consequences of the spin-quadrupole fluctuations, we first consider the saddle-point solution of the effective action. Differentiating $S_{\text{eff}}$ with respect to $\phi_i^\alpha (\tau)$, we obtain

$$\phi_i^\alpha (\tau) = -\sqrt{\frac{U}{2}} \sum_{\alpha\beta} G_{\alpha\beta}(r_i, \tau; r_i, \tau) \Gamma_{\beta\alpha}^\ast,$$

(12)

corresponding to a mean field result [11]. It is expected that the lowest energy state is given by a staggered phase of the SO(5) vector, namely, a staggered spin-quadrupole (spin nematic) ordered phase with an order parameter $\phi_i (\tau) \rightarrow \phi_i e^{iQ \cdot r_i}$, where $d$ vector corresponds to one of the five spin-quadrupole components resulting from spontaneous symmetry breaking of SO(5) to SO(4). On a two-dimensional (2D) square lattice $Q = (\pi, \pi)$ corresponds to the reciprocal wave vector. Compared with the conventional magnetic long-range ordered states, there is no time reversal symmetry breaking in the spin-quadrupole ordering state. Then the effective action at the saddle point becomes
with respect to terms in momentum space due to the umklapp processes 

\[ \langle \pm \rangle \]

By Fourier transformation, the gap equation is given by

\[ \Delta = 2t \sum_{\mathbf{k}, \omega_n, \alpha} \left( \psi^\dagger_{\mathbf{k}, \omega_n} \phi^\dagger_{\alpha\mathbf{k}} \mathbf{\hat{d}} \right) \left( \psi_{\mathbf{k}, i\omega_n} \phi_{\overline{\alpha\mathbf{k}}} \right) + \frac{1}{2} \beta N \phi^2, \]

(13)

where the summation over momentum is limited to the reduced Brillouin zone, the dispersion relation is \( \varepsilon_{\mathbf{k}} = -2t(\cos k_x + \cos k_y) \), and \( \omega_n \) is the fermionic Matsubara frequency.

Due to the doubled unit cell, the fermion GF generally has to be defined as \( G_{\alpha\beta}(\mathbf{k}, \mathbf{k}'; \tau) = -\langle T\psi_{\alpha\mathbf{k}}(\tau)\psi_{\beta\mathbf{k}'}(0) \rangle \), which has non-zero off-diagonal terms in momentum space due to the umklapp processes with respect to \( \mathbf{Q} \). Explicitly, the expression of the single-particle GF can be written as

\[ G_{\alpha\beta}(\mathbf{k}, -\mathbf{k}'; i\omega_n) = \frac{1}{(i\omega_n + \varepsilon_{\mathbf{k}}) - \varepsilon_{\mathbf{k}} \delta_{\mathbf{k} \mathbf{k'}} + \sqrt{\frac{\sqrt{\frac{\varepsilon^2_{\mathbf{k}} + \frac{\Delta^2}{\beta N}}{\varepsilon_{\mathbf{k}}^2}}}} \]

(14)

where GF poles lead to the quasiparticle spectra \( E_{\mathbf{k}} = \pm \sqrt{\frac{\varepsilon^2_{\mathbf{k}} + \frac{\Delta^2}{\beta N}}{\varepsilon_{\mathbf{k}}^2}} \). At half filling, the upper band is empty while the lower band is completely filled. Thus an energy gap \( \Delta = 2\sqrt{\frac{\varepsilon^2_{\mathbf{k}} + \frac{\Delta^2}{\beta N}}{\varepsilon_{\mathbf{k}}^2}} \) opens up in the quasiparticle spectrum.

By Fourier transformation, the gap equation is given by

\[ 1 - \frac{2U}{\beta N} \sum_{\mathbf{k}, i\omega_n} \frac{1}{\omega_n^2 + E_{\mathbf{k}}^2} = 0. \]

(15)

At \( T = 0K \), for arbitrarily small \( U \) there is always a finite energy gap. Particularly, in the limit of \( U \ll t \), it gives rise to

\[ \Delta \approx 2te^{-\pi\sqrt{2t/U}}. \]

(16)

However, for \( U \gg t \), we have \( \Delta \approx 2U \), i.e., the Mott gap in the single-particle excitations.

Moreover, the sublattice spin-quadrupole moment is related to the energy gap and is given by

\[ |\mathbf{n}| = \frac{\Delta}{2U}. \]

(17)

We have numerically calculated the energy gap and plotted the spin-quadrupole moment in Fig.1. For a large value of \( U \), \( \Delta \approx 2U \) and \( |\mathbf{n}| \rightarrow 1 \), i.e., a saturated spin-quadrupole moment.

