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Nonlinear effects in the spin-liquid phase

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A *spin-liquid* phase in which long-range order at the absolute zero is destroyed by quantum fluctuations may occur in two-dimensional frustrated Heisenberg antiferromagnets. However, the possible existence of the spin liquid is intriguing, because a first-order phase transition is expected to suppress it. Here we present a nonperturbative approach to explore quantum fluctuations in the whole parameter space of the square Heisenberg model with competing interactions up to third-nearest neighbors. Our results rule out the existence of the spin liquid when the exchange-interaction competition allows only collinear spin configurations, but we find that spin liquid exists when the exchange interactions also produce helix configurations.

I. INTRODUCTION

A spin liquid (SL) is an exotic phase revealed as a divergent spin reduction[1,2] within the simple spin-wave (SSW) approximation in the square Heisenberg antiferromagnet with a suitable competing next-nearest-neighbor (NNN) interaction. We recall that the classical ($S \rightarrow \infty$) zero-temperature phase diagram of the square lattice with a nearest-neighbor (NN) antiferromagnetic interaction ($J_1 < 0$) and NNN interaction (J_2) consists[1] of the two-sublattice Néel (N) configuration for $j_2 = J_2/J_1 < \frac{1}{2}$ and of the two-interpenetrating-antiferromagnetic-sublattice configuration for $j_2 > \frac{1}{2}$, the angle θ between the spin directions of the two antiferromagnetic sublattices being arbitrary. For $\theta = 0$ ($\theta = \pi$), one has the so-called "columnar" (C) phase with alternating columns (alternating rows) of parallel spins. Quantum fluctuations, when accounted for in the SSW approximation,[1,2] suggest the existence of the spin-liquid phase via a divergent spin reduction. Consequently, the occurrence of this phase in a finite range around $j_2 = \frac{1}{2}$ was proposed. The existence of magnetically disordered phases around $j_2 = \frac{1}{2}$ was also proposed on the basis of series expansions about explicitly dimerized models[3] as well as on the basis of finite-cluster diagonalization.[4] On the other hand, this expectation runs counter to results of direct considerations on the zero-point-motion energy which suggest that the N phase should expand at the expense of the C phase. Notice that the C phase is selected by quantum fluctuations which favor the maximum of antiparallelism in the spin patterns, so that the arbitrariness of the angle θ between the two interpenetrating antiferromagnetic sublattices is removed.[1] The zero-point-motion energy of the N configuration is lesser than the zero-point-motion energy of the C configuration so that a first-order phase transition is expected to prevent the occurrence of the SL phase. This picture was indeed substantiated[5,6] by a self-consistent treatment of the leading zero-point-motion contributions to the N and C ground-state energies. Notice that any divergence of the spin reduction disappears in such approaches.

However, the SL phase could survive nonlinear effects in a wider parameter space. For instance, the addition of a third-nearest-neighbor (TNN) interaction J_3 ($3N$ model) enters two helix phases we call H_1 and H_2 in the classical phase diagram.[3,7] The N-H_1-C triple point corresponds to $j_2 = \frac{1}{2}$, $j_3 = J_3/J_1 = 0$. The SSW approach suggests the existence of the spin liquid in the neighborhood of this point as well as in strips encompassing the N-H and H_1-H_2 phase boundaries as shown in Fig. 4 of Ref. 8. The existence of a spin liquid is also expected on the H_1-C phase boundary, but only for $S = \frac{1}{2}$.[9] A controlled perturbation approach[10] shows that crucial nonlinear contributions shift the existence region of the SL phase well inside the classical existence region of the H_1 and H_2 phases. Indeed, the spin reduction as obtained by SSW theory diverges on the classical N-H phase boundary because of a soft k^2 behavior of the magnon dispersion curve in the long-wavelength limit, but we have found that the Hartree-Fock contribution restores the customary linear k dependence.[10] However, the vanishing of the magnon velocity is reached inside the H_1 and H_2 existence regions so that SL can exist unless a first-order N-H phase transition occurs. Unfortunately, a systematic perturbation approach cannot be performed for vanishing j_3 because of unphysical divergencies that appear around the N-H_1-C triple point. This drawback is due to soft lines in the SSW dispersion curve: The perturbation contributions are enhanced in a catastrophic way because of lines of zeros appearing in the denominators of the perturbation expansion terms. Indeed, in the neighborhood of the N-H_1-C triple point, a suitable approach is the self-consistent approach, in which artifacts that enter by the classical approximation can be avoided in a natural way.

