Spontaneous dimer order, excitation spectrum, and quantum phase transitions in the $J_1 - J_2$ Heisenberg model

Valeri N. Kotov

Department of Physics, University of Florida
Gainesville, Florida 32611-8440, USA

Jaan Oitmaa, Oleg Sushkov, and Zheng Weihong

School of Physics, University of New South Wales,
Sydney 2052, Australia

Abstract

We overview some recent work and present new results on the ground state properties and the spectrum of excitations of the two-dimensional frustrated Heisenberg antiferromagnet. Spontaneous dimer order is present in the quantum disordered phase of this model. We study the stability and analyze the structure of the spectrum, including the two-particle singlet excitation branch throughout the disordered phase, as well as in the vicinity of the Néel critical point. The variation of the dimer order parameter is also given, and it is argued that near the critical point it reflects the presence of the low-energy singlet bound state.
I. INTRODUCTION

Many of the fundamental problems in the field of low-dimensional quantum spin systems are those related to the nature of the ground states, the spectrum of excitations, and the stability of quantum phases without long-range order (Sachdev 1999). The major interest boost which these topics have enjoyed is certainly connected to the discovery of the high-Tc superconductivity. The high-Tc materials are doped antiferromagnetic insulators where doping destroys the Néel long-range order, present in the parent compounds (Manousakis 1991). It is believed that the antiferromagnetic correlations are intimately tied to superconductivity - a connection that emphasizes the need for good understanding of the magnetically disordered ground state. In a proposal, advocated by Anderson (1973, 1987), the ground state is viewed as a coherent superposition of spins paired into singlet bonds, which can "resonate" between different configurations. This resonating valence bond (RVB) state is an example of a true spin-liquid - a state that is both translationally and spin-rotationally invariant. Depending on the nature of the correlations between the valence bonds, different varieties of RVB’s have been considered (Fradkin 1991). The spectrum of these systems is however far from being well understood.

A major recent development in the field of high-Tc materials was the discovery of stripes - a spatial modulation of the spin and charge densities, which seems to be related to superconductivity (Tranquada et al. 1995, 1997). In this connection the study of dimerized, valence bond solid-like (VBS) ground states with spontaneously broken translational invariance, has become an important issue. The stability of such dimerized phases was recently investigated in the framework of the $t-J$ model (Sachdev and Vojta 1999a,b, Sushkov 1999, Vojta and Sachdev 1999). Within the approximation schemes used by these authors, spontaneously dimerized ground states were found to be stable in the region of doping, relevant to the high-Tc cuprates. More detailed predictions, relevant to experiments, are currently being developed (Sachdev and Vojta 1999b).

Spontaneous dimer order can also occur due to spin frustrating interactions - this con-
stitutes the subject of the present work. The basic model which shows such behavior is the two-dimensional \( J_1 - J_2 \) Heisenberg model on a square lattice. There are at least two major reasons why this model holds a special place in the physics of spin systems: (1.) By itself it is one of the simplest models which exhibits quantum transitions between long-range ordered phases and a quantum disordered phase - a topic of fundamental interest (Sachdev 1999), (2.) Even though the \( J_1 - J_2 \) model does not contain charge dynamics, the understanding of how translational symmetry is broken in a purely insulating spin background is crucial to the finite doping situation as well. Indeed, the techniques used in the analysis of the \( t - J \) model by Vojta and Sachdev (1999) and Sushkov (1999) have been tested first on purely spin models and rely heavily on the good handling of the zero doping case. Finally, due to its simpler nature, the \( J_1 - J_2 \) model allows for a comprehensive and accurate description of the ground state properties and the spectrum of excitations, which so far is lacking for the \( t - J \) model.