**B. Spin-quadrupole correlations at the saddle point**

The longitudinal and transverse spin-quadrupole correlation functions are defined as

\[ \chi_\sigma(\mathbf{p}, \mathbf{p}'; i\omega_n) = \frac{1}{N} \text{Tr} \left[ (\mathbf{T} \cdot \mathbf{\hat{d}}) \mathbf{G}(\mathbf{k}, -\mathbf{k}'; i\omega_n) \right] \]

\[ \chi_\tau(\mathbf{p}, \mathbf{p}'; i\omega_n) = \frac{1}{N} \text{Tr} \left[ (\mathbf{T} \cdot \mathbf{\hat{d}}) \mathbf{G}(\mathbf{k}, -\mathbf{k}'; i\omega_n) \right] \]

\[ \chi_\pi(\mathbf{p}, \mathbf{p}'; i\omega_n) = \frac{1}{N} \text{Tr} \left[ (\mathbf{T} \cdot \mathbf{\hat{d}}) \mathbf{G}(\mathbf{k}, -\mathbf{k}'; i\omega_n) \right] \]

\[ \chi_\pi'(\mathbf{p}, \mathbf{p}'; i\omega_n) = \frac{1}{N} \text{Tr} \left[ (\mathbf{T} \cdot \mathbf{\hat{d}}) \mathbf{G}(\mathbf{k}, -\mathbf{k}'; i\omega_n) \right] \]

(18)

After some straightforward algebra, the spin-quadrupole correlation functions are expressed as

\[ \chi_\sigma(\mathbf{p}, \mathbf{p}'; i\omega_n) = \frac{\delta_{\mathbf{p}\mathbf{p}'} \omega_n}{\beta N} \sum_{\mathbf{k}, i\omega_n} \left( \omega_n + i\omega_n + \varepsilon_{\mathbf{k}+\mathbf{p}'} - \frac{\Delta^2}{4} \right) \]

\[ \chi_\pi(\mathbf{p}, \mathbf{p}'; i\omega_n) = \frac{\delta_{\mathbf{p}\mathbf{p}'} \omega_n}{\beta N} \sum_{\mathbf{k}, i\omega_n} \left( \omega_n + i\omega_n + \varepsilon_{\mathbf{k}+\mathbf{p}'} - \frac{\Delta^2}{4} \right) \]

(19)

Performing the Matsubara frequency summation and analytical continuation, the imaginary part of \( \chi(\mathbf{p}, \mathbf{p}'; \omega) \) can be derived. Since the off-diagonal terms in momentum space vanish, we can only consider \( \chi(\mathbf{p}, \mathbf{p}; \omega) \).
To display the spatial correlations, the static spin-quadrupole structure factors are evaluated through the fluctuation-dissipation relation $S(p) = \frac{1}{\beta N} \int_{-\infty}^{+\infty} d\omega [1 + n_B(\omega)] \text{Im} \chi(p, p; \omega)$, where $n_B(\omega)$ is the Bose-Einstein distribution function. At $T = 0K$, the static structure factors are obtained

$$\left( \frac{S_x(p)}{S_y(p)} \right) = \frac{1}{4N} \sum_k (1 - \frac{\varepsilon_{k,p} \pm \delta^2}{E_k E_{k+p}}).$$

(20)

The static spin-quadrupole structure factors are numerically calculated and displayed in Fig. 2, where broad peaks appear at $p = (\pi, \pi)$, indicating antiferroquadrapolar long-range correlations.

C. Anisotropic spin fluctuations at the saddle point

It is interesting to examine the spin-spin correlation functions in the presence of staggered spin-quadrupolar ordering background. Let us first look at the dipole moment,

$$\langle S_\gamma^\alpha \rangle = -\frac{1}{\beta N} \sum_{k,i,l} \text{Tr} [S^{\gamma \alpha} G(k, Q - k; i, \omega_n)] = 0,$$

(21)

where $S_\gamma (\gamma = x, y, z)$ denotes the corresponding spin-$\frac{1}{2}$ matrices. This result implies that there is no spin-dipole long-range order in the saddle point solution. To get the collective excitations, the spin-correlation functions have to be evaluated,

$$\chi^{\alpha \beta}(p, p'; \tau) = \frac{1}{N} \langle \text{Tr}_p S^\alpha_p(\tau) S^\beta_{p'}(0) \rangle.$$

(22)

By expressing the spin-density operators in terms of Nambu spinor as $S^\alpha_p = \sum_{k,\alpha,\beta} \psi^\dagger_{k+p,\alpha} S^{\gamma}_{\alpha \beta} \psi_{k,\beta}$, the spin-correlation functions are given by

$$\chi^{\alpha \beta}(p, p'; i, \omega_l) = -\frac{1}{\beta N} \sum_{kk',i,l} \text{Tr} [S^{\alpha} G(k, -k'; i, \omega_n) \times S^{\beta} G(k' + p', -k - p; i, \omega_n + i\omega(l))].$$

(23)

Without loss of generality, we may assume the spin-quadrupole ordering is along $d = \hat{d}_4$. Then the order parameter is denoted by $(-1)^{l} n^d_4 \neq 0$, where

$$2n^d_4 = \psi^\dagger_{i,\tau} \psi_{i,\tau} + \psi^\dagger_{i,-\tau} \psi_{i,-\tau} - \psi^\dagger_{i,\tau} \psi_{i,-\tau} - \psi^\dagger_{i,-\tau} \psi_{i,\tau}.$$

(24)

(corresponding to the difference between the $S^x = \pm 3/2$ and $S^y = \pm 1/2$ spin densities in the site singlet state). The physical picture corresponding to this particular spin-quadrupole ordering state is displayed in Fig. 3. Inserting the fermionic GF into the above expression of the spin correlations, we find that

$$\chi^{zz}(p, p'; i, \omega_l) = \chi^{yy}(p, p'; i, \omega_l) \neq \chi^{zz}(p, p'; i, \omega_l),$$

