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Bond-operator representation of quantum spins: Mean-field theory of frustrated quantum Heisenberg antiferromagnets

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We introduce a new representation of $S=\frac{1}{2}$ quantum spins in terms of bond operators. The bond operators create and annihilate singlet and triplet bonds between a pair of spins. The representation is useful in describing the transition between dimerized and magnetically ordered phases of quantum antiferromagnets. It is used to obtain a mean-field theory of the two-dimensional frustrated quantum Heisenberg antiferromagnets considered recently by Gelfand, Singh, and Huse. The method should also be useful in the analysis of random quantum antiferromagnets.

I. INTRODUCTION

The discovery of high-temperature superconductivity in $La_{2-x}Sr_xCuO_4$ has led to a resurgence of interest in the properties of two-dimensional quantum Heisenberg antiferromagnets.[1] There now appears to be a consensus that the nearest-neighbor $S=\frac{1}{2}$ SU(2) Heisenberg antiferromagnet on a square lattice displays long-range Néel order in its ground state.[2] Attention has therefore focused on different methods of perturbing the nearest-neighbor model to enhance the effect of quantum fluctuations, leading to a destruction of magnetic order in the ground state.[3–8] There is evidence[4,9] that nearest-neighbor, square lattice SU(N) antiferromagnets with quadratic exchange have two candidate ground states: (i) a Néel ordered state and (ii) a quantum disordered state that is *spontaneously dimerized* for $2S \neq 0 \pmod 4$. Gelfand *et al.*[5] have recently examined *frustrated* $S=\frac{1}{2}$, SU(2) quantum antiferromagnets on a square lattice by a series expansion method. They found evidence for spontaneous dimerization in the ground state. Additional supporting evidence for spontaneous dimerization in frustrated SU(2) antiferromagnets has emerged in the exact diagonalization studies of Dagotto and Moreo.[6] (We note, however, that alternative proposals on the nature of the quantum disordered phase have also been made.[8]) Dimerized phases were actually considered several years ago by Bhatt and Lee[10] in their study of *random* spin-$\frac{1}{2}$ quantum antiferromagnets which modeled the insulating phase of doped semiconductors. We note that, in contrast to the square-lattice models just discussed, the dimerization in these random antiferromagnets is not spontaneous in that no symmetry of the Hamiltonian is broken by the ground state.

In this paper we shall introduce a new bond-operator representation of quantum $S=\frac{1}{2}$ spins that is specifically designed to understand the properties of dimerized phases. The method applies to both spontaneously and intrinsically dimerized phases. It also displays the boundary of instability of these phases to magnetically ordered states. We will illustrate the method by applying it to frustrated antiferromagnets considered by Gelfand *et al.*[5] These are square-lattice antiferromagnets with first (J_1), second (J_2), and third (J_3) neighbor exchange interaction. In addition a parameter λ multiplies some of the interactions and is used to interpolate between an exactly solvable dimerized limit $\lambda=0$ and the limit with the full symmetry of the square lattice $\lambda=1$. Our results are summarized in Figs. 3, 4, and 5 and are in close agreement with those of Ref. 5. Promising applications of the methods of this paper to random antiferromagnets also exist but will be postponed for future investigation.

The outline of the rest of the paper is as follows: In Sec. II we introduce the bond-operator representation. Sections III and IV presents results of a mean-field theory calculation on the frustrated quantum Heisenberg antiferromagnets considered by Gelfand *et al.*[5] Finally, Sec. V recapitulates and outlines proposals for further work.

II. BOND OPERATORS

We introduce the representation by considering two $S=\frac{1}{2}$ spins, $\mathbf{S}_1$ and $\mathbf{S}_2$. The four states in the Hilbert space can be combined to form the singlet state $|s\rangle$ and the three triplet states $|t_x\rangle$, $|t_y\rangle$, and $|t_z\rangle$. We introduce singlet and triplet creation operators that create these states out of the vacuum $|0\rangle$. Thus

$$\begin{aligned} |s\rangle &= s^\dagger|0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)\,, \\ |t_x\rangle &= t_x^\dagger|0\rangle = \frac{-1}{\sqrt{2}}(|\uparrow\uparrow\rangle - |\downarrow\downarrow\rangle)\,, \\ |t_y\rangle &= t_y^\dagger|0\rangle = \frac{i}{\sqrt{2}}(|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle)\,, \\ |t_z\rangle &= t_z^\dagger|0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)\,. \end{aligned} \tag{2.1}$$