The \( J_1 - J_2 \) model has been discussed in numerous works over the last ten years and some of the important issues that have been addressed are: (1.) How is the Néel order, present for small frustration \( (J_2) \), destroyed as frustration increases, and (2.) Is a quantum disordered phase present in a finite window of frustration, and what is the structure of this phase. Let us mention several representative papers which discuss these points, without attempting to give a comprehensive literature review. Spin-wave calculations, both at the non-interacting level as well as including interactions perturbatively in powers of \( 1/S \) (\( S \) is the spin value), have found that the magnetization decreases with increasing frustration, ultimately vanishing at a critical value (Igarashi, 1993). These calculations however cannot predict the structure of the phase beyond the instability point, or the location of the phase boundary with high accuracy, since as the magnetization decreases more and more powers of \( 1/S \) have to be included (strong spin-wave interactions). Exact diagonalization (ED) of clusters as large as \( N = 36 \) (Schulz et al. 1994, 1996) have found a finite region of quantum disordered (gapped) phase, but have failed to determine with certainty what the dominant correlations or type of order (e.g. dimer, plaquette, etc.) are dominant in this phase. The ED calculations also
suffer from large finite-size corrections, especially for strong frustration. An insight into the structure of the disordered phase was possible with the help of the large $N$ expansion technique (Read and Sachdev 1990, 1991, Sachdev and Read 1991). These authors predicted the quantum disordered phase to be spontaneously dimerized in a particular (columnar) configuration (see figure 1(a)). High order dimer series expansions around this configuration were performed (Gelfand et al. 1989, Gelfand 1990, Kotov et al. 1999a), all confirming its stability in a window of frustration. Thus the spontaneously dimerized state has emerged as the most probable candidate for a disordered ground state. Let us mention in this connection that in one-dimensional systems the Lieb-Schultz-Mattis (LSM) theorem guarantees that a gapped phase in a quantum spin system breaks translational symmetry (Lieb et al. 1961). Extension of the LSM theorem to two-dimensions was proposed (Affleck 1988) but not proven in the most general case. The large $N$ and dimer series results however seem to confirm the validity of the LSM theorem in 2D as well, including, in fact, the case of finite doping (Sachdev and Vojta 1999b).

The present work is devoted to discussion of several aspects of the spontaneously dimerized phase, with particular emphasis on the excitation spectrum, including collective modes (Section II), and the evolution of the spin-spin correlations and the dimer order parameter throughout the disordered phase (Section III). Properties near the Néel ordered - quantum disordered phase are also discussed. A preliminary account of some of the results has been given by Kotov et al. (1999a) and Kotov and Sushkov (1999).

**II. SINGLE-PARTICLE AND COLLECTIVE LOW-ENERGY EXCITATIONS IN THE SPONTANEOUSLY DIMERIZED PHASE.**

The $J_1 - J_2$ model is defined via the Hamiltonian:

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

where $J_1 \geq 0$ is the nearest-neighbor (NN), and $J_2 \geq 0$ - the frustrating diagonal next-nearest-neighbor (NNN) coupling on a square lattice (see figure 1(a)).
phases for small (large) $J_2$ are shown in figure 1(b)(figure 1(c)), and are referred to as the Néel and collinear phase, respectively. The phase diagram of the model, as determined by dimer series expansions in the disordered phase (this work, see below), and Ising series expansions in the two ordered phases (Oitmaa and Zheng 1996), is shown in figure 2. The disordered phase is stable in the interval $0.38 \leq (J_2/J_1) \leq 0.62$, although there is uncertainty in the exact location of the boundaries due to the poorer convergence near them. We proceed with a description of the spectrum of excitations and its evolution in the quantum disordered phase.

A. Triplet spectrum.

In order to calculate the spectrum we use the "strong-coupling" philosophy, i.e. develop perturbative expansions around a "perfect" dimer configuration (represented as a direct product of singlets). The columnar dimerization pattern used in the expansions is shown in figure 1(a). Unlike systems where the dimerization is explicit, i.e. caused by alternation in the Heisenberg exchange, in the $J_1 - J_2$ model there is no formally small expansion parameter (both inter- and intra-dimer couplings are of the same order in the relevant parameter regime). To achieve maximum reliability of our results we have developed and compared two techniques which take into account the inter-dimer interactions. The first one is the dimer series expansion, where a series for the appropriate observable is generated in powers of the inter-dimer interaction to high order (typically around 10). The generation and resummation of such a series as well as other types of linked-cluster expansions involve a lot of technical details (Gelfand et al. 1990). In what follows we will present the results only. Our second approach is based on diagrammatic resummation of selected (infinite) series which we find to give the dominant contribution. The two approaches are quite similar in spirit, but technically very different, thus allowing us to make sure that the (strong) interaction effects are treated properly.

