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Dimer stability region in a frustrated quantum Heisenberg antiferromagnet

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We study the stability region for the columnar dimer state proposed as a candidate ground state for the square-lattice quantum antiferromagnet with first- and second-neighbor antiferromagnetic couplings (J_1-J_2 model). We use a boson representation of the spin operators suited to the perturbative expansion around a dimer ground state. At lowest order, the columnar dimer is found to be stable only at the classical critical value $J_2/J_1=\frac{1}{2}$. However, we show that the leading anharmonic corrections stabilize the dimerized phase in a region of a finite width around $J_2/J_1=\frac{1}{2}$. A comparison of the ground-state energies shows that among the possible dimerized states the columnar dimer is the most favorable candidate to separate the two ordered states in the $S=\frac{1}{2}$ antiferromagnet with first- and second-neighbor exchange.

The discovery of the remarkable magnetic properties of high-T_c superconductors has led to an intense activity in the subject of antiferromagnetic quantum spin systems mainly in two dimensions. One of the crucial issues is the nature of the ground state. Magnetically disordered resonating-valence-bond (RVB) states have been proposed as possible candidates for the ground states of the Heisenberg antiferromagnets (AFM) in two dimensions.[1] However, the spin-$\frac{1}{2}$ Heisenberg AFM with nearest-neighbor exchange have been studied by several techniques and there is now convincing evidence that they have ordered ground states on both square and triangular lattices.[2] The situation is far from being understood in the case of frustrated spin systems. The addition of next-to-nearest-neighbor antiferromagnetic couplings certainly destabilizes Néel order on a square lattice and it has been suggested that beyond some critical values of the couplings the Néel order is replaced by a new magnetically disordered phase.[3] There are presently several candidates for this phase. Large-N techniques[4,5] suggest that spin-Peierls order can occur. This idea has received some support from numerical[6] and series[7] studies on a $S=\frac{1}{2}$ model with first- and second-neighbor couplings (the so-called J_1-J_2 model). Lanczös studies[6] have measured a susceptibility suited to dimerized states showing an enhancement near $J_2/J_1=\frac{1}{2}$. However, recent results[8] on a 36-site cluster indicate that this quantity does not scale properly with the system size. More exotic proposals also have been made, such as chiral[9] or spin nematic[10] ground states for antiferromagnets with strong enough frustration.

On the contrary, perturbative calculations for J_1-J_2 model within the standard $1/S$ expansion do not reveal any intermediate phase. One finds a first-order transition between two ordered phases with ordering wave vector (π,π) (Néel phase) and $(\pi,0)$ [or $(0,\pi)$], respectively.[11-15] While this is certainly correct in the large-S limit, the $S=\frac{1}{2}$ case can be quite different as suggested by the numerical results. It is also worth mentioning that the perturbative approach based on the Schwinger bosons representation shows[14,15] that $S=\frac{1}{2}$ is very close to the critical value of S which separates a first-order transition between two ordered states from a two-step transition via an intermediate magnetically disordered phase, which is most likely to be dimerized.

In this paper we present the results of a study of the stability regions of various dimerized states of the spin-$\frac{1}{2}$ square lattice antiferromagnet with nearest- and next-to-nearest neighbors. We have used a bosonization technique suited to the perturbative expansion around dimer states and have shown that zero-point fluctuations favor energetically the columnar dimer in Fig. 1(a). At the quadratic order in bosons, this configuration is stable only for the particular ratio of the couplings $J_2/J_1=\frac{1}{2}$. However, when the interactions between bosonic excitations are treated in a self-consistent manner, the columnar dimer becomes stable in a *finite* region around $J_2/J_1=\frac{1}{2}$. This means that this state is likely to separate two ordered states in $S=\frac{1}{2}$ AFM with first- and second-neighbor exchange, though the question of whether columnar dimer actually is a global minima in the energy is still unanswered.