By considering the various matrix elements $\langle s|\mathbf{S}_1|t_\alpha\rangle$, $\langle s|\mathbf{S}_2|t_\alpha\rangle, \ldots,$ it can be seen in a straightforward manner that the action of $\mathbf{S}_1$ and $\mathbf{S}_2$ on the singlet and triplet states leads to the representation

$$S_{1\alpha}=\tfrac{1}{2}(s^\dagger t_\alpha+t_\alpha^\dagger s-i\epsilon_{\alpha\beta\gamma}t_\beta^\dagger t_\gamma)\ , \tag{2.2}$$

$$S_{2\alpha}=\tfrac{1}{2}(-s^\dagger t_\alpha-t_\alpha^\dagger s-i\epsilon_{\alpha\beta\gamma}t_\beta^\dagger t_\gamma)\ , \tag{2.3}$$

where α,β,γ take the values x,y,z, repeated indices are summed over and ϵ is the totally antisymmetric tensor. The restriction that the physical states are either singlets or triplets leads to the constraint

$$s^\dagger s+t_\alpha^\dagger t_\alpha=1\ . \tag{2.4}$$

So far we have not specified the algebra of the s and t_α operators. it turns out that either the canonical commutation ("bosonic") or the canonical anticommutation ("fermionic") relations reproduce the correct algebra of the spin operators. However, the anticommuting operators were not found to lead a physically meaningful mean-field theory and will therefore not be discussed further. We will therefore use operators satisfying the bosonic commutation relations

$$[s,s^\dagger]=1,\ \ [t_\alpha,t_\beta^\dagger]=\delta_{\alpha\beta},\ \ [s,t_\alpha^\dagger]=0\ . \tag{2.5}$$

Now it can be verified that as a consequence of the representation [(2.2) and (2.3)], the constraint [(2.4)], and the commutation relations [(2.5)], we have

$$\begin{aligned}&[S_{1\alpha},S_{1\beta}]=i\epsilon_{\alpha\beta\gamma}S_{1\gamma},\ \ [S_{2\alpha},S_{2\beta}]=i\epsilon_{\alpha\beta\gamma}S_{2\gamma}\ ,\\&[S_{1\alpha},S_{2\beta}]=0\ ,\\&\mathbf{S}_1^2=\tfrac{3}{4},\ \ \mathbf{S}_2^2=\tfrac{3}{4}\ ,\\&\mathbf{S}_1\cdot\mathbf{S}_2=-\tfrac{3}{4}s^\dagger s+\tfrac{1}{4}t_\alpha^\dagger t_\alpha\ .\end{aligned} \tag{2.6}$$

These are just the properties expected of spin-$\frac{1}{2}$ SU(2) operators.

The group-theoretical interpretation of this representation is straightforward. The tensor product of the two spin-$\frac{1}{2}$ states on sites 1 and 2 can be considered as a realization of a representation of the group SU(2)×SU(2). The well-known homomorphism[11] between SU(2)×SU(2) and SO(4) is now realized by noting that $\mathbf{S}_1+\mathbf{S}_2$ and $\mathbf{S}_1-\mathbf{S}_2$ are the usual generators of SO(4) "rotations" and "translations." From Eqs. (2.2) and (2.3) we now see that the s and t_α bosons merely yield the canonical "Schwinger boson" representation of the generators $\mathbf{S}_1+\mathbf{S}_2$ and $\mathbf{S}_1-\mathbf{S}_2$ of SO(4). Moreover, the constraint (2.4) is invariant under SO(4) transformations.

The application of the bond-operator formalism to the lattice begins by the choice of a close-packed dimerization of the lattice. The spin operators on the sites are now expressed in terms of the singlet and triplet bosons on the dimer connected to the site. The choice of the lattice dimerization necessarily breaks the symmetry of any Hamiltonian with the full symmetry of the lattice. In any exact calculation this should have no effect on the form of the final result. The symmetry will, however, not be fully restored in an approximate mean-field calculation. For the calculations in this paper to be meaningful for Hamiltonians with the full symmetry of the square lattice, it is necessary that the magnetically disordered phase be spontaneously dimerized. As noted in Sec. I, there is evidence[4–6] that such a spontaneous dimerization does occur. For Hamiltonians that do not have the full symmetry of the lattice, or for random systems, there is no such arbitrariness in the choice of dimerization. It is then appropriate to choose a dimerization in which the exchange constants on the dimers are as large as possible and lead to the smallest ground-state energy. Our calculations yield two different classes of ground states.

(i) *Dimerized phases.* These are magnetically disordered states. The dimerization can be either spontaneous or intrinsic. Such states have the following anomalous expectation values:

$$\langle s\rangle\neq 0,\ \ \langle t_\alpha\rangle=0,\ \ \langle t_\alpha t_\beta\rangle=C\delta_{\alpha\beta}\ , \tag{2.7}$$

where C is a nonzero constant. The relationships imply that the t_α bosons have condensed in *pairs*, but there is no single t_α boson condensation.

(ii) *Magnetically ordered phases.* Condensation of *single* t_α bosons leads to long-range magnetic order. The anomalous expectation values are now

$$\langle s\rangle\neq 0,\ \ \langle t_\alpha\rangle\neq 0\ . \tag{2.8}$$

The wave vector and polarization of the mode at which the t_α bosons condense determines the nature of the magnetic ordering. We note, however, that the mean-field approach of this paper does not restore the equivalence (if any) between the bonds connected to a site, and will always lead to a coexistence of dimerization and magnetic order.