In order to get a feeling for the types of interactions between the dimers, it is useful to
write the Hamiltonian, Eq.(1) in terms of operators creating triplets \((t^\dagger_i)_\alpha\), where \(i\) is the dimer site index, and \(\alpha = x, y, z\) from the "perfect" dimer configuration. Such a representation was discussed by Sachdev and Bhatt (1990), and leads to the effective Hamiltonian:

\[
H = H_2 + H_3 + H_4, \tag{2}
\]

\[
H_2 = \sum_k \left\{ A_k t^\dagger_{k\alpha} t^\dagger_{k\alpha} + \frac{B_k}{2} \left( t^\dagger_{k\alpha} t^\dagger_{-k\alpha} + \text{h.c.} \right) \right\}, \tag{3}
\]

\[
H_3 = \sum_{k_1} R(k_1, k_2) \epsilon_{\alpha\beta\gamma} t^\dagger_{k_1\alpha} t^\dagger_{k_2\beta} t_{k_1+k_2\gamma} + \text{h.c.}, \tag{4}
\]

\[
H_4 = \sum_{k_i} T(k_1 - k_3) (\delta_{\alpha\delta} \delta_{\beta\gamma} - \delta_{\alpha\beta} \delta_{\gamma\delta}) t^\dagger_{k_1\alpha} t_{k_1+k_2-\alpha} t^\dagger_{k_2\beta} t_{k_1+k_2-\alpha}. \tag{5}
\]

where:

\[
A_k = J_1 - \frac{J_1}{2} \cos k_x + (J_1 - J_2) \cos k_y - J_2 \cos k_x \cos k_y = J_1 + B_k, \tag{6}
\]

\[
T(p) = \frac{J_1}{4} \cos p_x + \frac{J_1 + J_2}{2} \cos p_y + \frac{J_2}{2} \cos p_x \cos p_y, \tag{7}
\]

\[
R(p, q) = -\frac{J_1}{4} \sin p_x - \frac{J_2}{2} \sin p_x \cos p_y - \{p \rightarrow q\}. \tag{8}
\]

An important point to be made is that the Hamiltonian, Eq.(2) in terms of the triplets is exact, i.e. the cubic \(R(p, q)\) and the quartic \(T(p)\) vertices are the only ones generated. However in addition one has to make sure that no double occupancy on a single site is allowed, i.e. \(t^\dagger_{i\alpha} t^\dagger_{i\beta} = 0\), which follows from the fact that a triplet is composed of two spins \(S = 1/2\) and consequently it is impossible to create higher on-site spins. When the dimer series is developed, this constraint is taken into account explicitly in every order of the perturbative expansion. On the other hand if we choose to treat the interactions \(H_3\) and \(H_4\) diagrammatically, which is conveniently done in momentum space, it is useful to introduce an (infinite) on-site repulsion \(U\) as an additional vertex in the theory (Kotov et al. 1998):

\[
H_U = U \sum_i t^\dagger_{i\alpha} t^\dagger_{i\beta} t_{i\beta} t_{i\alpha}, \quad U \rightarrow \infty. \tag{9}
\]
Since the interaction is infinite, it has to be treated to infinite order of perturbation theory, i.e. replaced by an effective scattering vertex $\Gamma^{\alpha\beta,\gamma\delta}(k,\omega)$. The latter turns out to have the structure $\Gamma^{\alpha\beta,\gamma\delta}(k,\omega) = \Gamma(k,\omega)(\delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma})$ and has the explicit form (Kotov et al. 1998):

$$[\Gamma(k,\omega)]^{-1} = -\sum_{q} \frac{u_{q}^{2}u_{k-q}^{2}}{\omega - \omega_{q} - \omega_{k-q}} + \left\{ \begin{array}{c} u \to v \\ \omega \to -\omega \end{array} \right\} . \quad (10)$$

