Properties of the doped spin-$\frac{3}{2}$ Mott insulator near half filling

Stellan Östlund*
Chalmers Technical University/Gothenburg University, Gothenburg 41296, Sweden

T. H. Hansson† and A. Karlhede‡
Department of Physics, Stockholm University, AlbaNova University Center, SE-106 91 Stockholm, Sweden
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We develop an exact generalized Bogoliubov transformation for the spin 3/2 Hubbard model with large anti-Hunds rule coupling near half filling. Since the transformation is unitary, we can employ standard approximate mean-field theory methods in the full Hilbert space to analyze the doped Mott insulator, in contrast to a conventional approach based on truncated Hilbert spaces complemented with hard core constraints. The ground state at exactly half filling is an insulating (Mott) singlet, and according to our analysis an order parameter Δ, usually associated with extended *s*-wave superconductivity, will appear self-consistently as soon as a finite density n of holes is introduced. This is a consequence of the nonlinear nature of the unitary transformation mapping the Mott singlet state to a Fock vacuum which introduces anomalous terms such as Δn in the effective Hamiltonian. Our analysis uses an approach that generalizes readily to multiband Hubbard models and could provide a mechanism whereby a superconducting order parameter proportional to density develops in Mott insulators with locally entangled ground states. For more complicated systems, such an order parameter could coexist naturally with a variety of other order parameters.

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I. INTRODUCTION

A Mott transition is expected to occur when the overlap between atomic orbitals in an insulator becomes large enough for the hopping energy to overcome the energy associated with charge fluctuations. To properly describe this transition is very difficult, and a variety of techniques have been used, with dynamic mean-field theory being an important recent contribution.[1–7] In this work we develop an approach, more in the spirit of BCS theory, for doped Mott insulators where the parent state has an even number of electrons, and thus integer spin, at each site. Examples of such models are two-band Hubbard models considered in the context of ruthenate alloys,[8] and multiband Hubbard models for doped C_{60}.[2,3,6,9,10] We present evidence for superconducting behavior in these systems even for weak doping. We focus on the simplest system to which our conclusions apply, namely, the spin-3/2 Hubbard model on a square lattice with anti-Hunds rule couplings.[8–13] This model neatly illustrates our core idea, namely, that a nonlinear canonical transformation can be used to attack the Mott transition. We do not believe that this method initially should be tested through direct comparison with experiment but rather by other independent calculations and simulations on the same theoretical model.

We define the particle density by $n_r = \sum_s c^\dagger_{s,r} c_{s,r}$ and the spin density by $S_r = \sum_s c^\dagger_{s,r} S_{s,s'} c_{s',r}$, where $-\frac{3}{2} \leqslant s \leqslant \frac{3}{2}$ and $S_{s,s'}$ are the generators of spin-3/2 rotations. Furthermore, we define the operator $P^\dagger_{2,m}(r)$ which creates an $l=2$, $l_z=m$ state with two fermions[12] $P^\dagger_{2,m}(r) = \sum_{\alpha,\beta} \langle \frac{3}{2}\frac{3}{2}\alpha\beta \,|\, \frac{3}{2}\frac{3}{2};2,m\rangle c^\dagger_{\alpha,r} c^\dagger_{\beta,r}$ and the SU(2) invariant $P_r^2 \equiv \sum_m P^\dagger_{2,m}(r) P_{2,m}(r)$. The Hamiltonian containing the maximal number of SU(2) invariant onsite terms is

$$H = -t \sum_{\langle r,r'\rangle,\delta} (c^\dagger_{s,r} c_{s,r+\delta} + \text{H.c.}) + \sum_r [U n_r (n_r - 2) + J P_r^2], \tag{1}$$

where the chemical potential is absorbed in U and $\langle r, r'\rangle$ are all pairs of nearest neigbors counted once.[20] We will consider the case $J \gg U \gg t$, which makes the singlet state heavily favored near $n=2$. The reason for this restriction will be explained in Sec. II.

The single site spectrum consists of sixteen states: an empty site, four equivalent spin 3/2 singly charged states, a singlet and five spin-2 doubly charged sites, four spin-3/2 charge three states and a charge four singlet. The energy $E_g(n)$ of the atomic ground state for n particles is $E_g(0) = E_g(2) = 0$, $E_g(1) = -U$, $E_g(3) = 5J + 3U$, $E_g(4) = 10J + 8U$ and the quintet state has energy $2J$. The lowest energy states obey $E_g(n+1) + E_g(n-1) - 2E_g(n) \geqslant U \gg 0$ so there is no tendency for superconducting pair formation from any of the local interactions.

With our choice of parameters, standard arguments imply that the ground state for $n=2$ and small t should be a spin singlet. For small doping near zero filling, the ground state will most likely be a normal Fermi liquid, at least for small U and J. If U and/or J are sufficiently large, a spin-symmetry breaking state may appear according to the Stoner criterion. Whether or not this happens depends on the density of states, which at least in the case of two dimensions remains finite even down to $n=0$. Numerical simulations indicate that the tendency towards spin ordering is grossly exaggerated in mean-field theory.[14] In any case, a deeper discussion of this point is not within the scope of the present article, which is to investigate the analog of a Fermi liquid near half filling.