III. MEAN-FIELD THEORY

This section will use the bond-boson representation of Sec. II to developed a mean-field theory of the frustrated square-lattice antiferromagnetic of Gelfand *et al.*[5] The definition of these models requires the choice of a dimerization $\mathcal{D}$ of the square lattice. All of the calculations in this paper will use either the "columnar" [$\mathcal{D}=\mathcal{D}_c$, Fig. 1(a)] or "staggered" [$\mathcal{D}=\mathcal{D}_s$, Fig. 1(b)] dimerizations. Each dimerization $\mathcal{D}$ is now associated with a square-lattice antiferromagnet

$$H_{\mathcal{D}}=J_1\sum_{\langle ij\rangle\in\mathcal{D}}\mathbf{S}_i\cdot\mathbf{S}_j+\lambda\left[J_1\sum_{\langle ij\rangle\notin\mathcal{D}}\mathbf{S}_i\cdot\mathbf{S}_j+J_2\sum_{2\mathrm{NN}}\mathbf{S}_i\cdot\mathbf{S}_j+J_3\sum_{3\mathrm{NN}}\mathbf{S}_i\cdot\mathbf{S}_j\right]\ , \tag{3.1}$$

where $\langle ij\rangle$ denotes nearest neighbors, 2NN denotes second neighbors (e.g., i and $i+\hat{x}+\hat{y}$), 3NN denotes third neighbors (e.g., i and $i+2\hat{x}$), and J_1,J_2,J_3 are positive exchange constants.[12] The parameter λ controls the strength of the intrinsic dimerization. At $\lambda=0$, $H_{\mathcal{D}}$ consists of independent dimers and is therefore trivially solvable. At $\lambda=1$, the full symmetry of the square lattice is restored and the properties of $H_{\mathcal{D}}$ should be independent of $\mathcal{D}$.

We now substitute the operator representation in Eq. (2.2) on the sites of the A sublattice and the representation in Eq. (2.3) on the sites of the B sublattice into the Hamiltonian $H_{\mathcal{D}}$. The bond bosons are chosen to reside on a dimerization $\mathcal{D}'$ which is not necessarily the same as $\mathcal{D}$. It is clear that for small λ, the choice $\mathcal{D}=\mathcal{D}'$ will minimize the energy; however, it is possible that for values of λ near 1, a different choice may be appropriate. This procedure transforms $H_{\mathcal{D}}$ into the following rather complicated form

$$\begin{aligned}
H_{\mathcal{D}}&=H_0+H_1+H_2+H_3\ ,\\
H_0&=\sum_{i\in\mathcal{D}'} J_i(-\tfrac{3}{4}s_i^\dagger s_i+\tfrac{1}{4}t_{i\alpha}^\dagger t_{i\alpha})-\sum_{i\in\mathcal{D}'}\mu_i(s_i^\dagger s_i+t_{i\alpha}^\dagger t_{i\alpha}-1)\ ,\\
H_1&=\sum_{i,j\in\mathcal{D}'} a(i,j)(t_{i\alpha}^\dagger t_{j\alpha}s_j^\dagger s_i+t_{i\alpha}^\dagger t_{j\alpha}^\dagger s_j s_i+\text{H.c.})\ , \qquad (3.2)\\
H_2&=\sum_{i,j\in\mathcal{D}'} b(i,j)(i\epsilon_{\alpha\beta\gamma}t_{j\alpha}^\dagger t_{i\beta}^\dagger t_{i\gamma}s_j+\text{H.c.})\ ,\\
H_3&=\sum_{i,j\in\mathcal{D}'} c(i,j)(t_{i\alpha}^\dagger t_{j\alpha}^\dagger t_{j\beta}t_{i\beta}-t_{i\alpha}^\dagger t_{j\beta}^\dagger t_{j\alpha}t_{i\beta})\ .
\end{aligned}$$

We have introduced site-dependent chemical potentials μ_i to impose the constraint (2.4) and the functions a,b,c, which depend on $\mathcal{D},\mathcal{D}'$ and the exchange constants.

The mean-field theory of $H_{\mathcal{D}}$ is obtained by decoupling the quartic terms to yield an effective quadratic Hamiltonian $H_{m\mathcal{D}}$. For simplicity we will limit further discussion in the next two paragraphs to the cases $\mathcal{D}=\mathcal{D}'=\mathcal{D}_s$ or $\mathcal{D}_c$. In this case we can use the translation invariance of the problem to perform a Fourier transformation with a single bond per unit cell.