(25)

which implies that in the spin-quadrupole long-range ordered state the time reversal symmetry is preserved but the spin rotational symmetry is broken. This is the main characteristics of the spin-quadrupole or spin nematic ordered state. Thus the corresponding expressions of the spin-correlation functions are written as

$$\chi^{zz}(p, p'; i, \omega_l) = \frac{5\delta_{pp'}}{\beta N} \sum_{k,i,l} \frac{\omega_n (\omega_n + \omega_l) - \varepsilon_{k,p} + \Delta^2}{(\omega_n + \omega_l)^2 + E^2_{k+p}}.$$

(26)

$$\chi^{zz}(p, p'; i, \omega_l) = \frac{2\delta_{pp'}}{\beta N} \sum_{k,i,l} \frac{5\omega_n (\omega_n + \omega_l) - 5\varepsilon_{k,p} + \Delta^2}{(\omega_n + \omega_l)^2 + E^2_{k+p}}.$$

(27)

It should be emphasized that unlike the half-filled spin-$\frac{1}{2}$ Hubbard model [23], all off-diagonal spin correlations in the momentum space vanish, which will be discussed in the next subsection. At $T = 0K$, we may rewrite $\chi(p, p'; \omega) = \chi(p, \omega) \delta_{pp'}$, and the static spin structure
factors can be derived as
\[
S^{zz}(p) = \frac{5}{4N} \sum_k \left( 1 - \varepsilon_k \xi_{k+p} + \frac{\Delta^2}{4} \right), \\
S^{+-}(p) = \frac{1}{2N} \sum_k \left( 9 - \frac{5\varepsilon_k \xi_{k+p} - \Delta^2}{2} \right). \tag{28}
\]

We have plotted the corresponding numerical results in Fig. 4. For both \(S^{zz}(p)\) and \(S^{+-}(p)\), much broader peaks at \((0, \pi)\) and \((\pi, 0)\) show collinear spin-dimer correlations. Actually, these peculiar features of spin and spin-quadrupole structure factors can be detected by polarized light Bragg scattering or spatial quantum noise interferometry experiments \(^{27}\), which are reliable experimental probes to detect magnetic correlations of ultracold atoms in optical lattices.

D. Spin-quadrupole density waves

To further study the spin-quadrupole collective excitation modes, we consider Gaussian fluctuations around the saddle point solution, \(\bar{\phi} = \bar{\phi}_c + \delta\bar{\phi}\). Then, according to
\[
\text{Tr} \ln[\partial_{\tau} + M] = \text{Tr} \ln[-G^{-1}(1 - GV)] = \text{Tr} \ln(-G^{-1}) - \frac{1}{n} \text{Tr}(GV)^n, \tag{29}
\]
the effective action \(S_{\text{eff}}\) can be expanded as \(S_{\text{eff}} = \sum_{n=0}^{\infty} S^{(n)}(\bar{\phi}_c, \delta\bar{\phi})\), where \(G\) represents the fermionic GF at the saddle point and the matrix element of \(V\) is given by
\[
(r_i, \tau, \alpha|V|r_j, \tau', \beta) = \delta_{\tau\tau'} \delta_{ij} \sqrt{\frac{U}{2}} \delta_{\beta}(\tau) \cdot \tilde{\Gamma}_{\alpha\beta}. \tag{30}
\]
Since \(V\) only contains a linear term in \(\delta\bar{\phi}\), the above procedure is indeed an expansion in the spin-quadrupole fluctuation field \(\delta\bar{\phi}\). The first-order term in \(\delta\bar{\phi}\) vanishes due to the saddle point condition. After some algebra, up to the second-order expansion, we arrive at
\[
S^{(2)}(\delta\bar{\phi}) = \frac{1}{2} \sum_{pp',i\omega_n} \sum \delta\phi^a(p', -i\omega_n) \delta\phi^b(p, i\omega_n) \times K^{ab}(p, p'; i\omega_n), \tag{31}
\]
where
\[
K^{ab}(p, p'; i\omega_n) = \delta_{ab}\delta_{p', -p} + \frac{U}{2\beta N} \sum_{kk',i\omega_n} \text{Tr}[G(k, -k'; i\omega_n) \times \Gamma^a G(k' - p', -k - p; i\omega_n + i\omega_l) \Gamma^b], \tag{32}
\]
\(1 \leq a, b \leq 5\) and \(\omega_l\) are bosonic Matsubara frequencies. With the help of the following relations for the Dirac gamma matrices
\[
\text{Tr}1 = 4, \quad \text{Tr}\Gamma^a = 0, \quad \text{Tr} (\Gamma^a \Gamma^b) = 4\delta_{ab}, \quad \text{Tr} (\Gamma^a \Gamma^b \Gamma^c) = 0, \quad \text{Tr} (\Gamma^a \Gamma^b \Gamma^c \Gamma^d) = 4 (\delta_{ab}\delta_{cd} - \delta_{ac}\delta_{bd} + \delta_{ad}\delta_{bc}), \tag{33}
\]
the above kernel functions are expressed as
\[
K^{ab}(p, p'; i\omega_l) = [K_0(p, i\omega_l) \delta_{ab} + K_1(p, i\omega_l)] d_{a'b'} d_{p'p}, \tag{34}
\]
with
\[
K_0(p, i\omega_l) = 1 - 2U\chi_{\sigma}(p, p; i\omega_l), \quad K_1(p, i\omega_l) = 2U [\chi_{\sigma}(p, p; i\omega_l) - \chi_{\pi}(p, p; i\omega_l)], \tag{35}
\]
where \(\chi_{\sigma}(p, p; i\omega_l)\) and \(\chi_{\pi}(p, p; i\omega_l)\) correspond to the spin-quadrupole correlation functions Eq. (19) derived at the saddle-point approximation.