The Mott singlet $|\Phi_s(r)\rangle = \Delta^\dagger{}_r|0\rangle$ at site r is created from the vacuum by the operator $\Delta^\dagger{}_r$ given by

$$\Delta^\dagger{}_r = \frac{1}{2\sqrt{2}}\sum_s e^{i\pi(s+1/2)} c^\dagger_{s,r} c^\dagger_{-s,r}. \tag{2}$$

A natural first attempt to understand the system for small hole doping is to try the same method as for the nearly half-filled spin-1/2 Hubbard model, i.e., to make a particle-hole transformation $\hat{c}^\dagger_{s,r} = \hat{c}_{-s,r}$, where $\hat{c}^\dagger_{s,r}$ and $\hat{c}_{s,r}$ are new local fermionic creation and destruction operators. In contrast to the case of the filled spin-1/2 Hubbard model, however, this canonical transformation fails a number of criteria if the Mott singlet is to act as a vacuum for the new operators. In particular, we see that

$$\hat{c}_{s,r}|\Phi_s(r)\rangle \neq 0, \tag{3}$$

$$\langle \Phi_s(r)|\hat{c}_{s,r}\hat{c}^\dagger_{s,r}|\Phi_s(r)\rangle \neq 1. \tag{4}$$

The first inequality is perhaps not so severe; after all the operator $\hat{c}_{s,r}$, being fundamentally a fermion creation operator creates a state with three fermions on site r, and we could argue that we could ignore this problem by suitably projecting onto states with at most two particles. This is in fact the approach usually taken in attempts to perturbatively construct a new vacuum near $n=2$. The second inequality is much worse; it is a consequence of the singlet being entangled, i.e., it cannot be written as a product state in any one-particle basis. As a result, the putative creation operator does not generate a normalized state from the vacuum. The entanglement property implies that the destruction operator on the created state does not recreate the ground state—not even in the two-particle space since it projects into the $S=2$ two-particle states.

II. CANONICAL TRANSFORMATION TO THE MOTT SINGLET VACUUM

We now show how to systematically construct creation and annihilation operators with correct local properties by a canonical transformation that fulfills the following criteria: (a) it maps $|\Phi_s(r)\rangle$ to $|0\rangle$ and (b) it maps the singly charged states to themselves. Due to our choice of interaction parameters, where $J > U$, we also expect that the state composed of two holes will play the same role in the hole doped Mott insulator near $n=2$, as the doubly charged singlet does for small filling. The canonical transformation that we desire should therefore interchange the Mott singlet and the vacuum, leaving all other states invariant. The $S=2$ doubly charged states will have the same charge as the Mott singlet. However, due to the constraints imposed by a canonical transformation utilizing only the spin-3/2 fermion operators, these doubly charged states must be obtained by two applications of the new creation operator, while the singlet created by another double combination of these creation operators forms a state with relative charge minus 2. These considerations force the canonical transformation to be charge nonconserving.

The canonical transformation that accomplishes this and similar mappings can be systematically obtained through the method in Ref. 15. However, we can get it without much formalism as follows. Our desired operator is "almost" $\Delta^\dagger_r$. The problem is that this operator generates unwanted side effects in the $n=1$ and $n=2$ particle subspaces by mapping these to new states with $n=3$ and $n=4$. We get rid of these unwanted overlaps by using a projection operator, defining $Q^\dagger_r = \Delta^\dagger{}_r(1-n)(1-n/2)$. It is straightforward to check that $Q^\dagger_r|0\rangle = |\Phi_s(r)\rangle$, $Q_r|\Phi_s(r)\rangle = |0\rangle$, and that Q_r and $Q^\dagger_r$ annihilate all other states. A canonical transformation that rotates the states $|0\rangle$ and $|\Phi_s(r)\rangle$ at each site r into each other without affecting the other states is provided by the unitary operator

$$U(\tilde{\phi}_r) \equiv e^{iG(\tilde{\phi}_r)} = \prod_r U_r(\tilde{\phi}_r), \tag{5}$$

where

$$U_r(\tilde{\phi}_r) \equiv e^{iG_r(\tilde{\phi}_r)} = e^{i(\tilde{\phi}_r Q^\dagger_r + \tilde{\phi}^\star_r Q_r)} \tag{6}$$

with $\tilde{\phi}_r \equiv \phi_r e^{i\chi_r}$ (ϕ_r and χ_r are real). On $|0\rangle$ and $|\Phi_s(r)\rangle$ the transformation becomes

$$U_r(\tilde{\phi}_r)|_{0,\Phi} = \cos\phi_r + i\sin\phi_r(e^{i\chi_r}Q^\dagger_r + e^{-i\chi_r}Q_r), \tag{7}$$

whereas it is unity on all other states. Choosing $\phi_r = \pi/2$, for all r, we obtain the canonical transformation that fulfills our criteria, i.e., it interchanges the empty state and the Mott state at each site without affecting the other states.

Applying the unitary transformation Eq. (5), with $\phi_r = \pi/2$, to the vacuum state $|0\rangle$, the phase factors $e^{i\chi_r}$ enter the obtained Mott state $|\Phi_s\rangle$ only as an overall phase $\Sigma_r\chi_r$, and can be neglected. In general, however, it is obvious from Eq. (5) that the unitary transformation gives a state where the phase factors enter in a nontrivial way. In particular, this is the case for the slightly doped Mott insulator, which we will consider below. This will be mapped onto a state near the true vacuum state, which can then be analyzed with standard methods. Note that the phase factors $e^{i\chi_r}$ are crucial to retain local gauge symmetry in the same way as the complex phases introduced into the Bogoliubov transformations are necessary to restore gauge invariance in BCS theory.

III. GENERALIZATIONS

Our analysis in this article assumes $0 < U \ll J$. As a consequence the ground state for the on-site terms in Eq. (1) at $n=2$ is nondegenerate—the Mott state is a simple product of local spin singlets. Remember that even though in this case the ground state at $n=2$ is trivial, this is no more true as soon as the Mott insulator is doped so $n=2-\delta<2$. Here we find, using mean field theory, a highly nontrivial result, namely a superconducting order parameter proportional to δ. In the physically more interesting case when $J<0$ or $0<J<U$, the state at $n=2$ is more complicated and the analysis in this article needs modification—we briefly discuss this below. However, we believe that it is important to first establish what the method predicts in the simpler but still very nontrivial case studied here.