Here $u_{k}, v_{k}$ are Bogoliubov coefficients, arising from the diagonalization of the quadratic part $H_{2}$, Eq.(3), which are given by the standard expression $u_{k}^{2}, v_{k}^{2} = \pm 1/2 + A_{k}/2\omega_{k}$. We denote by $\omega_{q}$ the one-particle dispersion, which on a quadratic level is: $\omega_{q} = \sqrt{A_{q}^{2} - B_{q}^{2}}$, and then gets renormalized by the interaction terms. Needless to say, the vertex $\Gamma(k,\omega)$ does not replace exactly $H_{UV}$, but can be viewed as the best approximation for low density of triplets. It is intuitively clear that if the density increases the hard-core constraint becomes harder to satisfy, leading, on a technical level, to generation of additional vertices. We have found however that the triplet density $N_{T} = \langle t_{\alpha i}^{\dagger}t_{\alpha i} \rangle = 3\sum_{q} v_{q}^{2}$ stays around 0.3 throughout the disordered phase, thus justifying our approximation Eq.(10). Next, the corresponding Dyson equation with one-loop self-energies arising from the three vertices Eqs.(4,5,10) is solved self-consistently for the triplet spectrum $\omega(k)$. Explicit formulas and the corresponding diagrams for Hamiltonians with similar structure to Eq.(2) can be found, e.g. in the works by two of us and co-authors (Kotov et al. 1999b, Shevchenko et al. 1999). Here we only present the results. Let us note that, as pointed out by Chubukov and Jolicoeur (1991), the interaction effects are responsible for creating a finite window of frustration where the gap is non-zero, i.e. the dimer phase is stable.

In figure 3 the spectrum calculated by the dimer series expansion (order 8) is plotted. As frustration increases, the minimum of the dispersion shifts from the Néel ordering wave

---

$^{1}$If one neglects the interactions Eqs.(4,5,10), the region of stability is limited to the point $J_{2}/J_{1} = 1/2$ only.
vector $Q_{AF}$ to the collinear ordering $Q_{COL}$. We work in the Brillouin zone of the dimerized lattice (doubled unit cell in the x-direction), where $Q_{AF} = (0, \pi)$ and $Q_{COL} = (0, 0)$. The convergence of the dimer series is quite poor in certain regions of k space which we attribute to decay, caused by an overlap with the two-particle continuum (seen clearly in figure 4). The evolution of the gaps at the two ordering wave-vectors is shown in figure 2. The points where the gap vanishes are the two quantum critical points where transitions take place into the corresponding ordered phases. We estimate the locations of these points, within errorbar, to be $(J_2/J_1)_{c}^{(1)} \approx 0.38$ and $(J_2/J_1)_{c}^{(2)} \approx 0.62$.

In figure 4 we present a comparison between our diagrammatic and dimer series results for the triplet spectrum for a fixed value of frustration $J_2/J_1 = 0.4$. We find that the agreement is excellent. This figure also shows that the error bars on the dimer series curve are the largest (i.e. the series does not converge well) in the k interval where the dispersion enters the scattering continuum, as discussed in the previous paragraph. One can also compare the gaps at $Q_{AF}$ calculated by the two methods (Kotov et al. 1999a). Both approaches produce an almost linear variation of $\omega(Q_{AF})$ in the disordered phase, and give very similar values for $(J_2/J_1)_{c}^{(1)}$, although generally the diagrammatic method gives larger gap values compared to the dimer series. We believe that the combination of the two methods leads to a very accurate description of the spectrum in the quantum disordered phase.

B. Singlet bound state spectrum.

We now turn to the description of the spectrum of collective two-particle excitations with spin $S = 0$. Our motivation for the study of this branch of the spectrum is two-fold: (1.) Quasi one-dimensional systems, such as spin chains and ladders (Sushkov and Kotov 1998, Shevchenko et al. 1999) were found to have well defined singlet bound states. Frustration was also generally found to increase the binding energy of these modes. Collective states in frustrated 2D systems however have not been studied. (2.) A gapped spinless collective mode, generated non-perturbatively via instanton effects, appears in the large-N field theory
solution (Read and Sachdev 1990). Moreover, the energy scale (gap) of this mode determines the variation of the dimer order parameter near the Néel critical point \((J_2/J_1)_c^{(1)}\). It is important to explore this non-trivial connection from the dimer expansion point of view, which takes into account fluctuation effects differently from the large-N approach, and is expected to perform much better numerically.