Decomposing the spin-quadrupole fluctuation field \(\delta\bar{\phi}\) into one longitudinal component \(\bar{\sigma}\) parallel to the direction \(d\) and four transverse components \(\bar{\pi}\) perpendicular to the direction \(d\), i.e., \(\delta\bar{\phi} = (\bar{\sigma} + \bar{\pi})\). In terms of \(\bar{\sigma}\) and \(\bar{\pi}\), the effective action with Gaussian fluctuations is expressed as
\[
S^{(2)}(\delta\bar{\phi}) = \sum_{p, i\omega_l} \frac{1}{2} [K_0(p, i\omega_l) + K_1(p, i\omega_l)] |\bar{\sigma}(p, i\omega_l)|^2 + \sum_{p, i\omega_l} \frac{1}{2} K_0(p, i\omega_l) |\bar{\pi}(p, i\omega_l)|^2. \tag{36}
\]
In the low energy limit \(i\omega_l \rightarrow 0\), for the momentum transfer \(p = Q\), we find \(K_0(Q, 0) = 0\) and \(K_1(Q, 0) > 0\) following from the quasiparticle gap equation. Hence the corresponding four transverse modes \(\bar{\pi}\) are nothing but the Goldstone boson modes induced by the spontaneous symmetry breaking from SO(5) to SO(4), living on the space \(SO(5)/SO(4) = S^4\), while the \(\bar{\sigma}\) mode is gapful. Thus we identify these Goldstone collective excitation modes with spin-quadrupole density waves.

For the spin-density wave (SDW) in the half filled spin-\(\frac{1}{2}\) Hubbard model, it had been noticed that the coupled

FIG. 4: (Color online) Static spin structure factor \(S^{zz}(p)\) (a) and \(S^{+-}(p)\) (b) in 2D staggered spin nematic ordered phase with \(U/t = 0.686\) and \(\Delta = 0.3\).
vibrations of $S_x(q)$ and $S_y(Q + q)$ produce the spin density wave [28][29]. In the functional integral approach, up to the Gaussian fluctuations, the coupled transverse vibrations are generated and allowed by the symmetry breaking of SO(3) to SO(2) in the effective action [24][26], which is also the manifestation of an identity for Pauli matrices: $\text{Tr}(\sigma^a \sigma^b \sigma^c) = 2i \epsilon^{abc}$. Due to such coupled transverse vibration modes, the SDW velocity is strongly suppressed in the limit of $U \gg t$, and there exist the off-diagonal transverse spin-correlation functions in the momentum space. To the contrary, we have found that the transverse mode couplings in spin-quadrupole density waves are absent, because the SO(4) invariance of the effective action under the Gaussian fluctuations does not allow such couplings. This is also the consequence of an identity for the Dirac gamma matrices: $\text{Tr}(\Gamma^a \Gamma^b \Gamma^c) = 0$. Actually, the same reason leads to vanishing of the off-diagonal transverse spin-correlation functions in the momentum space. To the contrary, we have found that the off-diagonal transverse spin-correlation functions in the limit of $U \gg t$, which is also the manifestation of an identity for Pauli matrices: $\text{Tr}(\rho) = 0$. These properties are based on the symmetry consideration. Therefore, the above results obtained in the Gaussian approximation around the saddle point should be valid even in the presence of higher-order fluctuations.

Below $T \sim \Delta$, the single-particle excitations are gapped, and the only contributions to the entropy are due to spin-quadrupole density waves with a linear dispersion. On a 2D square lattice, the thermal energy per unit volume is easily calculated as $E/T \approx 4.8(k_B T)^3/(\pi \hbar^2 v_s^2)$. When measuring the low temperature special heat, we expect that $c \sim T^2$, from which $v_s$ can be determined. We would like to mention that such a quadratic temperature dependence of the specific heat is consistent with recent experiments on Ni$_2$Ga$_3$S$_4$ [30], a rare example which, probably, has a spin-quadrupole long-range ordered ground state [31][32][33].

\section{Effective Hyperfine Spin-Exchange Interactions of the Mott States}

So far, a functional integral approach has been developed to consider the generalized half filled spin-$\frac{1}{2}$ Hubbard model, and we have obtained a number of interesting results. In this Section, we derive the low-energy effective hyperfine spin model Hamiltonian in the strong-coupling limit. In the Mott insulating state with a small, but finite atomic tunneling parameter, the virtual tunneling of ultracold atoms can induce effective spin-exchange interactions between the nearest neighbor sites, leading to possible magnetic multipolar ordering or disordered states. In order to derive such interactions, we have to carefully consider the strong coupling limit and perform the second-order perturbation calculation.