A. The degenerate case: $J<0$

When $J<0$, the problem cannot be addressed with the method presented above, since there is no unique ground state at $n=2$ to map to an equivalent $n=0$ problem. Let us, however, suggest a generalization of our strategy: Using a mapping similar to the one in Eq. (5) we can map an arbitrary state in the $n=2$, $L=2$ ground-state multiplet to the $n=0$ state. The arbitrariness of this choice can then be coded in a non-Abelian (gauge) potential in much the same way the arbitrary local phase χ_r was introduced in Eq. (7) above. Our hope is that the resulting gauge theory will be more tractable than the original degenerate Mott problem.[21]

B. The case $0<J<U$

For this case, the $n=2$ ground state is still unique and the mapping using Eq. (5) is well defined and meaningful when the hopping vanishes. Our mean field treatment of the resulting doped $n=2$ system when hopping is introduced is, however, not to be trusted for the following reason. Under the mapping given by Eq. (5), the density operator n transforms as

$$n \to 2 - n + P^2 - n(n-1)(n-2)/6, \tag{8}$$

where P^2 is the operator defined above Eq. (1). When the two-particle singlet has the same free energy as the singly occupied sites, the chemical potential obeys $\mu \approx U$. Reintroducing μ in Eq. (1) and using Eq. (8) shows that that $J \to J_{\text{eff}}=J-U$ under the transformation. Thus for $J<U$ we find that the effective Hamiltonian after the transformation will have $J_{\text{eff}}<0$, and we have the same situation as in Sec. III A.

These generalizations of the model, although of legitimate interest, take us outside what we are able to handle without introducing additional approximations and uncertainties. For that reason they will not be further considered in this paper.

IV. A VARIATIONAL ANSATZ

We now turn to a systematic variational analysis of the slightly doped Mott insulator using the canonical transformation in Eq. (5). In analogy with ordinary Fermi-liquid theory, as well as the BCS theory of superconductivity, we search for a variational state with particle number given by $n=2-\delta$ that is obtained from the vacuum by a canonical transformation $e^{i\mathcal{G}_u}$ depending on a set of parameters u. We define the functions $E(u)$ and $N(u)$ by

$$E(u) = \langle 0|e^{-i\mathcal{G}_u} H e^{i\mathcal{G}_u}|0\rangle, \tag{9}$$

$$N(u) = \langle 0|e^{-i\mathcal{G}_u} N e^{i\mathcal{G}_u}|0\rangle. \tag{10}$$

The values $\{u\}$ which minimize $E(u)$ define our variational ground state $e^{i\mathcal{G}_u}|0\rangle$ with particle number $N(u)$. We have seen that for $n=2$, the transformation $\mathcal{G}_u$ is simply $G[(\pi/2)e^{i\chi_r}] \equiv G_0(\chi_r)$ given by Eq. (5). We hence expect that near the Mott insulator, the relevant transformation will be given by a further transformation close to the identity. We therefore make the ansatz $e^{i\mathcal{G}_u}=e^{iG_0(\chi_r)}e^{i\mathcal{G}'_u} \equiv U_0 U'$.

Note that since we can continuously rotate the Mott state at $n=2$ to the true vacuum by letting ϕ go from $\pi/2$ to zero, we can generate a Mott singlet on a site either by having $U'_r=1$ and $\phi_r=\pi/2$, or by having $U'_r=U_r(\pi/2)$ and $\phi_r=0$. In general, we can make a coherent superposition of empty and doubly occupied singlet sites, both by letting ϕ vary and by adding an onsite s-wave order parameter. As could be expected, this indeterminacy leads to a numerical instability in the variational equations which we resolve by simply taking $\phi_r=\pi/2$ for all r, and not further exploiting these variational parameters.

In order to construct an ansatz for $e^{i\mathcal{G}'_u}$ we first work out $e^{-iG_0(\chi_r)}He^{iG_0(\chi_r)}$. This operator is obtained by replacing each occurence of the fermion operator $c^\dagger_{r,s}$ by $e^{-iG_0(\chi_r)}c^\dagger_{r,s}e^{iG_0(\chi_r)}$ and similarly for $c_{s,r}$. This expression is complicated, but it can nonetheless be worked out exactly in terms of polynomials of $c_{s,r}$ and $c^\dagger_{s,r}$, since the fermion algebra at a site is closed. The exact expression, written here for reference only, is given by

$$\begin{aligned} c^\dagger_{s,r} \to c^\dagger_{s,r}&\left\{[\Delta^\dagger e^{-2i\chi}(1-n) - e^{2i\chi}\Delta] + \left(\frac{:S^2:}{3} + n + \frac{:n^3:}{6}\right)\right\} \\ &+ (-1)^{(s+1/2)}e^{2i\chi}2^{-1/2}\Bigg[-1 + e^{-2i\chi}\Delta^\dagger \\ &+ \left(\frac{:S^2:}{3} + n + \frac{:n^2:}{4}\right)\Bigg]c_{-s,r}, \end{aligned} \tag{11}$$

where the subscripts are dropped on the right-hand side. The notation: O: indicates a normal ordered operator, i.e., strings of fermion operators where all creation operators are anticommuted to the left and annihilation operators to the right taking only into account the sign of the permutation. In this case, $:n^2:=n^2-n$ and $:S^2:=S^2-15n/4$.

The onsite interaction is zero in the vacuum and two particle singlet subspace. Since these are the only two states affected by the canonical transformation, this interaction remains invariant, while the chemical potential transforms according to

$$n \to 2 - \left(n - \frac{5:n^2:}{4} - \frac{:S^2:}{3} + \frac{:n^3:}{6}\right). \tag{12}$$