Because of the $-(J_1/4)s^\dagger s$ term in H_0, it is clear that single s bosons will condense, $\langle s\rangle=\bar{s}$. The terms in H_1 will in turn lead necessarily to a nonzero expectation value of $\langle t_\alpha t_\alpha\rangle$. Moreover, the translation invariance of the problem implies that we may assume $\mu_i=\mu$, site independent. This procedure leads finally to

$$\begin{aligned}
H_{m\mathcal{D}}(\mu,\bar{s})=N&\left[-\frac{3J_1}{4}\bar{s}^2-\mu\bar{s}^2-\mu\right]\\
&+\sum_{\mathbf{k}}\left[\frac{J_1}{4}-\mu+2\lambda A_{\mathbf{k}}(\bar{s})\right]t_{\mathbf{k}\alpha}^\dagger t_{\mathbf{k}\alpha}\\
&+\lambda\sum_{\mathbf{k}}B_{\mathbf{k}}(\bar{s})(t_{\mathbf{k}\alpha}t_{-\mathbf{k}\alpha}+t_{\mathbf{k}\alpha}^\dagger t_{-\mathbf{k}\alpha}^\dagger)\ , \qquad (3.3)
\end{aligned}$$

where N is the number of dimers on the lattice. The functions $A_{\mathbf{k}},B_{\mathbf{k}}$ will have contributions from H_1 and H_3 while all the quadratic decouplings of H_2 are identically zero. $A_{\mathbf{k}}$ and $B_{\mathbf{k}}$ therefore depend upon $\langle t_{\mathbf{k}\alpha}t_{-\mathbf{k}\alpha}\rangle$ and $\langle t_{\mathbf{k}\alpha}^\dagger t_{\mathbf{k}\alpha}\rangle$, where the expectation values have to be determined self-consistently in the ground state of $H_{m\mathcal{D}}$. The parameters μ and $\bar{s}$ are determined by the saddle-point equations

$$\left\langle\frac{\partial H_{m\mathcal{D}}}{\partial\mu}\right\rangle=0,\quad \left\langle\frac{\partial H_{m\mathcal{D}}}{\partial\bar{s}}\right\rangle=0\ . \qquad (3.4)$$

This decoupling procedure just discussed is equivalent to determining the best variational wave function of the form

$$|\Omega\rangle=C'\exp\left[\sum_i\bar{s}s_i^\dagger-\sum_{\mathbf{k}}f_{\mathbf{k}}t_{\mathbf{k}\alpha}^\dagger t_{-\mathbf{k}\alpha}^\dagger\right]|0\rangle \qquad (3.5)$$

as the ground state of $H_{m\mathcal{D}}$, while satisfying the constraint (2.4) on the average.

The Hamiltonian $H_{m\mathcal{D}}$ can be diagonalized by Bogoliubov rotation $\gamma_{\mathbf{k}\alpha}=u_{\mathbf{k}}t_{\mathbf{k}\alpha}+v_{\mathbf{k}}t_{-\mathbf{k}\alpha}^\dagger$ leading to the result

$$H_{m\mathcal{D}}=E_G+2\sum_{\mathbf{k}}\omega_{\mathbf{k}}\gamma_{\mathbf{k}\alpha}^\dagger\gamma_{\mathbf{k}\alpha}\ , \qquad (3.6)$$

where

$$\omega_{\mathbf{k}}=\left[\left[\frac{J_1}{8}-\frac{\mu}{2}+\lambda A_{\mathbf{k}}\right]^2-(\lambda B_k)^2\right]^{1/2}\ . \qquad (3.7)$$

For small values of λ we will always find a solution of the self-consistency equations (3.4) such that $\omega_{\mathbf{k}}$ is real and positive everywhere in the Brillouin zone. Under such conditions, the system is in a *magnetically disordered phase* with $\langle t_\alpha\rangle=0$. It is possible, however, that at a critical value of $\lambda=\lambda_c,\omega_{\mathbf{k}}$ will vanish first at $\mathbf{k}=\mathbf{k}_0$. This will lead to a condensation of single t_α bosons at $\mathbf{k}=\mathbf{k}_0$ for $\lambda>\lambda_c$. At nonzero value of $\langle t_\alpha\rangle$ implies that the system is in a *magnetically ordered phase.* If $\epsilon_{\alpha\beta\gamma}\langle t_\beta^\dagger\rangle\langle t_\gamma\rangle=0$, we deduce from the bond-boson representation of the spins in Eqs. (2.2) and (2.3) that the wave vector for magnetic ordering is $\mathbf{k}_0+(\pi,\pi)$ [the (π,π) term arises from the relative minus sign between Eqs. (2.2) and (2.3)]. The mean-field theory in the magnetically ordered phase was obtained by taking the zero-temperature limit of a finite temperature theory. In $d=2$ there is no magnetic ordering at any finite temperature, and so the

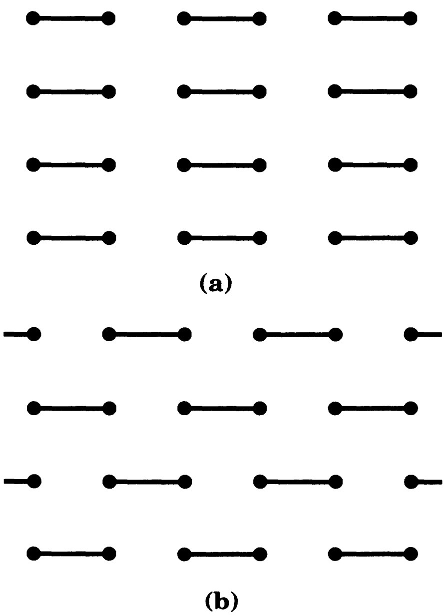

FIG. 1. (a) Columnar ($\mathcal{D}=\mathcal{D}_c$) and (b) staggered ($\mathcal{D}=\mathcal{D}_s$) dimerizations.

decouplings of the theory are identical to those already discussed. At $T=0$, $\langle t_\alpha \rangle$ acquires a nonzero expectation value.