\subsection{Single lattice site}

Let us first look at the extreme limit of $t = 0$. Each lattice site decouples from its nearest neighbor sites, and
the single site Hamiltonian is obtained
\[ H_0 = \frac{c_0}{2} N(N-1) + \frac{c_2}{2} (S^2 - \frac{15}{4}N). \] (41)

The corresponding energy eigenstates labelled by |n, S, m⟩ with good quantum numbers n, S and its z component m are given by
\[ E_0 = \frac{c_0}{2} n(n-1) + \frac{c_2}{2} (S(S+1) - \frac{15}{4}n), \] (42)

which has 2S+1-fold degeneracy. According to the Pauli principle, we can construct the eigenstates with zero and even number of particles as
\[ |0, 0, 0\rangle = |\Omega\rangle; \]
\[ |2, 0, 0\rangle = \frac{1}{\sqrt{2}} (|\psi^{\dagger}_1 \psi^{\dagger}_2 - \psi^{\dagger}_1 \psi^{\dagger}_2\rangle - |\psi^{\dagger}_1 \psi^{\dagger}_2 - \psi^{\dagger}_1 \psi^{\dagger}_2\rangle) |\Omega\rangle, \]
\[ |2, 2, 0\rangle = \frac{1}{\sqrt{2}} (|\psi^{\dagger}_1 \psi^{\dagger}_2 + \psi^{\dagger}_1 \psi^{\dagger}_2\rangle) |\Omega\rangle, \]
\[ |2, 2, 2\rangle = |\psi^{\dagger}_1 \psi^{\dagger}_2\rangle |\Omega\rangle, \]
\[ |2, 2, 1\rangle = |\psi^{\dagger}_1 \psi^{\dagger}_2\rangle |\Omega\rangle, \]
\[ |4, 0, 0\rangle = |\psi^{\dagger}_1 \psi^{\dagger}_2 \psi^{\dagger}_3 \psi^{\dagger}_4\rangle |\Omega\rangle, \] (43)

while the eigenstates with odd number of particles are given by
\[ |1, 3, 3\rangle = |\psi^{\dagger}_1 \psi^{\dagger}_2 \psi^{\dagger}_3\rangle |\Omega\rangle, \]
\[ |1, 3, 3\rangle = |\psi^{\dagger}_1 \psi^{\dagger}_2 \psi^{\dagger}_3\rangle |\Omega\rangle, \]
\[ |3, 3, 3\rangle = |\psi^{\dagger}_1 \psi^{\dagger}_2 \psi^{\dagger}_3\rangle |\Omega\rangle, \]
\[ |3, 3, 3\rangle = |\psi^{\dagger}_1 \psi^{\dagger}_2 \psi^{\dagger}_3\rangle |\Omega\rangle, \]
\[ |3, 3, 3\rangle = |\psi^{\dagger}_1 \psi^{\dagger}_2 \psi^{\dagger}_3\rangle |\Omega\rangle, \] (44)

Here it is worth noticing that the number of the possible eigenstates are substantially reduced by the Pauli exclusion principle. The corresponding eigen-energies are listed in Table I.

From the experimental point of view, the Mott insulating states with one or two particles per site are the most interesting cases because they are free of three-body decays.

### B. One atom per site

To derive the effective model Hamiltonian, we can simply consider a two-site problem. For one atom per site, the unperturbed ground state will be given by |1, 3/2, m⟩(i) |1, 3/2, m⟩(j). Then, four intermediate states are allowed:
\[ |\Omega\rangle^{(i)} |2, 2, 0\rangle^{(j)} \]
\[ |\Omega\rangle^{(i)} |2, 2, 0\rangle^{(j)} \]
\[ |\Omega\rangle^{(i)} |2, 2, 0\rangle^{(j)} \]
\[ |\Omega\rangle^{(i)} |2, 2, 0\rangle^{(j)} \]
\[ \epsilon_S = -t^2 \sum \langle v | H_t | n, S, m \rangle \langle 2 | E_v - E_g, \] (46)

where |n, S, m⟩ and |v⟩ denote the initial state and the possible intermediate states in the representation of the two-site system, respectively. The total number of particles n, the total spin S, and its z component S_z are good quantum numbers, while E_v and E_g correspond to the zeroth-order eigenenergies of these states, which are calculated from the single-site eigenenergies in the Table I. According to the allowed intermediate states, the channels with total spin S = 0 and S = 2 have to be considered in this case.

For the energy shift in the total spin S = 0 channel, there are two possible intermediate states: S_i = 0, n_i = 0; S_j = 0, n_j = 2 and i ≠ j. With the help of the Clebsch-Gordon coefficients, the unperturbed state and intermediate states are expressed in the form
\[ |2, 0, 0\rangle = \frac{1}{2} \left( |1, 3, 3\rangle^{(i)} |1, 3, 3\rangle^{(j)} - |1, 3, 3\rangle^{(i)} |1, 3, 3\rangle^{(j)} - |1, 3, 1\rangle^{(i)} |1, 3, 1\rangle^{(j)} + |1, 3, 1\rangle^{(i)} |1, 3, 1\rangle^{(j)} \right), \]
\[ |2, 0, 0\rangle_{\text{int}} = |\Omega\rangle^{(i)} |2, 0, 0\rangle^{(j)}, \]