Anticipating a mean-field calculation under the assumption of no spontaneously broken global symmetries, we do a Wick decomposition of the onsite term, and calculate the expectation value according to

$$\langle Un(n-2) + JP^2\rangle = -\hat{n}U + \hat{n}^2\left(\frac{5J}{8} + \frac{3U}{4}\right) + 2\hat{\Delta}^2 U, \tag{13}$$

where a hat indicates the expectation value of an operator composed of ordinary fermion operators evaluated in the state $U'|0\rangle$ near the physical vacuum. Similarly, the expectation values for the density and s-wave order parameter $\Delta^\dagger_r$ become exactly

$$\begin{aligned} \langle\Delta^\dagger\rangle = \frac{(\hat{\Delta}^*)^2}{2}e^{-i\chi} - \hat{\Delta}e^{2i\chi}&\left(1 + \frac{|\hat{\Delta}|^2}{2} - \frac{\hat{n}}{2} + \frac{\hat{n}^2}{16}\right) \\ &- \hat{\Delta}^*\left(|\hat{\Delta}|^2 - \frac{\hat{n}}{2} + \frac{\hat{n}^2}{8}\right), \end{aligned} \tag{14}$$

$$\langle n\rangle = \hat{n} + 2\left(1-\frac{\hat{n}}{4}\right)^2\left(1-\frac{\hat{n}}{2}\right) - \frac{\hat{n}|\hat{\Delta}_r|^2}{2}. \quad (15)$$

We now derive a similar expansion of the hopping operator. In this case the expressions become a terrible mess—far too complicated to write down in their entirety. It is, however, possible to construct a systematic expansion in the number of fermion operators, which is appropriate for small doping. To this respect, we take the entire expression, and rewrite it as a sum of normal ordered terms. We then truncate this expression at fourth order in fermion operators and keep expectation values of on-site and nearest neighbor s-wave pairing amplitudes, ordinary hopping and density operators. Defining

$$\Delta^\dagger_{r,r'} = \sum_s \frac{(-1)^{(s+1/2)}}{2\sqrt{2}} c^\dagger_{s,r} c^\dagger_{-s,r'}, \quad (16)$$

$$h_{r,r'} = \sum_s c^\dagger_{s,r} c_{s,r'}, \quad (17)$$

where r,r' are nearest neighbors, we find that the hopping operator, truncated at fourth order in fermion operators, becomes

$$-t\langle h_{r,r'}\rangle = -\frac{5t}{4}\hat{n}(e^{2i\chi_r}\hat{\Delta}_{r,r'} + e^{-2i\chi_{r'}}\hat{\Delta}^*_{r,r'}) + \frac{t}{2}e^{2i(\chi_r-\chi_{r'})} \times(1-\hat{n})\hat{h}_{r',r} - t(e^{-2i\chi_r}\hat{\Delta}^*_r - e^{2i\chi_r}\hat{\Delta}_r)e^{-2i\chi_{r'}}\hat{\Delta}^*_{r,r'} + t(e^{-2i\chi_{r'}}\hat{\Delta}^*_{r'} - e^{2i\chi_{r'}}\hat{\Delta}_{r'})e^{2i\chi_r}\hat{\Delta}_{r,r'} + \cdots. \quad (18)$$

Decomposing the expectation values as

$$\hat{\Delta}_{r,r'} = e^{-i(\eta_{r,r'}+\chi_r+\chi_{r'})}\tilde{\Delta}_{r,r'},$$

$$\hat{\Delta}_r = e^{-i2(\eta_r+\chi_r)}\tilde{\Delta}_r,$$

$$\hat{h}_{r',r} = e^{-i(\xi_{r',r}+\chi_r-\chi_{r'})}\tilde{h}_{r',r}, \quad (19)$$

where $\tilde{\Delta}_{r,r'}$, $\tilde{\Delta}_r$, and $\tilde{h}_{r',r}$ are real and non-negative, and the phases η_r, $\eta_{r,r'}$, and $\xi_{r',r}$ are invariant under local gauge transformations [as can be seen from Eqs. (6) and (19)], we obtain

$$-t\langle h_{r,r'}\rangle = te^{i(\chi_r-\chi_{r'})}\Bigg[-\frac{5}{2}\hat{n}\tilde{\Delta}_{r,r'}\cos\eta_{r,r'} + \frac{1}{2}(1-\hat{n})\tilde{h}_{r',r}e^{-i\xi_{r',r}} - 2i\tilde{\Delta}_r\tilde{\Delta}_{r,r'}e^{i\eta_{r,r'}}\sin 2\eta_r + 2i\tilde{\Delta}_{r'}\tilde{\Delta}_{r,r'}e^{-i\eta_{r,r'}}\sin 2\eta_{r'}\Bigg] + \cdots. \quad (20)$$

[Note that what enters the energy is the real term $\langle h_{r,r'}\rangle + \langle h_{r',r}\rangle = 2\,\mathrm{Re}\langle h_{r,r'}\rangle$.] Written in this way, the invariance under local gauge transformations is manifest provided that the original hopping term is supplemented with the usual electromagnetic phase factor $\exp[i2e\int_r^{r'} d\vec{r}''\cdot\vec{A}(\vec{r}'')] = e^{i2eA_{r,r'}}$.

Since the phases η_r, $\eta_{r,r'}$, and $\xi_{r',r}$ are gauge invariant, they are genuine physical quantities (the overall phase is fixed since $\hat{n}_r$ is real). We first discuss the gauge invariant phases and later return to χ_r. In our search for self-consistent mean-field solutions that is described in Sec. IV below, we have only explored homogeneous time-reversal invariant solutions corresponding to $\eta_r = \eta_{r,r'} = 0$, $\xi_{r,r'} = \pi$ and $2\eta_r = -\eta_{r,r'} = \pi/2$, $\xi_{r,r'} = \pi$. These solutions amount to keeping $\hat{n}\tilde{\Delta}_{r,r'}$ or $\tilde{\Delta}_r\tilde{\Delta}_{r,r'}$ in Eq. (20), respectively. For large U, $\tilde{\Delta}_r$ is very small and the first solution, keeping the terms $\sim\hat{n}\tilde{\Delta}_{r,r'}$ and $\sim(1-\hat{n})\tilde{h}_{r,r'}$, is energetically favored. In the mean-field approach the energy to minimize, subject to total particle number given by Eq. (15), is thus given by the sum of the onsite energy Eq. (13) and the hopping energy

$$-t\langle h_{rr'}\rangle = -\frac{t}{2}\tilde{h}_{r,r'}(1-\hat{n}) - \frac{5t}{2}\tilde{\Delta}_{r,r'}\hat{n} + \cdots - \frac{t}{16}\tilde{\Delta}_r\tilde{h}\hat{n}^2. \quad (21)$$

For future reference we have also included the lowest higher order term which couples linearly to $\tilde{\Delta}_r$.