Note that the features of the phase diagram of the $3N$ model were anticipated on the basis of a reasonable guess.[11] Here we try to obtain *quantitative* results by performing an extension of the self-consistent approach so far restricted to $j_3 = 0$,[5,6] and to study the effect of quantum fluctuations on helical configurations with $j_3 \neq 0$. This extension is not trivial and requires an ansatz concerning the magnon spectrum that we justify on the basis of the Goldstone theorem.[12] The results we obtain confirm the main features of the phase diagram that were suggested in Ref. 11, in particular, the first-order N-C phase transition for j_3 small enough, but we find an interesting discrepancy concerning the $S=\frac{1}{2}$ case. Indeed, we find that the SL phase for $S=\frac{1}{2}$ gets rid of the H_1 configuration for intermediate values of j_3, while for $S=1$ we find that the SL and H_1 configurations appear simultaneously as j_3 increases, in agreement with Ref. 11, even though the region of existence of the SL phase is substantially reduced.

II. MAGNON SPECTRUM AND QUANTUM FLUCTUATIONS IN THE 3N MODEL

The Hamiltonian we consider reads

$$\mathcal{H}=-2J_1\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j-2J_2\sum_{\langle il\rangle}\mathbf{S}_i\cdot\mathbf{S}_l-2J_3\sum_{\langle im\rangle}\mathbf{S}_i\cdot\mathbf{S}_m\ , \quad (1)$$

where J_1, J_2, J_3 are the NN, NNN, TNN exchange coupling, respectively. The NN coupling J_1 is antiferromagnetic ($J_1<0$), while J_2 and J_3 can have either signs $\langle ij\rangle$, $\langle il\rangle$, and $\langle im\rangle$ mean distinct NN, NNN, and TNN pairs of spin, respectively. We refer the reader to Refs. 12 and 13 to get the explicit expression of the bosonic equivalent Hamiltonian obtained by the Holstein-Primakoff transformation[14] when both collinear and helix configurations are taken into account.

We are interested in the zero-temperature phase diagram for which the classical approximation[3,7] suggests the existence of four phases: N, C, and the two helical configurations H_1 and H_2. The $\mathbf{Q}$ wave vectors characterizing the N, C, and H_2 phases are

$$(\pi,\pi)\ ,$$

$$(0,\pi)\ ,$$

$$\left[\cos^{-1}\frac{2j_2-1}{4j_3},\pi\right]\ ,$$

and

$$\left[\cos^{-1}\frac{-1}{2j_2+4j_3},\cos^{-1}\frac{-1}{2j_2+4j_3}\right]\ ,$$

respectively. Long-range order (LRO) is expected on the basis of SSW theory except for a strip encompassing the N-H (Refs. 8 and 9) and H_1-H_2 (Refs. 2 and 8) boundaries for any finite S, while LRO might be suppressed along the H_1-C boundary for $S=\frac{1}{2}$ only.[9]

Higher-order quantum corrections cannot be treated in a systematic perturbation approach throughout parameter space, even though significant results have been obtained on the persistence of the Goldstone mode[12,15] for $\mathbf{k}=\pm\mathbf{Q}$, where $\mathbf{Q}$ is the helix wave vector, and on the lifting of the accidental soft modes[15] for $\mathbf{k}=\mathbf{Q}'$, where $\mathbf{Q}'$ is obtained from $\mathbf{Q}$ by symmetry transformations of the underlying square lattice. However, the perturbation approach suffers from certain shortcomings. For instance, no thermal and quantum renormalization of the helix wave vector can be obtained since the SSW energy spectrum is well defined only for $\mathbf{Q}=\mathbf{Q}_c$, where $\mathbf{Q}_c$ is the helix wave vector obtained in classical approximation ($S\to\infty$). Variational approaches have been worked out,[13] but the equations involved are too cumbersome for systematic use in parameter space, unless a suitable ansatz can be introduced to simplify the formulas. Here we propose a description of the modulated phases that agrees with the variational approach for large S (Ref. 13) and reduces to the self-consistent approach for the collinear phases, where the bilinear bosonic Hamiltonian contributions arising from the normal ordering of the quartic potential are fully accounted for by the generalized Bogoliubov transformation[16] (GBT). For $j_3=0$, our approach agrees with recent calculations based on a self-consistent Hartree-Fock approximation[5,6] and on the Schwinger-boson mean-field theory (SBMFT).[17]

Notice that the magnon spectrum $\hbar\omega_{\mathbf{k}}$ that we propose for helix configurations shows Goldstone modes at $\mathbf{k}=\pm\mathbf{Q}$ and quantum gaps[15] at $\mathbf{k}=\pm\mathbf{Q}'$ so that the main quantum corrections are embodied in an effective way:

$$\hbar\omega_{\mathbf{k}}=\sqrt{S_{\mathbf{k}}D_{\mathbf{k}}}\ , \quad (2)$$

where

$$S_{\mathbf{k}}=J(\mathbf{Q})-J(\mathbf{k})\ , \quad (3)$$

$$D_{\mathbf{k}}=J(\mathbf{Q})-\tfrac{1}{2}[J(\mathbf{k}+\mathbf{Q})+J(\mathbf{k}-\mathbf{Q})]\ , \quad (4)$$

with

$$J(\mathbf{k})=4J_1(S+\tfrac{1}{2}-a_{1x})\cos k_x+4J_1(S+\tfrac{1}{2}-a_{1y})\cos k_y+4J_2(S+\tfrac{1}{2}-a_2^{+})\cos(k_x+k_y)+4J_2(S+\tfrac{1}{2}-a_2^{-})\cos(k_x-k_y)$$
$$+4J_3(S+\tfrac{1}{2}-a_{3x})\cos(2k_x)+4J_3(S+\tfrac{1}{2}-a_{3y})\cos(2k_y)\ . \quad (5)$$