We shall study the J_1-J_2 model defined by the Hamiltonian

$$H=J_1\sum_{NN}\mathbf{S}_i\cdot\mathbf{S}_j+J_2\sum_{NNN}\mathbf{S}_i\cdot\mathbf{S}_k\,. \tag{1}$$

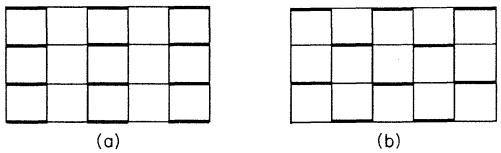

FIG. 1. Two dimer coverings of the square lattice: (a) columnar dimer and (b) staggered dimer.

The first sum runs over nearest-neighbor links of a square lattice and the second over next-to-nearest-neighbor links. The $\mathbf{S}_i$ are spin-$\frac{1}{2}$ operators and the couplings J_1 and J_2 are both positive, i.e., antiferromagnetic. It is convenient to introduce $\alpha = J_2/J_1$ which is the unique dimensionless parameter of this model. Let us first discuss the properties of the classical model related to (1) when the spin value $S \to \infty$. For values of α smaller than $\frac{1}{2}$ the classical ground state is Néel ordered, i.e., the ordering wave vector is $\mathbf{Q} = (\pi, \pi)$. For $\alpha > \frac{1}{2}$ there is a continuous degeneracy at the classical level which is lifted by quantum fluctuations, the so-called "order from disorder" phenomenon.[16] The ordering selected by quantum fluctuations is defined by the wave vector $\mathbf{Q} = (0, \pi)$ or $(\pi, 0)$. It is then possible to study the model (1) by a perturbative expansion in powers of $1/S$ starting from the classical ground states: this is the standard spin-wave theory *à la* Holstein-Primakoff. In the classical description, there is no hysteresis at the transition between the (π, π) and $(\pi, 0)$ [or $(0, \pi)$] phases. At the classical transition point $\alpha = \frac{1}{2}$ the dispersion relation for spin waves softens; entire lines of zero modes appear in the Brillouin zone. Since this is more than we expect from the Goldstone theorem, this degeneracy is accidental and should be lifted when the interactions between spin waves are taken into account. Simultaneously, quantum fluctuations should produce a finite hysteresis width for the $\alpha_c \approx \frac{1}{2}$ transition since there is no exact *quantum* degeneracy at criticality. This has been shown by explicit self-consistent calculations:[11–15] for finite S one finds a conventional first-order transition between the ordered phases for a critical value[11,15] $\alpha_c = \frac{1}{2} + O(1/S)$ or[12,14] $\alpha_c \simeq 0.6$ for spin $\frac{1}{2}$.

However, numerical[6] and series[7] analysis suggest that for $S = \frac{1}{2}$ the situation may be different and, in particular, there may be an intermediate dimerized phase. To study this possibility, we use a bosonization of the SU(2) spin algebra for spin-$\frac{1}{2}$ suited to the perturbative expansion around a spin-Peierls state. Specifically, if $\mathbf{S}_1$ and $\mathbf{S}_2$ are two neighboring spins to be involved in a dimer, then we introduce the ferromagnetic and antiferromagnetic vectors

$$\mathbf{M} = \mathbf{S}_1 + \mathbf{S}_2 \text{ and } \mathbf{L} = \mathbf{S}_1 - \mathbf{S}_2 . \tag{2}$$