This resulting effective Hamiltonian looks very similar to an ordinary BCS Hamiltonian, corresponding to Eq. (1), but with one dramatic difference, namely the presence of a term proportional to $\tilde{\Delta}_{r,r'}\hat{n}$, as well as a higher order term which couples linearly to the *s*-wave pairing operator $\tilde{\Delta}_r$. Nonzero doping implies, according to Eq. (15), that $\hat{n}>0$. The hopping energy (21) then leads to nonvanishing superconducting order parameters $\tilde{\Delta}_r$, $\tilde{\Delta}_{r,r'}$ as will be further discussed below.

The set of nontrivial variational parameters are χ_r, which characterize the transformation U_0, together with a set of parameters $\{u\}$ that characterize $U' = e^{i\mathcal{G}'_u}$. In the self-consistent mean-field calculation presented below, we make the simplest natural choice for the u's namely the quantities $\hat{n}$, $\tilde{\Delta}_{r,r'}$, $\tilde{\Delta}_r$, and $\tilde{h}_{r,r'}$, which appear in Eqs. (13) and (20) for the energy. A more complicatedvariational ansatz is of course possible, but that would not in general allow for a self-consistent mean-field solution.

Anticipating that we have found a self-consistent solution, we now address the question of stability of this solution to fluctuations. First notice that χ_r only enters as a (lattice) derivative $\chi_r - \chi_{r'}$, so for the homogeneous solution we can always put $\chi_r = 0$ by a global gauge transformation. Concerning the gauge invariant phases, we only studied the time-reversal invariant solutions corresponding to real effective Hamiltonians. Thus, we have, for instance, not excluded the possibility of a magnetized mean-field state with lower energy nor have we explored the possibility of nonzero terms $\sim\sin 2\eta_r$ in Eq. (19) which may or may not lead to time reversal symmetry breaking. Clearly the presence of such a state close to the nonmagnetic Mott state at half filling would in itself be very interesting and would not violate the main conclusions of this paper.

Finally, to study the stability against local fluctuations in the phases χ_r, we expand Eq. (20) to quadratic order in the difference $\chi_r - \chi_{r'}$. In the continuum limit and to leading order in a gradient expansion we have the following effective Hamiltonian for the phase variable:

$$H_\chi = \left[t\tilde{h}_{r,r'}(1-\hat{n}) + 5t\tilde{\Delta}_{r,r'}\hat{n} + \cdots + \frac{t}{8}\tilde{\Delta}_r\tilde{h}\hat{n}^2 \right](\vec{\nabla}\chi - 2e\vec{A})^2, \tag{22}$$

where we also introduced the external electromagnetic vector potential $\vec{A}$. The expression in the parenthesis is positive since it is minus the negative condensation energy Eq. (21) due to the hopping term. Thus our solution is stable against local χ_r fluctuations, and it follows that we have phase coherence in a charge $2e$ field, and thus superconductivity.

V. MEAN-FIELD ANALYSIS

The mean-field Hamiltonian can be analyzed by several equivalent methods. In the spirit of what was just developed, we could, e.g., make a Bogoliubov-Valatin canonical transformation and minimize the energy of the retransformed vacuum. This variational procedure would precisely correspond to the canonical transformation $e^{iG'_u}$ alluded to earlier. The method we actually use generalizes easily to finite temperatures and arbitrary large number of terms in the polynomial expansion of the mean-field Hamiltonian. It uses that the density matrix $\rho = e^{\beta[\Omega(T,\mu)-(H-\mu N)]}$ minimizes the free energy $F=\langle H-\mu N\rangle_\rho - kT\langle S\rangle_\rho$ for all values of ρ ($e^{-\beta\Omega}$ is the partition function and S the entropy). We can then take ρ to be the exponential of an expression linear in $\hat{n}_r$, $\hat{\Delta}_{rr'}$, $\hat{\Delta}_r$, and $h_{r,r'}$, where the prefactors are varied to minimize F. This method yields the ordinary BCS theory when applied to a Hamiltonian of the form in Eq. (1) and gives a more complicated self-consistent calculation when more terms are kept.

We have performed the mean-field analysis numerically, both using the truncated expressions given explicitly above and the full mean-field theory containing polynomials to seventh order. Since we construct an effective Hamiltonian, we define the hatted operators whose expectation values give the values $\hat{\Delta}_r^\dagger$ in Eq. (18). The corresponding operators $\hat{\Delta}_r^\dagger, \hat{\Delta}_{r,r'}^\dagger, \hat{h}$ are therefore formally $\hat{\Delta}_r^\dagger = e^{-i\mathcal{G}_u}\Delta_r^\dagger e^{i\mathcal{G}_u}$, etc. but in the calculation this involves simply reinterpreting the original operator in Eq. (16) in terms of quasiparticle fermion operators. For the density matrix $\rho \propto e^{-\beta H_{\mathrm{eff}}}$ we choose the Hamiltonian $H=H_{\mathrm{eff}}$ as

$$H_{\mathrm{eff}} = \sum_{r,r'} (t' h_{rr'} + 2^{3/2}\gamma\Delta_{rr'}^\dagger) - \sum_r (\mu' n_r + 2^{3/2}\gamma_2\Delta_r^\dagger) + (CC). \tag{23}$$