The generalization of this formalism to the cases where $\mathcal{D} \neq \mathcal{D}'$ is straightforward. It is necessary to choose a larger unit cell, and the Bogoliubov rotation involves the *matrix* functions $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$. Details of this generalization are tedious and will not be presented explicitly. We turn instead to a presentation of the results of the numerical solution of the mean-field equations for various choices of the exchange constants and $\mathcal{D}$.

IV. NUMERICAL RESULTS

Numerical calculations were carried out to determine the best variational ground state of $H_{\mathcal{D}}$ for columnar ($\mathcal{D}=\mathcal{D}_c$) and staggered ($\mathcal{D}=\mathcal{D}_s$) dimerizations. For both choices of $\mathcal{D}$, we used mean-field wave functions obtained by placing the bosons on the bonds of the columnar ($\mathcal{D}'=\mathcal{D}_c$) or staggered ($\mathcal{D}'=\mathcal{D}_s$) dimerizations. The dimerization $\mathcal{D}'$, which minimized the energy, was chosen as the ground state. As already noted, for small values of λ, we expect $\mathcal{D}'=\mathcal{D}$. It is also clear that the procedure outlined cannot be reliable when λ is close to 1 *and* J_2 or J_3 is large. We then expect a dimerization of the lattice based upon second or third nearest neighbors to be appropriate; such ansatzes were not used in the calculations of this paper.

The staggered and columnar dimerizations are the only possible dimerizations of the lattice with a unit cell of two sites. Other dimerizations of the lattice with larger unit cells are shown in Fig. 2. It is clearly possible to extend the calculations of this paper to such dimerizations but the large increase in numerical complexity has kept us from doing so.

A further numerical simplification has been made in the solutions of the mean-field equations presented in the following: the dependence of $A_{\mathbf{k}}$ and $B_{\mathbf{k}}$ [Eq. (3.3)] on $\langle t_\alpha t_\alpha \rangle$ and $\langle t_\alpha^\dagger t_\alpha \rangle$ was omitted. This is expected to be a good approximation because $\bar{s}^2 \gg \langle t_\alpha^\dagger t_\alpha \rangle$ for small λ. This approximation is equivalent to omitting H_2 and H_3 from the Hamiltonian. All of the calculations in the magnetically disordered phases were repeated without this additional simplification and were found to lead to shifts in the phase boundaries and energies smaller than 15% at the largest values of λ (See Fig. 3). In the magnetically ordered phases, the dropping of H_2 and H_3 makes the energy independent of the polarization of $\langle t_\alpha \rangle$. The splitting of the polarization degeneracy by terms in H_2 and H_3 has not been numerically investigated, but, from our results on the magnetically disordered phases, is expected to have little effect on the phase boundary of the magnetically disordered region.

The most detailed calculations were carried out for $J_3=0$. The results for the columnar dimerization, $\mathcal{D}=\mathcal{D}_c$, are discussed in Sec. IV A and shown in Fig. 3, while those for the staggered dimerization, $\mathcal{D}=\mathcal{D}_s$, are discussed in Sec. IV B and shown in Fig. 4. Finally, results for nonzero J_3 but with $J_2=0$ and $\mathcal{D}=\mathcal{D}_c$ are discussed in Sec. IV C and shown in Fig. 5.

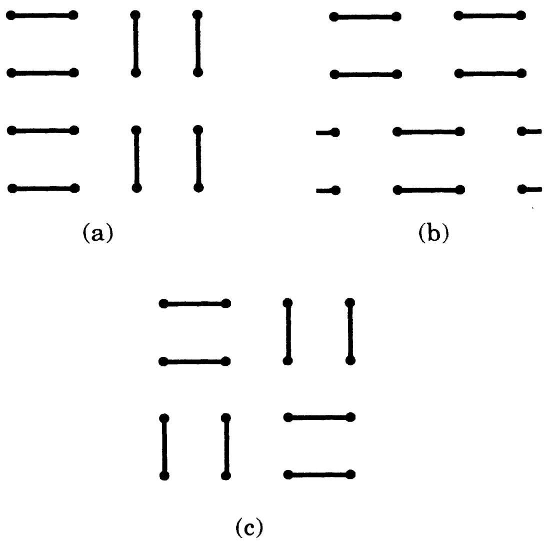

FIG. 2. Dimerizations with unit cells larger than two sites.