Here

$$a_{1x}=\frac{1}{2N}\sum_{\mathbf{q}}\sqrt{S_{\mathbf{q}}/D_{\mathbf{q}}}(1-\cos q_x)\ , \quad (6)$$

$$a_{1y}=\frac{1}{2N}\sum_{\mathbf{q}}\sqrt{S_{\mathbf{q}}/D_{\mathbf{q}}}(1-\cos q_y)\ , \quad (7)$$

$$a_2^{+}=\frac{1}{2N}\sum_{\mathbf{q}}\sqrt{S_{\mathbf{q}}/D_{\mathbf{q}}}[1-\cos(q_x+q_y)]\ , \quad (8)$$

$$a_2^{-}=\frac{1}{2N}\sum_{\mathbf{q}}\sqrt{S_{\mathbf{q}}/D_{\mathbf{q}}}[1-\cos(q_x-q_y)]\ , \quad (9)$$

$$a_{3x}=\frac{1}{2N}\sum_{\mathbf{q}}\sqrt{S_{\mathbf{q}}/D_{\mathbf{q}}}[1-\cos(2q_x)]\ , \tag{10}$$

$$a_{3y}=\frac{1}{2N}\sum_{\mathbf{q}}\sqrt{S_{\mathbf{q}}/D_{\mathbf{q}}}[1-\cos(2q_y)]\ . \tag{11}$$

The value of $\mathbf{Q}$ is obtained by maximization of $J(\mathbf{Q})$, which ensures the magnon spectrum energy to be real and positive. The N phase corresponds to $Q_x=\pi, Q_y=\pi$ for which one finds $a_{1x}=a_{1y}=a_1$, $a_2^+=a_2^-=a_2$, $a_{3x}=a_{3y}=a_3$. The C phase corresponds to $Q_x=0, Q_y=\pi$ for which one finds $a_{1x}\neq a_{1y}$, $a_2^+=a_2^-=a_2$, $a_{3x}\neq a_{3y}$. The H_1 phase corresponds to

$$Q_x=\cos^{-1}\frac{-(S+\frac{1}{2}-a_{1x})+2j_2(S+\frac{1}{2}-a_2)}{4j_3(S+\frac{1}{2}-a_{3x})}\ ,$$

$$Q_y=\pi\ ,$$

where $a_{1x}\neq a_{1y}$, $a_2^+=a_2^-=a_2$, $a_{3x}\neq a_{3y}$. The H_2 phase corresponds to

$$Q_x=\cos^{-1}\frac{-(S+\frac{1}{2}-a_1)}{2j_2(S+\frac{1}{2}-a_2^+)+4j_3(S+\frac{1}{2}-a_3)}\ ,$$

$$Q_y=Q_x$$

with $a_{1x}=a_{1y}=a_1$, $a_2^+\neq a_2^-$, $a_{3x}=a_{3y}=a_3$. The above relationships between the self-consistent coefficients a_{1x}, a_{1y}, a_2^+, a_2^-, a_{3x}, a_{3y} for the different phases are suggested by symmetry reasons and tested by direct numerical calculation. Notice that the relationship between the wave vector $\mathbf{Q}$ and the exchange integrals can be written in the same way as in a linear approximation if one defines renormalized exchange integrals. Indeed one has, for the H_1 configuration,

$$\cos Q_x=\frac{2\tilde{j}_2-1}{4\tilde{j}_3}\ ,\quad Q_y=\pi \tag{12}$$

with

$$\tilde{j}_2=j_2\frac{S+\frac{1}{2}-a_2}{S+\frac{1}{2}-a_{1x}},\quad \tilde{j}_3=j_3\frac{S+\frac{1}{2}-a_{3x}}{S+\frac{1}{2}-a_{1x}} \tag{13}$$

and, for the H_2 configuration,

$$\cos Q_x=-\frac{1}{2\tilde{j}_2+4\tilde{j}_3}\ ,\quad Q_y=Q_x \tag{14}$$

with

$$\tilde{j}_2=j_2\frac{S+\frac{1}{2}-a_2^+}{S+\frac{1}{2}-a_1},\quad \tilde{j}_3=j_3\frac{S+\frac{1}{2}-a_3}{S+\frac{1}{2}-a_1}\ . \tag{15}$$

Notice that these equations hold for the broken symmetry of the ground state that we have considered. For instance, if we had chosen for the H_1 configuration $\mathbf{Q}=(\pi,Q_y)$, all the subscripts x should be replaced by subscripts y in Eq. (13).