Our approximation is reasonable for small doping, and we confine the mean-field analysis to this regime. We let $\epsilon_k = (2-\cos k_x - \cos k_y)$ in two dimensions with a similar expression for $D=3$. With $\mu' = Dt' + \delta_\mu$ and $\gamma_2 = D\gamma + \delta_\gamma$ we define $e_k = t'\epsilon_k + \delta_\mu$, $d_k = \gamma\epsilon_k + \delta_\gamma$, and $E_k = \sqrt{e_k^2 + d_k^2}$. The energy gap Δ is the minimum in E_k, and it is easy to verify that for small doping an excellent approximation is given by $\Delta = |\gamma\delta_\mu - \delta_\gamma t'|/\sqrt{\gamma^2 + (t')^2}$. We define the momentum space sums as $f(\tilde{k}) = \Sigma_k[|t'|f(k)/E_k]$. Taking into account that there are four spin values we find the following expression for the doping $\delta \approx \hat{n}$, where expectation values of operators are dropped when the context is clear,

$$\hat{n} = 4 \times \frac{1}{2} \int \left(1 - \frac{e_k}{E_k}\right) d^2k = 2 - 2\tilde{e}_k/|t'|. \tag{24}$$

The expressions for t' and γ can be read off to lowest order from Eq. (21)

$$t' = -\frac{1}{2}t(1-\hat{n}), \tag{25}$$

$$\gamma = -\frac{5\hat{n}t}{4\sqrt{2}}, \tag{26}$$

while the definition of the on-site s-wave-order parameters can be read off from Eq. (13)

$$D\gamma - \delta_\gamma = 2\sqrt{2}U\langle\Delta_r\rangle. \tag{27}$$

In writing Eq. (23), we neglected the nonlocal repulsive interactions of type $n_r n_{r'}$ that will certainly be present in any realistic model with screened Coulomb interaction. Assuming a nearest-neighbor term $U_1 n_r n_{r'}$ and using the identity $\langle n_r n_{r'}\rangle = 1 + \frac{1}{2}\hat{\Delta}_{r,r'}\hat{\Delta}_{r,r'}^\dagger - \frac{1}{4}h_{r,r'}h_{r',r} - 2(\hat{n}_r + n_{r'}) + \frac{5}{4}(n_r^2 + n_{r'}^2) + n_r n_{r'}$, Eq. (26) would change to

$$\gamma = -\frac{5\hat{n}t}{4\sqrt{2}} + \sqrt{2}U_1\tilde{\Delta}_{r,r'}^\dagger. \tag{28}$$

Below we argue that this would not qualitatively change our conclusions.

As usual, self-consistency implies a gap equation which reads

$$\langle\Delta_r\rangle = -\frac{1}{\sqrt{2}t'}\tilde{d}_k. \tag{29}$$

After expanding to lowest order in δ and doing some algebra this can be recast as

$$\frac{t}{4U}\left(\frac{\Delta}{\sqrt{2}t} - \frac{5D\delta}{8}\right) = \frac{5\delta}{8} - \frac{\Delta}{t\sqrt{2}}\tilde{1}, \tag{30}$$

which is a closed equation that can be used to find the physical gap Δ as a function of δ.

Using the self-consistent equations, we can find expressions for the extended and onsite s-wave pairing amplitudes

$$\langle t\Delta_{r,r'}\rangle \approx \sqrt{2}\Delta(\tilde{1} - 1/D),$$

$$\langle\Delta_r\rangle \approx \frac{1}{2U}\left(\frac{\Delta}{\sqrt{2}} - \frac{5D\delta t}{8}\right). \tag{31}$$

Thus, not surprisingly we find that the onsite s-wave component is reduced by a factor t/U relative to the extended component.

We can now understand what would be the qualitative effect of adding extra repulsive interactions corresponding to the redefinition (28) of the variational parameter γ. For large U_1, γ will effectively be put to zero corresponding to $\tilde{\Delta}_{r,r'}$

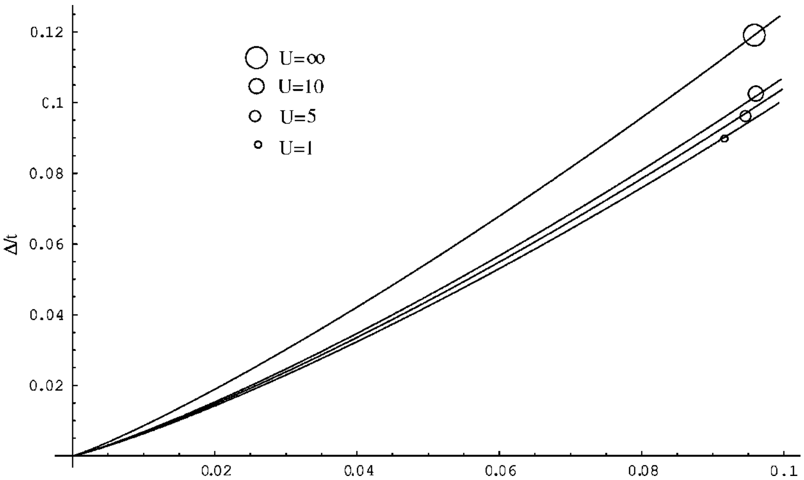

FIG. 1. Values of Δ/t as a function of doping δ for $U=(\infty,10,5,1)$ for $\delta \leqslant 0.1$, with the gap decreasing monotone with increasing U at given δ.

$\sim(t'/U_1)\hat{n}$, rather than $\tilde{\Delta}_{r,r'} \sim \delta$. We see that the scale of $\tilde{\Delta}_{r,r'}$ changes but it is still nonzero for arbitrary small doping.