The dimer (singlet) state is defined by $M = 0$ [$\mathbf{M}^2 = M(M+1)$]. We now introduce three bosons a, b, and c with canonical commutation relations in the following manner:[17]

$$\begin{aligned} M^z &= a^\dagger a - b^\dagger b , \\ M^+ &= \sqrt{2}\,(a^\dagger c - c^\dagger b) , \\ M^- &= \sqrt{2}\,(c^\dagger a - b^\dagger c) , \end{aligned} \tag{3a}$$

$$\begin{aligned} L^z &= -(c^\dagger U + U_c) , \\ L^+ &= \sqrt{2}\,(a^\dagger U + U b) , \\ L^- &= \sqrt{2}\,(b^\dagger U + U a) , \end{aligned} \tag{3b}$$

where $U = (1 - a^\dagger a - b^\dagger b - c^\dagger c)^{1/2}$. The physical subspace is obtained by considering states with at most one boson. The bosonic vacuum $a|0\rangle = b|0\rangle = c|0\rangle = 0$ is the singlet $M = 0$ state and the triplet states $M = 1$ are the states with one boson a, b, or c. In this physical subspace we have the correct commutation relations

$$\begin{aligned} [M_i, M_j] &= i\varepsilon_{ijk} M_k , \\ [L_i, L_j] &= i\varepsilon_{ijk} M_k , \\ [M_i, L_j] &= i\varepsilon_{ijk} L_k , \end{aligned} \tag{4}$$

together with the constraint $S^2 = \frac{3}{4}$. Moreover, the matrix elements of transitions between physical and nonphysical states are zero. This bosonization given by Eqs. (3a) and (3b) may thus be regarded as an exact at zero temperature. It is completely analogous to the Holstein-Primakoff expression for the spin operators in the case of $S = \frac{1}{2}$, the only difference is in the form of the vacuum state. The transformation [Eqs. (3a) and (3b)] can be generalized to spin S leading to $(2S+1)^2 - 1$ coupled bosons, allowing for the study of higher spin systems.[18]

If we consider any covering of the lattice with dimers, it is clear that it can be mapped into a Bose problem by introducing a, b, and c operators at each dimer "site." Since exchange also occurs between sites belonging to different dimers, the bosons are allowed to hop. The hopping induces a dispersion of the excitations as well as zero-point fluctuations which destroy the perfect dimer ordering.

We first focus on the columnar dimer ground state for the J_1-J_2 model [Fig. 1(a)]. The two spins belonging to a dimer are aligned along a lattice direction that will be referred to as "horizontal" (x). The exchange interaction (1) leads to couplings $\mathbf{M}_i \cdot \mathbf{M}_j$, $\mathbf{L}_i \cdot \mathbf{M}_j$, and $\mathbf{L}_i \cdot \mathbf{L}_j$, as well as on-site couplings. The last two terms contribute to the quadratic part of the bosonized Hamiltonian. After Fourier transformation the quadratic terms can be written as

$$\begin{aligned} H^{(2)} = \sum_k & A_k (a_k^\dagger a_k + b_k^\dagger b_k + c_k^\dagger c_k) \\ & + B_k [a_k b_{-k} + a_k^\dagger b_{-k}^\dagger + \tfrac{1}{2}(c_k c_{-k} + c_k^\dagger c_{-k}^\dagger)] , \end{aligned} \tag{5}$$

where

$$A_k = J_1(1 + v_{k_y}) - \frac{J_1}{2} v_{2k_x} - J_2 v_{k_y}(1 + v_{2k_x}) , \tag{6a}$$

$$B_k = J_1 v_{k_y} - \frac{J_1}{2} v_{2k_x} - J_2 v_{k_y}(1 + v_{2k_x}) , \tag{6b}$$

and $v_k = \cos k$. This quadratic form can be straightforwardly diagonalized by a Bogolyubov transformation leading to three degenerate bosons s, t, and u as, it should be, since all three excitations above the dimer singlet are completely equivalent.