The reduced ground-state energy reads

$$\begin{aligned} e_0=E_0/4|J_1|N=\tfrac{1}{2}[\,&(S+\tfrac{1}{2}-a_{1x})^2\cos Q_x+(S+\tfrac{1}{2}-a_{1y})^2\cos Q_y+j_2(S+\tfrac{1}{2}-a_2^+)^2\cos(Q_x+Q_y)\\ &+j_2(S+\tfrac{1}{2}-a_2^-)^2\cos(Q_x-Q_y)+j_3(S+\tfrac{1}{2}-a_{3x})^2\cos(2Q_x)+(S+\tfrac{1}{2}-a_{3y})^2\cos(2Q_y)]\ , \end{aligned} \tag{16}$$

where E_0 is the ground state of our model obtained by the GBT approach. For the N, C, H_1, phases, where $Q_y=\pi$ and $a_2^+=a_2^-=a_2$, one has

$$e_0=\tfrac{1}{2}[(S+\tfrac{1}{2}-a_{1x})^2\cos Q_x-(S+\tfrac{1}{2}-a_{1y})^2]-j_2(S+\tfrac{1}{2}-a_2^2)\cos Q_x+\tfrac{1}{2}j_3[(S+\tfrac{1}{2}-a_{3x})^2\cos(2Q_x)+(S+\tfrac{1}{2}-a_{3y})^2]\ , \tag{17}$$

while for the H_2, N phases, where $Q_x=Q_y$, $a_{1x}=a_{1y}=a_1$, and $a_{3x}=a_{3y}=a_3$, one has

$$e_0=(S+\tfrac{1}{2}-a_1)^2\cos Q_x+\tfrac{1}{2}j_2[(S+\tfrac{1}{2}-a_2^+)^2\cos(2Q_x)+(S+\tfrac{1}{2}-a_2^-)^2]+j_3(S+\tfrac{1}{2}-a_3)^2\cos(2Q_x)\ . \tag{18}$$

The zero-temperature spontaneous magnetization $M(0)$ reads

$$M(0)=S+\frac{1}{2}-\frac{1}{2N}\sum_{\mathbf{q}}(\sqrt{S_{\mathbf{q}}/D_{\mathbf{q}}}+\sqrt{D_{\mathbf{q}}/S_{\mathbf{q}}})\ . \tag{19}$$

While the above equations are consistent with respect to the GBT approach[15,16] for the N and C configurations, the same approach provides unpleasant features[12,15,18–20] as for the magnon spectrum $\hbar\Omega_{\mathbf{k}}$ of helix configurations. Indeed, the result of the GBT approach is [18]

$$\hbar\Omega_{\mathbf{k}}=\sqrt{\tilde{S}_{\mathbf{k}}\tilde{D}_{\mathbf{k}}}\ , \tag{20}$$

where

$$\mathbf{S}_{\mathbf{k}}=\sum_{\delta}2J_\delta S\left\{\cos(\mathbf{Q}\cdot\boldsymbol{\delta})-\cos(\mathbf{k}\cdot\boldsymbol{\delta})+\frac{1}{2S}\{\cos(\mathbf{Q}\cdot\boldsymbol{\delta})(1-\tilde{I}_A-\tfrac{3}{2}\tilde{I}_B+\tilde{I}_\delta^+)-\cos(\mathbf{k}\cdot\boldsymbol{\delta})[1-\tilde{I}_A+\tfrac{1}{2}\tilde{I}_B+\tilde{I}_\delta^-\cos(\mathbf{Q}\cdot\boldsymbol{\delta})]\}\right\}\ , \tag{21}$$

$$\tilde{D}_{\mathbf{k}}=\sum_{\delta} 2J_{\delta}S\cos(\mathbf{Q}\cdot\boldsymbol{\delta})[1-\cos(\mathbf{k}\cdot\boldsymbol{\delta})]\left[1+\frac{1}{2S}(1-\tilde{I}_A-\tfrac{1}{2}\tilde{I}_B+\tilde{I}_{\delta}^{+})\right] \tag{22}$$

with

$$\tilde{I}_A=\frac{1}{2N}\sum_{\mathbf{q}}(\sqrt{\tilde{S}_{\mathbf{q}}/\tilde{D}_{\mathbf{q}}}+\sqrt{\tilde{D}_{\mathbf{q}}/\tilde{S}_{\mathbf{q}}})\ , \tag{23}$$

$$\tilde{I}_B=\frac{1}{2N}\sum_{\mathbf{q}}(\sqrt{\tilde{S}_{\mathbf{q}}/\tilde{D}_{\mathbf{q}}}-\sqrt{\tilde{D}_{\mathbf{q}}/\tilde{S}_{\mathbf{q}}})\ , \tag{24}$$

$$\tilde{I}_{\delta}^{+}=\frac{1}{N}\sum_{\mathbf{q}}\sqrt{\tilde{S}_{\mathbf{q}}/\tilde{D}_{\mathbf{q}}}\cos(\mathbf{q}\cdot\boldsymbol{\delta})\ , \tag{25}$$

$$\tilde{I}_{\delta}^{-}=\frac{1}{N}\sum_{\mathbf{q}}\sqrt{\tilde{D}_{\mathbf{q}}/\tilde{S}_{\mathbf{q}}}\cos(\mathbf{q}\cdot\boldsymbol{\delta})\ . \tag{26}$$