Thus the corresponding energy shift is evaluated as
\[ \epsilon_0 = -\frac{16t^2}{4c_0 - 15c_2}. \] (48)
Then let us consider the total spin $S = 2$ channel. There are two possible intermediate states $S_i = 0$, $n_i = 0$, $S_j = 2, n_j = 2$ and $i \leftrightarrow j$. We notice that evaluation of the energy shift with maximal projection $m = S$ is sufficient for obtaining the needed results, because the tunneling Hamiltonian is SU(2) spin invariant and the tunneling processes do not mix the different $m_F$ states. In the representation of the two-site system with good quantum numbers, the unperturbed state and intermediate states with maximal spin polarization are written as

$$
|2, 2, 2\rangle = \frac{1}{\sqrt{2}} \left| \begin{array}{c} \frac{3}{2} \frac{3}{2} \frac{1}{2} \\
\frac{1}{2} \frac{1}{2} \frac{1}{2} \end{array} \right\rangle
$$

$$
- \frac{1}{\sqrt{2}} \left| \begin{array}{c} \frac{3}{2} \frac{3}{2} \frac{1}{2} \\
\frac{1}{2} \frac{1}{2} \frac{1}{2} \end{array} \right\rangle,
$$

$$
|2, 2, 2\rangle_{\text{int}} = |\Omega\rangle^{(i)} \left| \begin{array}{c} 2, 2, 2 \rangle^{(j)} \end{array} \right\rangle, \quad i \leftrightarrow j. \quad (49)
$$

Similar calculation leads to the energy shift in the total spin $S = 2$ channel as

$$
\epsilon_2 = -\frac{16t^2}{4c_0 - 3c_2}. \quad (50)
$$

Thus, up to the second order of the hopping term, the effective spin-exchange interactions for the two-site problem are obtained as

$$
H_{ij} = \epsilon_0 P_{ij}(0) + \epsilon_2 P_{ij}(2), \quad (51)
$$

where $P_{ij}(S)$ projects the two-spin states of $S_i = S_j = 3/2$ onto the total spin-$S$ state. The explicit form of $P_{ij}(S)$ are given as follows:

$$
P_{ij}(0) = \frac{(S_i S_j - \lambda_1) (S_i S_j - \lambda_2) (S_i S_j - \lambda_3)}{(\lambda_0 - \lambda_1)(\lambda_0 - \lambda_2)(\lambda_0 - \lambda_3)},
$$

$$
P_{ij}(2) = \frac{(S_i S_j - \lambda_0) (S_i S_j - \lambda_1) (S_i S_j - \lambda_3)}{(\lambda_0 - \lambda_2)(\lambda_0 - \lambda_1)(\lambda_0 - \lambda_3)}. \quad (52)
$$

with $\lambda_S = \frac{1}{2} [S(S+1) - \frac{15}{4}]$. For the lattice model, the effective spin-exchange interactions is finally obtained up to a constant

$$
H_{eff} = \epsilon_0 \sum_{<ij>} P_{ij}(0) + \epsilon_2 \sum_{<ij>} P_{ij}(2). \quad (53)
$$

Actually, a similar effective hyperfine spin model Hamiltonian was also given in the earlier work [16]. Here we would emphasize that the validity of the second-order perturbation theory is restricted to $c_0 \gg t$ to ensure the stability of the Mott insulating state. When $t$ becomes large and comparable to the energy difference between unperturbed ground state and the lowest intermediate states, higher-order perturbations should be included, and the spin exchange beyond the nearest neighbor sites should be considered.

In particular, when $c_2 = 0$, the original spin-$\frac{3}{2}$ Hubbard model displays an SU(4) symmetry [11, 22], and here we have $\epsilon_0 = \epsilon_2 = -\frac{4t^2}{c_0}$. Then the effective spin-exchange interaction becomes

$$
H_{eff} = -\frac{4t^2}{c_0} \sum_{<ij>} [P_{ij}(0) + P_{ij}(2)], \quad (54)
$$

with

$$
P_{ij}(0) + P_{ij}(2)
$$

$$
= -\frac{1}{9} (S_i S_j)^3 - \frac{11}{36} (S_i S_j)^2 + \frac{9}{16} S_i S_j + \frac{99}{64}. \quad (55)
$$

This effective spin model also exhibits a uniform SU(4) symmetry. In one dimension, an exact solution had been obtained by the Bethe ansatz method [33], and the ground state of the SU(4) spin-exchange model is a spin singlet with gapless spin excitations. Moreover, such a spin-$\frac{3}{2}$ exchange Hamiltonian is equivalent to the so-called SU(4) spin-orbital model, which has been extensively studied in solid-state systems [36].

### C. Two atoms per site

More interesting and complicated situations occur for a Mott insulator with two interacting atoms per site. Now the possible ground states on each site may depend on the spin dependent coupling parameter $c_2$. In the limit of $t = 0$, the single site state is a spin singlet ($S_i = 0$) for $c_2 > 0$ and a spin quintet ($S_i = 2$) for $c_2 < 0$. However, a small finite tunneling induces an exchange energy of order $t^2/c_0$ between the nearest neighbor sites. Since the energy difference between the spin singlet and quintet states is given by $c_2$, when $t^2/c_0$ is large compared with the absolute value of $c_2$, the quintet state is mixed with the singlet state, and both spin configurations of these two states have to be taken into account in the second-order perturbation calculations. However, in the present section we will not consider this situation and simply assume $t \ll \sqrt{c_0|c_2|}$. The discussion is divided into two parts.