We find however that the corresponding energies ε_k are real only at the special point $\alpha = \frac{1}{2}$. Thus there is at this order of approximation only one point of stability for the columnar dimer.[19] Exactly at $\alpha = \frac{1}{2}$ the energies of the quasiparticles are given by

$$\varepsilon_k = (A_k^2 - B_k^2)^{1/2} = J_1[(1 + \cos k_y)(1 - \cos 2k_x)]^{1/2} . \tag{7}$$

This dispersion relation has entire lines of zero modes in the Brillouin zone. The degeneracy is again not related to

any kind of broken symmetry and there is no reason to believe that $\alpha=\frac{1}{2}$ will be the critical point for the dimerization beyond the quadratic approximation. Anharmonic couplings should thus render the dimerized phase stable either in a finite region of α or nowhere.

To perform the perturbative calculations we need to expand the square root in the operator U. The simple approximation $\sqrt{1+x}\approx 1+\frac{1}{2}x$ is valid if both the density of particles and the interaction strength are small. There is, of course, no small parameter in the present problem to control our perturbative expansion. However, the low-density approximation can be avoided by summing all the quadratic terms in the Taylor expansion of U when we require normal ordering. This procedure is known from the Holstein-Primakoff expansion for $S=\frac{1}{2}$:

$$\left(1-\frac{a^\dagger a}{2S}\right)^{1/2}=1-a^\dagger a\left[1-\left(1-\frac{1}{2S}\right)^{1/2}\right]+O(a^\dagger a^\dagger aa)$$

(Ref. 20). In the present case a similar reasoning leads to

$$U\simeq U^2=1-a^\dagger a-b^\dagger b-c^\dagger c\,. \tag{8}$$

We believe that the first corrections will show the correct tendency since the degeneracy seen at the quadratic level is accidental and must be lifted immediately when quantum fluctuations are taken into account.[21] We will calculate the first correction in the coupling constant with the assumption, in analogy with the $1/S$ expansion, that quartic vertices are linear in the coupling while cubic vertices are proportional to the square root of the coupling constant.[21] Note also that with U given by Eq. (8) the matrix elements between physical and nonphysical states remain zero and nonphysical states thus do not contribute to the problem.

Expanding U as dictated by (8) we obtain vertices containing three and four bosons. At the first order in the coupling, we treat the quartic terms in a simple Hartree-Fock approximation: one inserts the zeroth-order contractions of the Bose operators allowed by (5) in the terms involving four bosons. This procedure changes the coefficients of the quadratic form (5): $A_k\rightarrow A_k+\delta A_k$ and $B_k\rightarrow B_k+\delta B_k$. Since $A_k-B_k=1$ we only need to compute $\delta A_k+\delta B_k$ to investigate what happens to the low-energy part of the dispersion relation ε_k. Our main result is that while the contributions coming from $\mathbf{M}_i\cdot\mathbf{M}_j$ terms are finite, the corrections produced by the $\mathbf{L}_i\cdot\mathbf{L}_j$ interaction are logarithmically divergent at $\alpha=\frac{1}{2}$. The corresponding contribution to the spectrum is *positive* along "soft" lines in Eq. (7) and thus leads to a finite region of stability for the columnar dimer. We find

$$[\delta A_k+\delta B_k]_{\rm div}=5\sum_k\frac{1}{\varepsilon_k}$$
$$=\tfrac{5}{2}\int\frac{d^2k}{(2\pi)^2}\frac{1}{|\sin k_x\cos k_y/2|}\propto\ln^2\Lambda\,, \tag{9}$$

where Λ is a gap in the spin-wave spectrum produced by quantum fluctuations. This gap should be calculated from the condition of self-consistency: $\Lambda\sim g\ln^2\Lambda$, where g is the (presumably small) coupling constant. At the leading order in g, $\Lambda\sim g\ln^2 g$. Since Λ is positive, a finite shift from $\alpha=\frac{1}{2}$ is required to destabilize the columnar dimer.

Another contribution, also linear in the coupling constant, comes from the second-order terms produced by the cubic vertices. The corresponding self-energy corrections are given by one-loop diagrams and are known to renormalize only the coefficient A_k ($A_k\rightarrow A_k+\Delta A_k$). By explicitly calculating the one-loop diagrams, we have found that ΔA_k is finite and cubic terms do not cancel the divergent contribution coming from the quartic interactions.