As one can verify, $\hbar\omega_{\mathbf{k}}$ given by Eq. (2) and $\hbar\Omega_{\mathbf{k}}$ given by Eq. (20) coincide for the N and C configurations, for which $\sum_{\mathbf{q}}\sqrt{\tilde{D}_{\mathbf{q}}/\tilde{S}_{\mathbf{q}}}=\sum_{\mathbf{q}}\sqrt{\tilde{S}_{\mathbf{q}}/\tilde{D}_{\mathbf{q}}}$, but for the H_1 and H_2 configurations $\hbar\Omega_{\mathbf{k}}$ suffers from spurious removal of the Goldstone modes at $\mathbf{k}=\pm\mathbf{Q}$. This drawback is due to neglect of the three-operator potential[12] of the bosonic equivalent Hamiltonian, whose second-order perturbation contribution is of the same order in $1/S$ as the first-order perturbation contribution of the quartic-operator potential (which is the only contribution accounted for by the GBT approach). On the other hand, the persistence of the Goldstone modes at $\mathbf{k}=\pm\mathbf{Q}$, when LRO is present, is ensured by symmetry reasons[12] and can be checked by numerical evaluation.[12,15] The magnon spectrum that we propose in Eq. (2) is the simplest modification of the spectrum (20) that can be made to satisfy the Goldstone theorem. Notice that the spectrum (2) allows the lifting of the accidental soft modes present in SSW theory [which corresponds to retention of the only terms proportional to S in (5)] and meets continuously the GBT spectrum for the N and C configurations. In Figs. 1 and 2 we show the magnon spectrum $\hbar\omega_{\mathbf{k}}$ given by Eq. (2) for the H_1 and H_2 configurations along the high-symmetry directions in reciprocal space. Notice the quantum gaps and Goldstone modes. We have compared the values of the quantum gap obtained from Eq. (2),

$$\hbar\omega_{\mathbf{Q}'}=\sqrt{D_{\mathbf{Q}'}S_{\mathbf{Q}'}}\ , \tag{27}$$

where

$$D_{\mathbf{Q}'}=4|J_1|\{-(S+\tfrac{1}{2}-a_{1x})\cos Q_x+(S+\tfrac{1}{2}-a_{1y})(1-\cos Q_x)+2j_2(S+\tfrac{1}{2}-a_2^{+})\cos Q_x(1+\cos Q_x)-j_3(S+\tfrac{1}{2}-a_{3y})[1-\cos(2Q_x)]\}\ , \tag{28}$$

$$S_{\mathbf{Q}'}=4|J_1|(1+\cos Q_x)[a_{1x}-a_{1y}-2j_3(a_{3x}-a_{3y})(1-\cos Q_x)] \tag{29}$$

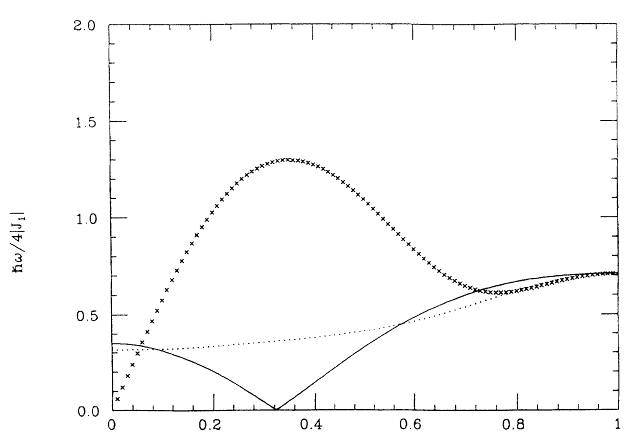

FIG. 1. Elementary excitation energies for the H_1 configuration ($j_2=0.6$, $j_3=0.1$) with $S=1$ as function of the reduced wave vector $\mathbf{q}=\mathbf{k}/\pi$ for $0<q_x<1$, $q_y=1$ (solid curve), for $q_x=1, 0<q_y<1$ (dots), and for $q_x=q_y$ (crosses). Notice the Goldstone modes at the zone center (crosses) and at the helix wave vector (solid curve). The accidental SSW soft mode is replaced by the quantum gap as shown by the dotted curve.