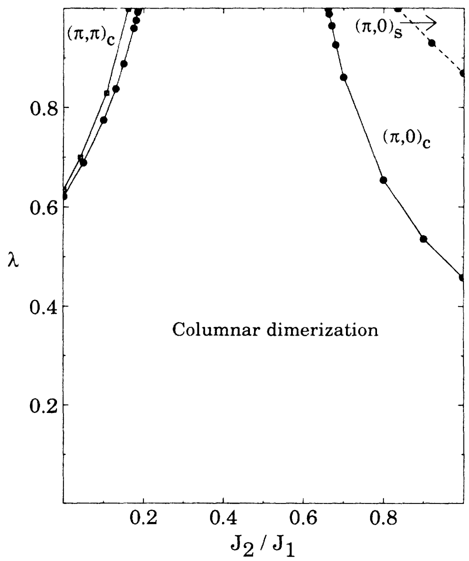

FIG. 3. Mean-field ground state of $H_{\mathcal{D}}$ for $\mathcal{D}=\mathcal{D}_c$, the columnar dimerization, and $J_3=0$. Only wave functions based upon bond bosons placed upon the dimers of $\mathcal{D}'=\mathcal{D}_c$ and $\mathcal{D}_s$ were examined. The magnetically ordered phases are denoted by the ordering wave vector and a subscript (c=columnar or s=staggered) denoting the type of coexisting dimerization. The solid circles denote the results of calculations carried out in the approximation that omitted H_2 and H_3. The open squares denote the position of the phase boundary between the magnetically disordered phase and $(\pi,\pi)_c$ ordering calculated with H_2 and H_3 included. The separation between the open-squares line and the solid-circles line is thus an estimate of the error due to the omission of H_2 and H_3. Solid lines denote second-order transitions, while dashed lines denote first-order transitions. The magnetically ordered phases break spin-rotation symmetry, while the $(\pi,0)_s$ phase and entire line $\lambda=1$ break square-lattice symmetries.

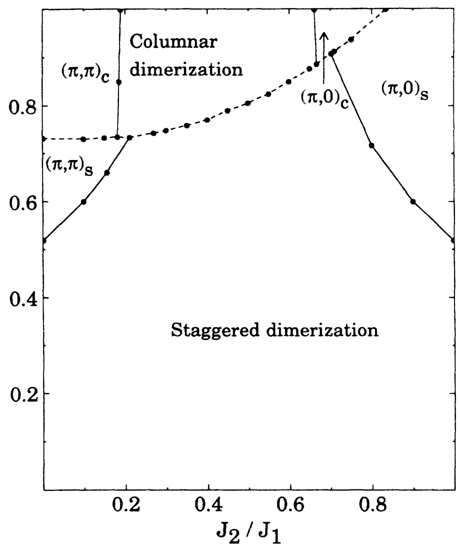

FIG. 4. As in Fig. 3 but for $\mathcal{D}=\mathcal{D}_s$, the staggered dimerization and $J_3=0$. Unlike Fig. 3, there is a region where the magnetically disordered state has $\mathcal{D}'=\mathcal{D}_c\neq\mathcal{D}$. The state with columnar dimerization, the states $(\pi,\pi)_c$ and $(\pi,0)_c$ and the line $\lambda=1$ all break lattice symmetries. The Hamiltonian is invariant under the full space group of the square lattice at $\lambda=1$: Notice the identity of the lines $\lambda=1$ between this figure and Fig. 3.

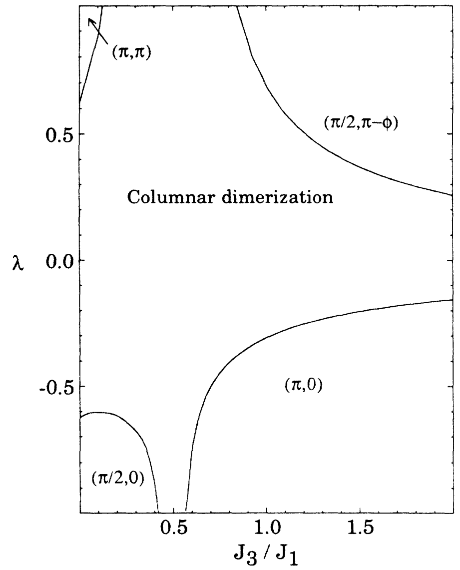

FIG. 5. Mean-field ground state of $H_{\mathcal{D}}$ as a function of J_3/J_1 and λ for $J_2=0$ and $\mathcal{D}$=columnar dimer configuration in Fig. 1(a). Unlike Figs. 3 and 4, the calculations have been performed *only* in the disordered phase. The dark lines denote instabilities towards the appearance of magnetic order. The ordered phase in the upper right is incommensurate and the ordering wave vector at the phase boundary is $(\pi/2,\pi-\phi)$ where $\phi=\cos^{-1}(J_1/4J_3)$.

A. $\mathcal{D}=\mathcal{D}_c, J_3=0$

The results of the calculation on $H_{\mathcal{D}}$ with $\mathcal{D}=\mathcal{D}_c$, the columnar dimerization are shown in Fig. 3. Wave functions based upon the columnar dimerization ($\mathcal{D}'=\mathcal{D}_c$) were the lowest-energy states over the entire phase diagram, except for a small region in the vicinity of $\lambda=1$ and $J_2/J_1=1$ where, as already noted, the mean-field theory is not expected to be reliable.