#### 1. $c_2 > 0$

In this case, the two-site unperturbed ground state corresponds to the site singlet state $\langle 2, 0, 0 \rangle^{(i)} \langle 2, 0, 0 \rangle^{(j)}$, and the allowed intermediate states are

$$
\left| \begin{array}{c} \frac{3}{2}, m_i \end{array} \right\rangle^{(i)} \left| \begin{array}{c} \frac{3}{2}, m_j \end{array} \right\rangle^{(j)} \quad \text{and} \quad i \leftrightarrow j. \quad (56)
$$

According to the initial state, only the total spin $S = 0$ channel is involved. The two-site unperturbed and inter-
mediate states are expressed as

\[
|4, 0, 0\rangle = \frac{1}{2} |\frac{3}{2}, 3\rangle^{(i)} |\frac{3}{2}, 3\rangle^{(j)}, \\
|4, 0, 0\rangle_{\text{int}} = \frac{1}{2} |\frac{3}{2}, \frac{3}{2}\rangle^{(i)} |\frac{3}{2}, \frac{3}{2}\rangle^{(j)} \\
- \frac{1}{2} |\frac{3}{2}, -\frac{3}{2}\rangle^{(i)} |\frac{3}{2}, \frac{3}{2}\rangle^{(j)} \\
- \frac{1}{2} |\frac{3}{2}, \frac{1}{2}\rangle^{(i)} |\frac{3}{2}, -\frac{1}{2}\rangle^{(j)} \\
+ \frac{1}{2} |\frac{3}{2}, -\frac{1}{2}\rangle^{(i)} |\frac{3}{2}, \frac{1}{2}\rangle^{(j)} , \quad (57)
\]

and the corresponding intermediate state with \(i \leftrightarrow j\). After some algebra, the energy shift is calculated as

\[
\epsilon_0 = -\frac{8t^2}{4c_0 + 15c_2} . \quad (58)
\]

Then, up to a constant, the effective lattice model is obtained as

\[
H_{\text{eff}} = -\frac{8t^2}{4c_0 + 15c_2} \sum_{<ij>} P_{ij}(0) , \quad (59)
\]

where \(P_{ij}(0)\) is the singlet projection operator between two site singlet states, which can not be expressed in terms of powers of hyperfine spin exchanges. In this sense, the functional integral approach developed in Sec. III is a more appropriate method to attack this problem.

2. \(c_2 < 0\)

In this case the two-site unperturbed ground state is described by the quintet state \(|\frac{2}{2}, m_j\rangle^{(i)} |\frac{2}{2}, m_j\rangle^{(j)}\). Two intermediate states have been given in [53], but the total spin \(S = 0, 1, 2, 3\) channels have to be considered, as the virtual tunneling process is forbidden in the \(S = 4\) channel due to the Pauli’s exclusion principle. With the help of the Clebsch-Gordan coefficients, the unperturbed two-site ground states with maximal spin polarization are written as

\[
|4, 3, 3\rangle = \frac{1}{\sqrt{2}} |\frac{2}{2}, 2\rangle^{(i)} |\frac{2}{2}, 1\rangle^{(j)} \\
- \frac{1}{\sqrt{2}} |\frac{2}{2}, 1\rangle^{(i)} |\frac{2}{2}, 2\rangle^{(j)} , \\
|4, 2, 2\rangle = \frac{1}{\sqrt{7}} |\frac{2}{2}, 2\rangle^{(i)} |\frac{2}{2}, 0\rangle^{(j)} \\
+ \frac{1}{\sqrt{7}} |\frac{2}{2}, 0\rangle^{(i)} |\frac{2}{2}, 2\rangle^{(j)} \\
- \frac{1}{\sqrt{7}} |\frac{2}{2}, 1\rangle^{(i)} |\frac{2}{2}, 1\rangle^{(j)} , \\
|4, 1, 1\rangle = \frac{1}{\sqrt{5}} |\frac{2}{2}, 2\rangle^{(i)} |\frac{2}{2}, -1\rangle^{(j)} \\
- \frac{1}{\sqrt{5}} |\frac{2}{2}, -1\rangle^{(i)} |\frac{2}{2}, 2\rangle^{(j)} \\
- \frac{3}{10} |\frac{2}{2}, 1\rangle^{(i)} |\frac{2}{2}, 0\rangle^{(j)} \\
+ \frac{3}{10} |\frac{2}{2}, 0\rangle^{(i)} |\frac{2}{2}, 1\rangle^{(j)} , \\
|4, 0, 0\rangle = \frac{1}{\sqrt{5}} |\frac{2}{2}, 2\rangle^{(i)} |\frac{2}{2}, -2\rangle^{(j)} \\
+ \frac{3}{\sqrt{5}} |\frac{2}{2}, 0\rangle^{(i)} |\frac{2}{2}, 0\rangle^{(j)} \\
- \frac{1}{\sqrt{5}} |\frac{2}{2}, 1\rangle^{(i)} |\frac{2}{2}, 1\rangle^{(j)} \\
- \frac{1}{\sqrt{5}} |\frac{2}{2}, 1\rangle^{(i)} |\frac{2}{2}, -1\rangle^{(j)} \\
+ \frac{1}{\sqrt{5}} |\frac{2}{2}, -2\rangle^{(i)} |\frac{2}{2}, 2\rangle^{(j)} . \quad (60)
\]