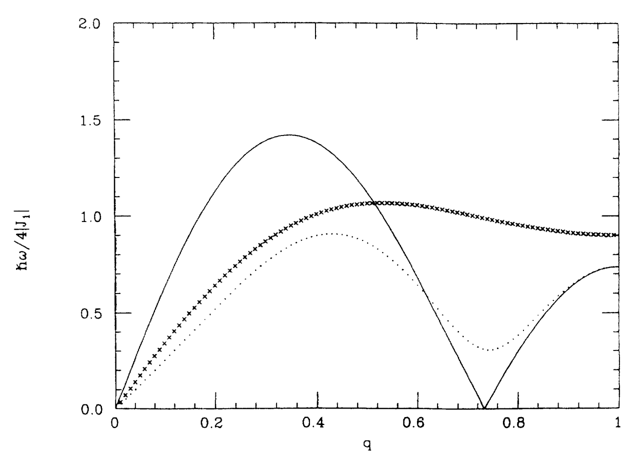

FIG. 2. Elementary excitation energies for the H_2 configuration ($j_2=0.4$, $j_3=0.25$) with $S=1$ as function of the reduced wave vector $\mathbf{q}=\mathbf{k}/\pi$ for $q_x=q_y$ (solid curve), for $q_x=-q_y$ (dots), and $0<q_x<1$, $q_y=0$ (crosses). Notice the Goldstone modes at the zone center and at the helix wave vector (solid curve). The accidental SSW soft mode is replaced by the quantum gap as shown by the dotted curve.

with that obtained by the perturbation approach at the leading order in $1/S$.[15] For $S=1$, $j_2=0.6$, and $j_3=0.1$, we obtain $\hbar\omega_{Q'}=4|J_1|\times 0.359$ from Eq. (27) to be compared with $\hbar\omega_{Q'}=4|J_1|\times 0.268$ as obtained in Ref. 15. Even more satisfactory is the agreement if one retains only terms proportional to S in (28). In this case one obtains $\hbar\omega_{Q'}=2|J_1|\times 0.307$. Notice that the leading contribution of $D_{Q'}$ is proportional to S, whereas $S_{Q'}$ is independent of S so that the SSW result corresponds to $S_{Q'}=0$. The helix wave vector we obtain from Eq. (12) is $\mathbf{Q}=(1.0198,\pi)$ to be compared with the classical value $\mathbf{Q}_c=(\pi/3,\pi)$. Notice that the perturbation approach prevents any renormalization of the helix wave vector $\mathbf{Q}_c$. This example shows that our effective spectrum (2) is in good quantitative agreement with that obtained from the perturbation theory;[15] besides, spectrum (2) accounts for quantum renormalization of the helix wave vector, which is essential to evaluate the effect of quantum fluctuations on the phase diagram.

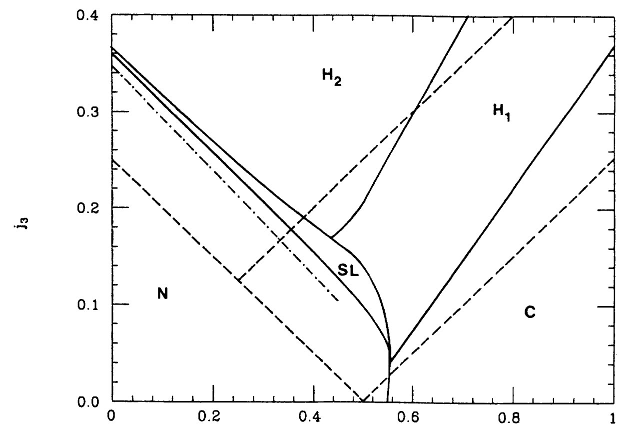

FIG. 4. The same as Fig. 3 but $S=1$. Notice the drastic shrinking of the region of existence of the SL with respect to the $S=\frac{1}{2}$ case.

III. ZERO-TEMPERATURE PHASE DIAGRAM OF THE SQUARE FRUSTRATED ANTIFERROMAGNET

We have numerically solved the self-consistent equations (6)–(11) for the N, C, H_1, and H_2 phases for assigned values of j_2, j_3, and S, then we have evaluated the ground-state energy and the spontaneous magnetization of those configurations in order to obtain the phase diagrams which are shown in Figs. 3 and 4 for $S=\frac{1}{2}$ and $S=1$, respectively.