A. Asymptotic behavior of the gap in $D=2$

In two dimensions, the expression (24) can be approximated by

$$\hat{n} = \approx \frac{1}{\pi|t'|}(\delta_\mu + \sqrt{\delta_\mu^2 + \Delta^2}). \tag{32}$$

The expression for $\tilde{1}$ is logarithmically singular but can be approximated by

$$\tilde{1} \approx \frac{-1}{2\pi} \ln\left(\frac{-\delta_\mu + \sqrt{\delta_\mu^2 + \Delta^2}}{32|t'|}\right) = \frac{1}{2\pi} \ln\left(\frac{32\hat{n}\pi(t')^2}{\Delta^2}\right), \tag{33}$$

where Eq. (32) was used. In spite of the logarithmic singularity, the self-consistent equation (30) can be solved in closed form. Defining $q=\Delta/(t\delta)$ the (inverse) equation is

$$\delta \approx \frac{8\pi}{q^2} e^{\pi[4q-5\sqrt{2}(1+2U/t)]/8qU/t}. \tag{34}$$

By plotting the pairs $(\delta, q\delta)$ according to the above formula as a function of q, we find the gap as a function of δ, shown for values of $U=(\infty,10,5,1)$ in Fig. 1. We can see an almost linear behavior of the gap as a function of doping that is quite insensitive to the value of U. For all values of δ and U the approximation $\Delta \approx t\delta$ is surprisingly good. A comparision with the numerical solution of the self-consistent equations is shown in Fig. 2.

B. Asymptotic solution for $D=3$ and small doping

In the case $D=3$, the vanishing density of states near $k=0$ makes the integral $\tilde{1}$ converge. In this case the self-consistent equation is Eq. (30) with $D=3$, and constant $\tilde{1}$. However, due to the vanishing density of states, even relatively small values of density lead to quite substantial values of μ and δ so the asymptotic value of this equation is far from being reached even for doping as low as 0.01. The corrections to $\tilde{1}$ are rather slowly varying, so the linear dependence of the gap upon doping is obtained for the $D=3$ case as well, as shown in Fig. 2.

VI. EFFECTIVE THEORY FOR SMALL δ

After applying a transformation that rigorously preserves the full Fock space, we have obtained a superconducting behavior for the doped spin 3/2 Mott insulator by using

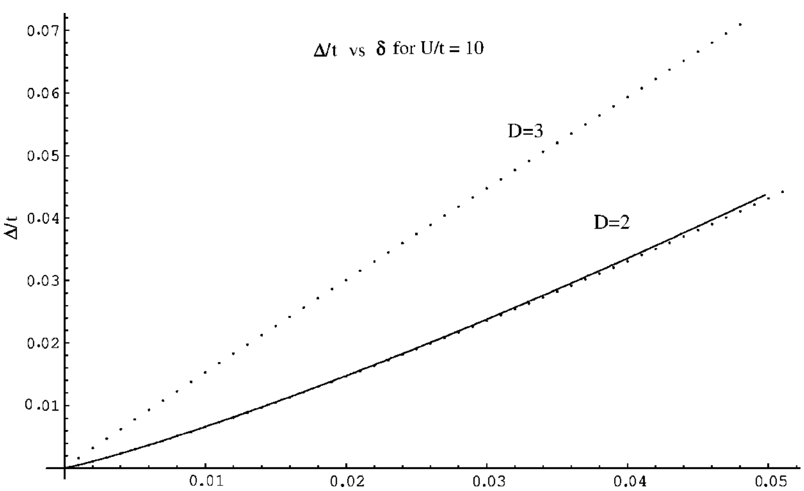

FIG. 2. Values of Δ/t as a function of doping δ for $U/t=10$ for $\delta<0.05$. The upper curve is for $D=3$ and the lower data points for $D=2$. The solid line is the fit to the $2D$ asymptotic curve according to Eq. (34). The 3D fits with no visible error to the curve $\Delta/t=1.4848\delta$ corresponding to $\tilde{1}=.6137$.

standard mean-field theory methods. Here we contrast this to an effective theory for the Hamiltonian in Eq. (1), derived in the limit of small doping $\delta \ll 1$ and small hopping $t \ll U, J$, using the more conventional approach of projection on a low-energy subspace. Ignoring hopping $t=0$, this low-energy sector consists of states where each site is occupied by either a Mott singlet $|\Phi_s\rangle$, which we choose as the Fock vacuum $|\Phi_s\rangle \to |0\rangle$ or by a single charge $|s\rangle = c_s^\dagger|0\rangle$ with spin 3/2 ($s = \pm 1/2, \pm 3/2$)—all other states are separated from these by a gap of order $X \sim U, J$. Restricting to this low-energy sector and including the hopping in perturbation theory gives to order t^2/X, the effective Hamiltonian

$$H_{\text{eff}} = -\tilde{t}\sum_{r,\delta,s} c_{sr}^\dagger c_{s,r+\delta} + \sum_{r,\delta,\alpha,\alpha',\beta,\beta'} \tilde{J}_{\alpha\alpha'\beta\beta'} c_{\alpha,r}^\dagger c_{\alpha' r} c_{\beta r+\delta}^\dagger c_{\beta',r+\delta} \tag{35}$$

subject to the constraint $\Sigma_s c_{sr}^\dagger c_{sr} \leq 1$ [$\tilde{J}_{\alpha\alpha'\beta\beta'}$ are SU(2) scalars]. This *t*-*J*-type model describes four species of fermions with nearest-neighbor hopping ($\tilde{t} \sim t$), nearest-neighbor exchange couplings ($\tilde{J} \sim t^2/X$), and with the hard-core constraint that no two fermions occupy the same site. Second-order perturbation theory guarantees a finite, albeit weak, attraction which opens the possibility of having a superconducting phase even for small doping. From this approach, however, we would expect such a phase to be destroyed by a nearest-neighbor repulsion that is normally present in a realistic model. Thus, our previous mean-field calculation is at odds with this approach. If the former turns out to be valid, it suggests that there are nonperturbative effects due to the hard-core constraints that are not easily accounted for in the conventional formulation. If, on the other hand, the hard-core constraints are not very important and the naive picture of four different species of weakly interacting fermions is essentially correct, it would suggest that our mean-field treatment of the phase phase fluctuations does not capture the correct physics.