We now justify our choice of the columnar dimer. Indeed there is an infinite number of dimer configurations which at the mean-field level have the same ground-state energy $E_0=-3J_1N/8$ as the columnar dimer. Moreover, one can find a whole family of configurations which are stable in the quadratic approximation only at $\alpha=\frac{1}{2}$ (for example, one can transform a part of the horizontal dimers into the vertical ones). However, the first quantum corrections to E_0 (produced by noninteracting bosons) already depend on the shape of the spectrum

$$\Delta E_0=-\frac{3}{4}\left(J_1-\frac{1}{N}\sum_k\varepsilon_k\right). \tag{10}$$

This clearly favors configurations with a maximum number of zero modes in the spectrum. This singles out the columnar dimer because all other dimer configurations have less zero modes at $\alpha=\frac{1}{2}$. For example, another widely discussed ground-state candidate—a staggered dimer[4,5,7] [Fig. 1(b)]—is also stable at the quadratic order only at $\alpha=\frac{1}{2}$ and has a line of soft modes in its spin-wave spectrum, but its dispersion relation at $\alpha=\frac{1}{2}$, $\varepsilon_k=J_1[(1-v_{2k_x})]^{1/2}$, is clearly less "soft" than that of Eq. (7). Thus, zero-point fluctuations favor energetically the columnar dimer: with ΔE as in (10), the gain in energy is $\Delta E=J_1(3/\pi)(1-2/\pi)$.

To summarize our findings, we have introduced a bosonization suited to the study of dimerized states and applied it to the J_1-J_2 model with spin $\frac{1}{2}$. We have found that the columnar dimer has the lowest energy among the dimer states and focused on the stability region of this configuration. At quadratic order in the bosons, the columnar dimer is stable only at $\alpha=\frac{1}{2}$, the classical transition point. However, the leading anharmonic corrections stabilize the dimerized phase in a region of finite width around $\alpha=\frac{1}{2}$.

Of course, the analysis given above does not manage to answer the question of whether the columnar dimer is actually the global minima in the system around $\alpha=\frac{1}{2}$ because short-range corrections to the ground-state energy are by no means small. Nevertheless, it seems worth pointing out that if to restrict with the leading quantum corrections, the energy of the columnar dimer at $\alpha=\frac{1}{2}$, $E_0\approx=-0.52J_1N$, practically coincides with that of the Néel phase ($E_{\rm Néel}\approx-0.54J_1N$) and significantly exceeds the ground-state energy of the $(\pi,0)$ phase ($E_{(\pi,0)}\approx-0.43J_1N$). Also, this value of E_0 is very close to the results obtained in the series[7] ($E_0^s\approx-0.48J_1N$) and numerical[6] ($E_0^n\approx-0.52J_1N$) experiments. This means that the columnar dimer actually is a good candidate to a ground state close to $\alpha=\frac{1}{2}$, though this question definitely needs further investigation.

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[1]P. W. Anderson, Science **235**, 1196 (1987); P. Fazekas and P. W. Anderson, Philos. Mag. **30**, 423 (1974).

[2]D. Reger and A. P. Young, Phys. Rev. B **37**, 5978 (1988); S. Liang, *ibid.* **42**, 6555 (1990); E. Dagotto and A. Moreo, Phys. Rev. Lett. **63**, 2184 (1989); D. Huse and V. Elser, *ibid.* **60**, 2531 (1988); H. Nishimori and H. Nakanishi, J. Phys. Soc. Jpn. **57**, 626 (1988); S. Fujiki, Can. J. Phys. **65**, 489 (1987); Th. Jolicoeur, E. Dagotto, E. Gagliano, and S. Bacci, Phys. Rev. B **42**, 4800 (1990).