The SL region is localized by the vanishing of the spontaneous magnetization $M(0)$. Where the disordered phase is concerned, one can prevent the magnetization from becoming negative by introducing a suitable bound to fix the magnon number.[21] Different features from both a qualitative and a quantitative point of view are found for $S=\frac{1}{2}$ and $S=1$. For $S=\frac{1}{2}$, an N-C-SL triple point and an SL-H_1-C triple point occur, while for $S=1$ an N-H_1-C triple point and an N-SL-H_1 triple point are noticed. In both cases an SL-H_1-H_2 triple point occurs. A relevant common feature is the N-C first order phase transition for j_3 small enough. This agrees with previous results[5,6,17] obtained for $j_3=0$ and with previous expectations[11] for $j_3\neq 0$ as for the $S=1$ case, but the strong S dependence shown in Figs. 3 and 4 is unexpected. A comment about the results obtained for $j_3=0$ is in order. Some numerical discrepancies between Refs. 5, 6, and 17 are related to the form of the ground-state energy. The expressions used in Refs. 5 and 6 are the same, apart from an expansion in $1/S$ used in Ref. 5. This approximation leads to ground-state energies for the N and C phases that do not cross for $S=\frac{1}{2}$,[5] since no N-C coexistence region is found. On the contrary, we obtain, in agreement with Ref. 6, that the N and C phases have the same ground-state energy at $j_2=0.6$ for $S=\frac{1}{2}$, and so we believe that a first-order N-C phase transition occurs. In Ref. 6, this possibility is ruled out and a disordered phase (DO) is suggested on the basis of an argument to which we do not subscribe. We stress that both Ref. 6 and our calculations find a crossing between the N and C ground-state energies so that only the phase of the lowest energy is stable and the other one is metastable. This does not mean that both phases are simultaneously present even if this could occur in nonequilibrium configurations.

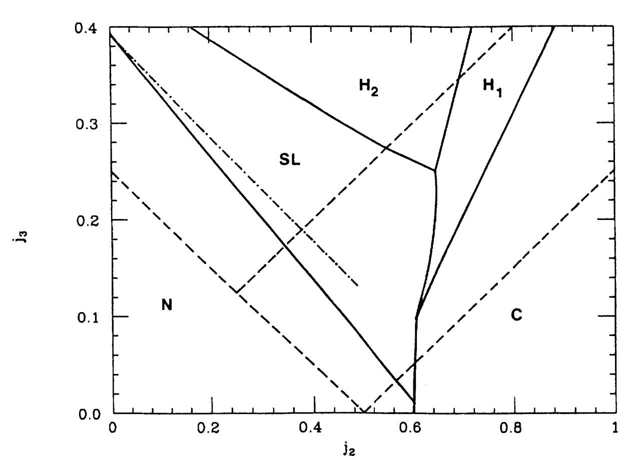

FIG. 3. Zero-temperature phase diagram for $S=\frac{1}{2}$ in the j_2-j_3 parameter space. The dashed curves show the SSW phase boundaries. The solid curves are the phase boundaries obtained in the present approximation. N, SL, C, H_1, and H_2 represent Néel, spin liquid, columnar, and two different helix configurations, respectively. The dash-dotted line is the N-SL phase boundary as obtained by the first-order perturbation theory.

In the Schwinger-boson mean-field theory (SBMFT) of Ref. 17, the drawback of the no crossing of the ground-state energies of the N and C phases for $S=\frac{1}{2}$ reappears. Moreover, the first-order phase transition for $S=1$ occurs at $j_2=0.65$, as compared with $j_2=0.55$ obtained by us and in Refs. 5 and 6. The SBMFT overestimates quantum fluctuations with respect to the self-consistent (or GBT) spin-wave theory. As for $j_3\neq 0$, our phase diagram (see Fig. 4) for $S=1$ agrees qualitatively with that suggested in Ref. 11 (see Fig. 1 of that paper). However, for $S=\frac{1}{2}$, we find a substantially wider region of existence for the SL configuration. Moreover, we find a substantial shrinking of the SL phase at large j_3 not suggested in Ref. 11. Anyway, the existence of the SL phase is confirmed when helical phases can exist owing to suitable exchange competition even if the SL phase is suppressed for $j_3=0$. A further nontrivial difference with respect to the scenario obtained by SSW theory[2,8,9] is the absence of SL near the H_1-H_2 phase boundary and near the H_1-C phase boundary even for $S=\frac{1}{2}$.

In Figs. 3 and 4 we have quoted (dash-dotted lines) the N-SL phase boundary as obtained by the first-order perturbation approach.[10] As one can see, the qualitative features of such a phase boundary agree with the present approach.

Notice that the renormalization-group approach[22] agrees with our conclusion that a disordered phase intervenes between the Néel and the helix configuration at finite j_3 even though it cannot be pushed at $j_3=0$. On the other hand, the renormalization-group approach cannot rule out the existence of a first-order N-H or N-C phase transition that would prevent the occurrence of the SL phase. A direct N-H phase transition is obtained for $S=\frac{1}{2}$ at any j_3, without the occurrence of a disordered phase in between, by extrapolating a large-N expansion to $N=1$.[23] In particular, for $j_3<0.005$ the first-order transition is between the N and C phases, in agreement with Fig. 3. The absence of a SL phase for any $j_3\neq 0$ is not clear.[11] Perhaps the extrapolation to $N=1$, which has to be performed in order to restore the physical picture, makes this result less reliable than the result obtained by the renormalization group[22] and by the self-consistent spin-wave theory[5,6,11] which do not suffer from such unreliable extrapolations.