The intermediate states in total spin \(S = 0\) channel have been expressed as \([57]\), while the other intermediate states in channel \(S = 3, 2, 1\) are expressed as

\[
|4, 3, 3\rangle_{\text{int}} = |\frac{3}{2}, \frac{3}{2}\rangle^{(i)} |\frac{3}{2}, \frac{3}{2}\rangle^{(j)} , \\
|4, 2, 2\rangle_{\text{int}} = \frac{1}{\sqrt{2}} |\frac{3}{2}, \frac{3}{2}\rangle^{(i)} |\frac{3}{2}, \frac{3}{2}\rangle^{(j)} \\
- \frac{1}{\sqrt{2}} |\frac{3}{2}, -\frac{3}{2}\rangle^{(i)} |\frac{3}{2}, \frac{3}{2}\rangle^{(j)} , \\
|4, 1, 1\rangle_{\text{int}} = \frac{3}{10} |\frac{3}{2}, \frac{3}{2}\rangle^{(i)} |\frac{3}{2}, -\frac{1}{2}\rangle^{(j)} \\
- \frac{3}{10} |\frac{3}{2}, -\frac{1}{2}\rangle^{(i)} |\frac{3}{2}, \frac{3}{2}\rangle^{(j)} \\
+ \frac{3}{10} |\frac{3}{2}, -\frac{1}{2}\rangle^{(i)} |\frac{3}{2}, -\frac{1}{2}\rangle^{(j)} , \quad (61)
\]

and corresponding intermediate states with interchange \(i \leftrightarrow j\). It is sufficient to calculate the energy shifts with
these maximal spin-polarized states. The final results are obtained as
\[
\epsilon_0 = -\frac{40t^2}{4c_0 - 9c_2}, \quad \epsilon_1 = \epsilon_3 = \frac{2}{5} \epsilon_0, \quad \epsilon_2 = 0.
\] (62)

Note that up to the second order of \( t \), the total spin \( S = 2 \) channel makes no contribution to the energy shift. Hence, the resulting effective spin interaction for the lattice model reads up to a constant
\[
H_{\text{eff}} = \epsilon_0 \sum_{<ij>} \left\{ P_{ij}(0) + \frac{2}{5} [P_{ij}(1) + P_{ij}(3)] \right\},
\] (63)

where \( P_{ij}(S) \) projects the \( S_l = S_r = 2 \) states onto the total spin-\( S \) states. They are given by

\[
P_{ij}(0) = \frac{(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_0)(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_2)(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_3)(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_4)}{(\lambda_0 - \lambda_1)(\lambda_0 - \lambda_2)(\lambda_0 - \lambda_3)(\lambda_0 - \lambda_4)},
\]
\[
P_{ij}(1) = \frac{(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_0)(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_2)(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_3)(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_4)}{(\lambda_1 - \lambda_0)(\lambda_1 - \lambda_2)(\lambda_1 - \lambda_3)(\lambda_1 - \lambda_4)},
\]
\[
P_{ij}(3) = \frac{(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_0)(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_2)(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_3)(\mathbf{S}_i \cdot \mathbf{S}_j - \lambda_4)}{(\lambda_3 - \lambda_0)(\lambda_3 - \lambda_1)(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_4)},
\] (64)

with \( \lambda_S = \frac{1}{2} [S(S + 1) - 12] \). Moreover, the effective spin-exchange model Hamiltonian can be simply written as
\[
H_{\text{eff}} = -\frac{\epsilon_0}{6} \sum_{<ij>} \left[ \frac{1}{15} (\mathbf{S}_i \cdot \mathbf{S}_j)^3 + \frac{2}{15} (\mathbf{S}_i \cdot \mathbf{S}_j)^2 - E_S \right].
\] (65)

Here we would like to emphasize that the spin operators correspond to \( S = 2 \) operators.

V. CONCLUDING REMARKS

To summarize, a functional integral approach has been applied to study the quantum spin-quadrupole long-range ordered state of the Mott insulating phase in the generalized half filled spin-\( \frac{3}{2} \) Hubbard model for the case \( a_2 > a_0 > 0 \). On a square lattice, the ground state shows a staggered spin-quadrupole long-range order from the saddle-point solution of the effective action. By including the Gaussian fluctuations, the four gapless collective modes have been found, corresponding to the spin-quadrupole density waves. Unlike spin density waves in the half filled spin-\( \frac{3}{2} \) Hubbard model, the spin-quadrupole density wave velocity is saturated in the limit of \( a_2 \gg a_0 \), because the transverse mode couplings of the spin-quadrupole collective excitations are not allowed by the SO(4) invariant effective action. Thus, our results obtained under the Gaussian approximation for the spin-quadrupole ordered state are robust even when the high order fluctuations are included. This is a remarkable property of the enlarged hyperfine spin space dimensionality and the higher symmetry of the local interactions in the generalized spin-\( \frac{3}{2} \) Hubbard model.

Moreover, the effective hyperfine spin-exchange interactions for the quarter filled and half filled cases have been derived from the second-order perturbation theory. Starting from these effective spin-exchange interactions, more efficient treatments may be carried out to characterize these multipolar magnetic states. Further investigations in this regard will be reported elsewhere.

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