[3]L. B. Ioffe and A. I. Larkin, Int. J. Mod. Phys. B **2**, 203 (1988); P. Chandra and B. Doucot, Phys. Rev. B **38**, 9335 (1988); T. Einarsson and H. Johannesson, *ibid.* **43**, 5867 (1991).

[4]N. Read and S. Sachdev, Nucl. Phys. **B316**, 609 (1989), Phys. Rev. Lett. **59**, 799 (1989).

[5]N. Read and S. Sachdev, Phys. Rev. Lett. **66**, 1773 (1991). Note that the $1/N$ expansion allows one to obtain various dimerized phases, but for $N=1$ it predicts a conventional first-order transition in the J_1-J_2 model.

[6]E. Dagotto and A. Moreo, Phys. Rev. Lett. **63**, 2148 (1989); R. R. P. Singh and R. Narayan, *ibid.* **65**, 1072 (1990). For a review, see E. Dagotto, Int. J. Mod. Phys. B **5**, 907 (1991).

[7]M. P. Gelfand, R. R. P. Singh, and D. A. Huse, Phys. Rev. B **40**, 10801 (1989); S. Sachdev and R. N. Bhatt, *ibid.* **41**, 4502 (1990).

[8]H. J. Schulz and T. A. L. Ziman (unpublished).

[9]V. Kalmeyer and R. B. Laughlin, Phys. Rev. Lett. **59**, 2095 (1987); X.-G. Wen, F. Wilczek, and A. Zee, Phys. Rev. B **39**, 11413 (1989).

[10]P. Chandra, P. Coleman, and A. I. Larkin, Phys. Rev. Lett. **64**, 88 (1990); P. Chandra and P. Coleman, Int. J. Mod. Phys. B **3**, 1729 (1989).

[11]A. V. Chubukov, Phys. Rev. B **44**, 392 (1991).

[12]L. Bergomi and Th. Jolicoeur (unpublished).

[13]A. F. Barabanov and O. A. Starykh, Pis'ma Zh. Eksp. Teor. Fiz. **51**, 271 (1990). [JETP Lett. **51**, 311 (1990)]; J. H. Xu and C. S. Ting, Phys. Rev. B **42**, 6861 (1990); H. Nishimori and Y. Saika, J. Phys. Soc. Jpn **59**, 4454 (1990).

[14]F. Mila, D. Poilblanc, and C. Bruder, Phys. Rev. B **43**, 7891 (1991).

[15]C. Bruder and F. Mila, Europhys. Lett. (to be published).

[16]E. Shender, Zh. Eksp. Teor. Fiz. **83**, 326 (1982) [Sov. Phys. JETP **56**, 178 (1982)]; P. Chandra, P. Coleman, and A. I. Larkin, Phys. Rev. Lett. **64**, 88 (1990); J. Phys. Condens. Matter **2**, 7933 (1990); A. Pimpinelli, E. Rastelli, and A. Tassi, *ibid.* **1**, 2131 (1989); A. Moreo, E. Dagotto, Th. Jolicoeur, and J. Riera, Phys. Rev. B **42**, 6283 (1990).

[17]A. V. Chubukov, Pis'ma Zh. Eksp. Teor. Fiz. **49**, 108 (1989) [JETP Lett. **49**, 129 (1989)].

[18]A. V. Chubukov, Phys. Rev. B **43**, 3337 (1991).

[19]A. V. Markelov, Pis'ma Zh. Eksp. Teor. Fiz. **51**, 320 (1990) [JETP Lett. **51**, 365 (1990)].

[20]See, for example, M. I. Kaganov and A. V. Chubukov, Usp. Fiz. Nauk. **153**, 537 (1987) [Sov. Phys. Usp. **30**, 1015 (1987)].

[21]Note that the approximation we use is opposite to what is adopted for $S=\frac{1}{2}$ *ferromagnets* at low temperatures when the density of bosons is small but the interaction between bosons is always strong.