The magnetically disordered columnar phase is stable for all $0\leq J_2/J_1\leq 1$ for $\lambda<0.46$. The solid lines indicate *second-order* transitions between the disordered and magnetically ordered phases. The spin ordering appears at wave vectors $\mathbf{k}=(\pi,\pi)$ at small values of J_2/J_1 and at $\mathbf{k}=(\pi,0)$ for J_2/J_1 near 1 and is driven by Bose condensation of the t_α bosons at the appropriate wave vectors. The dimerization order persists everywhere in the magnetically ordered phase; this dimerization is columnar except for a first-order transition into a staggered phase in the vicinity of $\lambda=1$, $J_2/J_1=1$. For $\lambda\neq 1$, the columnar order is natural and does not imply the existence of any additional broken symmetry. However, the coexistence of columnar and magnetic order at $\lambda=1$ does imply the existence of a broken lattice symmetry. The formalism of this paper clearly overestimates the tendency of the system to dimerize: Thus the results of this paper cannot conclusively settle the issue of whether spontaneous dimerization can occur in a Néel state.

There is a regime $0.19<J_2/J_1<0.66$ in Fig. 3, where the columnar dimerization, with no coexisting magnetic order, is stable at $\lambda=1$. Thus, consistent with the results of Refs. 4 and 5, we find a spontaneously dimerized columnar phase in the Hamiltonian which has the full symmetry of the square lattice ($\lambda=1$).

B. $\mathcal{D}=\mathcal{D}_s, J_3=0$

The results of the calculation on $H_{\mathcal{D}}$ with $\mathcal{D}=\mathcal{D}_s$, the staggered dimerization are shown in Fig. 4. Unlike Fig. 3, however, wave functions with both choices of the dimerization $\mathcal{D}'=\mathcal{D}_s$ and $\mathcal{D}'=\mathcal{D}_c$ were found to lead to stable magnetically disordered phases. For small λ the system is in a magnetically disordered staggered ($\mathcal{D}'=\mathcal{D}_s$) phase and eventually undergoes second-order transitions to magnetically ordered phases at wave vectors (π,π) and $(\pi,0)$. At still larger values of λ, the system undergoes a first-order transition to phases based upon columnar ($\mathcal{D}'=\mathcal{D}_c$) dimerization. The columnar dimerization is present without any magnetic order in the range $0.19<J_2/J_1<0.66$; this entire region therefore breaks a symmetry of the underlying Hamiltonian which is based upon $\mathcal{D}=\mathcal{D}_s$. The columnar dimerization also persists in the magnetically ordered phases, again breaking additional lattice symmetries.

We note finally that at $\lambda=1$, $H_{\mathcal{D}}$ is independent of $\mathcal{D}$. The phases along the line $\lambda=1$ in Figs. 3 and 4 are therefore identical and represent the best variational ground states of the spin-$\frac{1}{2}$ Heisenberg antiferromagnet with nearest-neighbor and second-neighbor interactions, which have the full symmetry of the square lattice.

C. $\mathcal{D}=\mathcal{D}_c, J_2=0$

Finally, we consider the effect of turning on a third-neighbor coupling J_3. We will restrict our analysis to the case $J_2=0$ and Hamiltonian $H_{\mathcal{D}}$ with $\mathcal{D}=\mathcal{D}_c$. Moreover, because of the presence of incommensurate wave vectors in the magnetically ordered phase, we will restrict our analysis to the magnetically disordered phase and determine its boundary of instability to the appearance of magnetic order. The results of the calculation are shown in Fig. 5. Wave functions with columnar dimerization ($\mathcal{D}'=\mathcal{D}_c$) and no magnetic order were found to be stable for $0.125<J_3/J_1<0.843$ at $\lambda=1$; in particular, they were lower in energy than states with staggered dimerization ($\mathcal{D}'=\mathcal{D}_s$). This conclusion presumably holds at all values of $\lambda<1$, as decreasing λ stabilizes the columnar ordering. The magnetically disordered columnar phase was found to have four different second-order phase boundaries to magnetically ordered phases with the following wave vectors: (i) (π,π) for $\lambda>0$ and small J_3; (ii) $(\pi/2,\pi-\cos^{-1}(J_1/4J_3))$ for $\lambda>0$ and J_3/J_1 near 1 (note that unlike other magnetically ordered phases the wave vector is a continuous function of the exchange constants); (iii) $(\pi/2,0)$ for $\lambda<0$ and small J_3; and (iv) $(\pi,0)$ for $\lambda<0$ and J_3/J_1 near 1. All of the magnetically ordered phases and their ordering wave vectors are consistent with the results of Gelfand *et al.*[5] We note that ordering wave vector has been determined only at the *boundary* between the ordered and disordered phases. It is possible that the wave vector will change in the interior of the magnetically ordered phases.