Finally we comment on the results for $S=\frac{1}{2}$ obtained from a series expansion about explicitly dimerized models[3] and from a finite-cluster diagonalization.[4,8] The former approach consists of a perturbative expansion starting from a set of unperturbed Hamiltonians where three of four NN interactions are neglected, while NNN and TNN interactions are neglected completely, so that the results obtained for the $3N$ model might be questionable. Indeed these results depend crucially on the choice of the dimer covering of the lattice which, in its turn, determines the unperturbed Hamiltonian (see, for instance, Figs. 3 and 4 of the first reference and Fig. 2 of the second reference quoted in Ref. 3), and the ratio method used by the author to locate the transition to an ordered phase (N or C for $j_3=0$) gives results affected by larger and larger error bars as one approaches the range $0.3<j_2<0.6$ where the occurrence of the disordered phase is suggested. For $j_2=0$, the N, disordered, and helix configurations are found,[3] while our results suggest the N, SL, and H_2 phases, where the disordered (SL) phase exists over a range less than that of the disordered (dimerized) configuration. We note that our ground-state energy lies below the ground-state energy of the columnar dimerized configuration in the range $0.3<j_3<0.5$, while the energy of the columnar dimerized phase is very close to our evaluation of the H_2 energy in the range $0.5<j_3<0.6$.

As for the approach consisting of a diagonalization of finite clusters of 16 and 20 sites using a Lanczos technique,[4,8] a clear conclusion about the existence of LRO cannot be achieved because extrapolation of the results for a finite lattice to the bulk limit has not been done. However, a resonating-valence-bond (disordered) state may be ruled out at least at $j_3=0$ and the stability of the columnar dimerized state cannot be shown convincingly.[4] In a subsequent paper,[8] finite-cluster diagonalization has been used to study the $3N$ model along the line $j_3=j_2/2$, $j_2>\frac{1}{4}$. The authors conclude that in the bulk limit the LRO should vanish in a region $0.25<j_2<0.5$ (we find the SL phase for $0.35<j_2<0.55$ along the line $j_3=j_2/2$ as shown in Fig. 3). Even though the existence of LRO in the thermodynamic limit remains an open question, the static structure factor $S(\mathbf{q})$ (see Fig. 8 of Ref. 8) shows peaks located in points that give spin configurations in overall agreement with those shown in Fig. 3.

IV. SUMMARY AND CONCLUSIONS

One of the more interesting features of frustrated Heisenberg antiferromagnets is the possible existence of the spin-liquid (SL) phase.[1,2] The occurrence of this exotic phase was suggested on the basis of simple spin-wave (SSW) theory for the square Heisenberg antiferromagnet with next-nearest-neighbor (NNN) competitive interaction.[1] However, higher-order quantum fluctuations[5,6] suppress the SL for this model entering a first-order phase transition between the Néel (N) and the columnar (C) configurations. On the other hand, the survival of the SL was guessed[11] in a wider parameter space. In this paper, we work out a self-consistent approach based on the generalized Bogoliubov transformation (GBT)[16,12] for the square Heisenberg antiferromagnet with competing interactions up to third-nearest neighbors (TNN). The zero-temperature phase diagram of this model obtained in the classical approximation is shown in Refs. 3 and 7. Our self-consistent calculation agrees with the previous ones[5,6] for $j_3=0$.

To describe the helix configurations we do not limit ourselves to the magnon spectrum obtained by the self-consistent approach because of the well-known unsatisfactory features obtained by this and analogous approaches.[18,19,12,20] This spectrum indeed violates the Goldstone theorem[12] since it does not show Goldstone modes at $\mathbf{k}=\pm\mathbf{Q}$. The origin of this drawback is well understood[20] and it is due to neglect of the three-operator magnon interaction, the inclusion of which unfortunately leads to results[12,13] that are too cumbersome to be used for detailed analysis in calculating the rich phase diagram of our model. So we do the ansatz of Eq. (2) that embodies all known features of the magnon spectrum due to quantum fluctuations, namely, the persistence of Goldstone modes at the zone center and at the helix wave vector $\mathbf{k}=\pm\mathbf{Q}$ and quantum gaps replacing the accidental SSW soft modes at $\mathbf{k}=\mathbf{Q}'$, where $\mathbf{Q}'$ is in the "star" of the wave vector **Q**. Notice that the spectrum (2) allows quantum renormalization of the helix wave vector that cannot be achieved by any perturbation approach.[12]

The phase diagrams we obtain for $S=\frac{1}{2}$ and $S=1$ are shown in Figs. 3 and 4. We confirm the existence of the SL phase when the exchange competition supports helix configurations in addition to the collinear ones. Our results confirm the guess of Ref. 11 for $S=1$ but we find a different scenario for $S=\frac{1}{2}$. The evidence of the S dependence of the phase diagram is clearly visible in Figs. 3 and 4. These features indicate that the region of existence of the SL is severely reduced with increasing S.

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