VII. DISCUSSION AND SUMMARY

A. The anomalous term $\Delta_{r,r'} n_r$

We see from Eq. (15) that in order to have a nonzero doping $\delta = 2 - \langle n \rangle$, we *must* have $\hat{n} > 0$, in fact $\hat{n} \approx \delta$ to lowest order in δ. Energetically we will also have $\hat{h} \neq 0$ for finite δ. According to Eq. (20) the extended *s*-wave pairing field $\hat{\Delta}_{r,r'}$ cannot vanish and in fact will be proportional to doping. This in turn generates a (much smaller) on-site pairing $\hat{\Delta}_r$ through the self-consistent equations. Note that this pairing field can never completely vanish because of the linear coupling to higher order terms. At finite temperature the mean-field theory will presumably eventually break down via an *xy* transition due to phase fluctuations that we have not taken into account. This has been discussed in a series of recent papers where the term "gossamer superconductivity"[16–18] has been used to describe a similar scenario.

It is admittedly not easy to understand the physical origin of these new anomalous terms of the type $\Delta_{r,r'} n_r$. On a technical level, they are forced by the fermion statistics which constrains the form of the canonical transformation necessary to map a local Mott ground state to the vacuum. In our case, this transformation must be (a) nonlinear in fermion operators and (b) charge nonconserving. Property (a) yields an effective interaction from the hopping term near a charged ground state and property (b) makes this interaction nongauge invariant. Property (a) is a necessary consequence of mapping a locally entangled state to the vacuum and property (b) is a consequence of mapping a charged state to the vaccum which breaks gauge symmetry. Very general arguments relying on long-range phase coherence and a finite range gap function then predict that the system should be a superconductor.[19] Our mean-field calculation, which suggests a superconducting ground state, supports this picture, given our assumptions about phase coherence.

We already pointed out the contradiction between our main result and what would be expected based on a conventional analysis of the type leading to Eq. (35), but also stressed the difficulties related to the hard core constraints inherent in the latter approach. Here we should note that more elaborate schemes for dealing with these nonholonomic hard core constraints face severe difficulties related to phase fluctuations. For example, in the spin-1/2 Hubbard model at half filling, one can turn the no double occupancy constraint into a holonomic gauge constraint by introducing spinons and holons. The resulting phase depends crucially on the fluctuations in the related gauge fields. By working in the full Hilbert space, we avoid these difficulties, but nevertheless our conclusions are still dependent on certain assumtions about phase coherence. Without a more sophisticated analysis of the phase fluctuations, we cannot rule out that these will be important and, e.g., destroy the superconduting state at low doping.

B. Range of validity and applicability

We now assume that our analysis is correct at low doping, and discuss its range of validity and applicability. At sufficiently large value of doping, the theory will yield a free energy which is unfavorable compared to that of a doped $n=1$ Mott insulator. The mean-field picture suggests there will be a coexistence region where a slightly hole doped $n=2$ Mott insulator will coexist with a hole doped $n=1$ Mott insulator. The $n=1$ Mott insulator will presumably have some sort of magnetic order at low temperature that breaks the large spin degeneracy of the uncorrelated odd filling Mott insulator. If a coexistence region really exists, or whether an intermediate phase which breaks translational invariance may exist, is beyond the scope of the present analysis. Our calculation thus makes assumptions about U which leave open the question if this behavior could really be seen in a physical system. On the one hand, U must be large enough (and J even larger) so that a Mott insulating state occurs at $n=2$ and furthermore triply occupied sites are effectively absent. On the other hand, U must be small enough so that the correlated state will have lower energy than a mixed state with an $n=1$ Mott insulator and an $n=2$ Mott insulator. The case $J<0$ or $J<U$ is briefly addressed in Sec. III.

Finally, it is relevant to ask whether the transformation used for the spin-3/2 case could be applied to the spin-1/2 systems. First consider the canonical transformation which maps between these the $n=0$ and $n=2$ states. This is the ordinary particle-hole transformation which is not charge conserving. However, the doubly occupied singlet is created by $c^{\dagger}_{\uparrow,r}c^{\dagger}_{\downarrow,r}|0\rangle$ and hence is factorizable in fermion operators. The canonical transformation is therefore linear and no new interaction terms are introduced in the transformation. The physical properties of the system are symmetric under charge conjugation, which is sufficient for the particle-hole transformation not to generate any new behavior and the present analysis is uninteresting. In the case of the half-filled Hubbard model, the Mott ground state corresponds to one electron per site. This cannot be mapped to the vacuum through a canonical transformation without violating the Fermi anticommutation relations.[15]

C. Summary

We have presented a new type of canonical transformation for the half-filled spin-3/2 Hubbard model that maps the Mott insulator at half filling to the vacuum. This canonical transformation is straighforward to generalize to multiband Hubbard models with a local spin singlet Mott insulating ground state. At finite doping, a self-consistent mean-field theory for such a system results in a phase with long-range phase coherence. An order parameter that is usually identified with extended s-wave superconductivity appears and is proportional to doping. We note that the resulting picture resembles the "gossamer superconductor" scenario that has been recently introduced by Laughlin and co-workers. Our approach appears to be in contradiction to other methods of attacking these kinds of problems, and we pointed out the difficulties with both approaches. Although we have only explored a specific spin-3/2 Hubbard model at half filling, we believe that our method could be useful for a variety of similar models with locally entangled Mott insulating ground states.

*Electronic address: ostlund@fy.chalmers.se
†Electronic address: hansson@physto.se
‡Electronic address: ak@physto.se

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[20] Another term which can be considered is of course S_r^2, but for spin 3/2 this obeys $P_r^2=\frac{1}{3}S^2-\frac{5}{2}n+\frac{5}{4}n^2$, so there is no loss of generality in the onsite term of our Hamiltonian aside from ignoring the possibility of a term proportional to n^3.
[21] In the weak interaction limit ($J\ll t$) conventional mean-field methods should be reliable and the $J<0$ problem has been addressed by T. L. Ho *et al.* (Ref. 11) and Wu *et al.* (Ref. 12) however, note that in our work we focus on the opposite limit of t small compared to J.