V. CONCLUSIONS

We have introduced a new bond-operator representation of quantum spin-$\frac{1}{2}$ operators. The representation is specifically designed to investigate the properties spontaneously or intrinsically "dimerized" magnetically disordered phases. A mean-field theory based upon this representation yields a low-lying spectrum of spin-1 bosons. At a critical value of the exchange constants the spin-1 bosons condense at a wave vector $\mathbf{k}_0$, leading to magnetic ordering at $\mathbf{k}=\mathbf{k}_0+(\pi,\pi)$.

The bond-boson representation was used to determine a mean-field phase diagram of the frustrated Heisenberg antiferromagnets on a square lattice of the type considered recently by Gelfand *et al.*[5] These are frustrated antiferromagnets characterized by a dimerization $\mathcal{D}$ and a parameter λ; at $\lambda=0$, the ground state consists of decoupled singlets on $\mathcal{D}$, while $\lambda=1$ restores the square-lattice symmetry. It is useful to distinguish the cases $\lambda=1$ and $\lambda<1$ in the subsequent discussion.

$(a)\lambda=1$. The mean-field theory displayed a region of values of J_2/J_1 and J_3/J_1 at $\lambda=1$ where a magnetically disordered, spontaneously dimerized columnar phase is the ground state. No other magnetically disordered phase was found to be stable at $\lambda=1$. Thus our results suggest that the magnetically disordered phase of certain frustrated spin-$\frac{1}{2}$ antiferromagnets whose exchange constants have the full symmetry of the square lattice, is spontaneously dimerized in a columnar pattern. This result is consistent with the series expansions of Gelfand *et al.*,[5] the numerical diagonalization of Dagotto and Moreo[6] and the large-N results of Read and Sachdev.[4] Magnetically ordered states were also found at $\lambda=1$, but always with coexisting dimerizations. In contrast, the large-N calculations[4] do not find coexisting magnetic and dimerization order. The calculations of this paper clearly overemphasize the stability of the dimerization order, and cannot be used to conclude that magnetic and dimerization order can coexist. It remains an open question as to whether there is a fundamental reason forbidding the coexistence of magnetic and dimerization order.

$(a)\lambda<1$. For $\lambda<1$ the models were found to display a large variety of magnetically ordered phases as shown in Figs. 3–5. All of these phases and their ordering wave vectors are consistent with the results of Gelfand *et al.*[5]

A promising line for future study is the application of this method to the disordered antiferromagnets considered by Bhatt and Lee.[10] The physical picture developed by them clearly indicates that a description in terms of singlet bond operators is natural. The bond bosons can be placed on the bonds with the largest exchange constants. Hartree-Fock decoupling of the Hamiltonian will lead to an effective Hamiltonian of the form

$$H_r=\sum_{ij}(2A_{ij}t^\dagger_{i\alpha}t_{j\alpha}+B_{ij}t_{i\alpha}t_{j\alpha}+B^*_{ij}t^\dagger_{i\alpha}t^\dagger_{j\alpha})\ , \qquad (4.1)$$

where A_{ij} and B_{ij} depend upon the random exchange constants. The localization properties of the bosonic excitations of H_r are not immediately apparent and are presently being investigated. This method offers, in principle, a systematic way of investigating the thermodynamic and transport properties of random antiferromagnets.

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[12] It is possible to consider $H_{\mathcal{D}_c}$ and $H_{\mathcal{D}_s}$ as special cases of a single Hamiltonian:

$$
\begin{aligned}
H = J_1 \sum_{\langle ij\rangle \in \mathcal{D}_c \cap \mathcal{D}_s} \mathbf{S}_i \cdot \mathbf{S}_j + \lambda_1 J_1 \sum_{\langle ij\rangle \in \mathcal{D}_c - \mathcal{D}_c \cap \mathcal{D}_s} \mathbf{S}_i \cdot \mathbf{S}_j \\
+ \lambda_2 J_1 \sum_{\langle ij\rangle \in \mathcal{D}_s - \mathcal{D}_c \cap \mathcal{D}_s} \mathbf{S}_i \cdot \mathbf{S}_j + \lambda_1 \lambda_2 J_1 \sum_{\langle ij\rangle \notin \mathcal{D}_c \cup \mathcal{D}_s} \mathbf{S}_i \cdot \mathbf{S}_j \\
+ \lambda_1 \lambda_2 J_2 \sum_{2\mathrm{NN}} \mathbf{S}_i \cdot \mathbf{S}_j + \lambda_1 \lambda_2 J_3 \sum_{3\mathrm{NN}} \mathbf{S}_i \cdot \mathbf{S}_j \ ,
\end{aligned}
$$

where $H_{\mathcal{D}_c}$ corresponds to $\lambda_1 = 1$, $\lambda_2 = \lambda$, and $H_{\mathcal{D}_s}$ to $\lambda_1 = \lambda$, $\lambda_2 = 1$. Other values of $0 < \lambda_1$, $\lambda_2 < 1$ can be used to interpolate between $H_{\mathcal{D}_c}$ and $H_{\mathcal{D}_s}$. For the case $J_3 = 0$, the phases of H are determined by the three parameters λ_1, λ_2, and J_2/J_1. The phase diagrams obtained in this paper are two separate two-dimensional cuts through this three-dimensional phase space.