The wave function of two triplets combined into a state with \(S = 0\) is given by (\(Q\) is the total momentum of the pair):

\[
|\Psi_Q\rangle = \sum_q \Psi(q, Q) t^\dagger_{\alpha, Q/2+q} t^\dagger_{\alpha, Q/2-q} |0\rangle.
\]  

The attraction between the triplets is mainly due to the two-particle scattering vertex \(T(k)\), relative to which the (second-order) contribution of \(R(k_1, k_2)\) turns out to be quite small and will be neglected from now on for simplicity. Diagrammatically the mutual scattering of two quasiparticles is shown in figure 5. The equation determining the bound state energy \(E_Q^S\) reads:

\[
\left[E_Q^S - \omega_{Q/2+q} - \omega_{Q/2-q}\right] \psi(q, Q) = \sum_p M^S(p, q, Q) \Psi(p, Q),
\]

where

\[
M^S(p, q, Q) = \left\{-2[T(p-q) + T(p+q)] + U\right\}.
\]

The hard-core repulsion \((U \to \infty)\) has to be also taken into account by imposing the following condition via a Lagrange multiplier:

\[
\sum_q \psi(q, Q) = 0.
\]

In figure 4 we present our results for the bound state spectrum \(E_k^S\) at \(J_2/J_1 = 0.4\), relatively to the two-particle scattering continuum. The bound state has a finite binding energy \(\epsilon(k)\), defined as \(\epsilon(k) = [(\text{Lower edge of continuum})(k) - E_k^S]\), throughout the Brillouin zone, with the exception of the point \(Q_{AF}\) and its vicinity. The binding for \(k = (0, 0)\) is quite small due to the closeness of the the Néel critical point (see discussion below). The strongest binding takes place for \(k = (\pi, \pi/2)\), where \(\epsilon(\pi, \pi/2) \approx J_1\).
In figure 6 we present the evolution of the bound state singlet gap for $k = (0, 0)$, $E^S(0, 0)$ which will be of particular interest to us in the next section. This gap is larger that the one-particle triplet gap $\omega(Q_{AF}) \equiv \Delta$, and appears to go to zero (within our resolution), along with $\Delta$, as the Néel transition point $(J_2/J_1)^{(1)}_c \approx 0.38$ is approached. We must note that our treatment breaks down at the very vicinity of $(J_2/J_1)^{(1)}_c$ since divergences in our diagrams start appearing for $\Delta \rightarrow 0$. Such divergences can presumably be summed by renormalization group techniques, which we have not attempted in the present work. Therefore the values of the gaps for $J_2/J_1 = 0.38$, shown in figure 6 should be viewed as being practically zero within the error of our calculation at that point. It is clear that since for $k = (0, 0)$ the binding energy is given by $\epsilon(0, 0) = 2\Delta - E^S(0, 0)$, it must approach zero at the critical point $(J_2/J_1)^{(1)}_c$. The full evolution of $\epsilon(0, 0)$ is shown in figure 6. Around the middle of the quantum disordered phase $J_2/J_1 \approx 0.5$ the binding is large ($\epsilon(0, 0) \approx 0.4J_1$), then it shows an almost linear decrease, and for $J_2/J_1 < 0.41$ becomes smaller than the accuracy of our calculation. Alternatively, in the regime $\Delta \rightarrow 0$ one can study the asymptotics of $\epsilon(0, 0)$ analytically (Kotov and Sushkov 1999) with the result:

$$\epsilon(k = 0) \sim \exp \left(-\frac{C_1}{\Delta}\right), \Delta \ll 1 \implies R \sim \frac{1}{\sqrt{\xi \Delta}} \sim \sqrt{\xi} \exp (C_2 \xi), \xi \sim \frac{1}{\Delta} \gg 1.$$  

Here we have also shown the asymptotic behavior of the radius $R$ of the bound state (defined as the spatial extent of the wave function $\psi(q, Q = 0)$). All energies are measured in units of $J_1$ and $C_1, C_2$ are constants of order unity. Thus we can see that $\epsilon(0, 0)$ vanishes exponentially fast as a function of the triplet gap, implying an exponentially large radius of the composite state. This behavior is certainly not captured well numerically in the vicinity of the critical point.

The properties of the spectrum near the spontaneously dimerized-Néel critical point are quite peculiar. In addition to the triplet gap $\Delta$, the singlet (bound state) gap goes to zero as well. However the spectral weight of the composite singlet, which is determined by
its binding energy (or size), approaches zero exponentially fast near the transition point. For this reason we do not expect that the singlet mode can affect the critical dynamics of the triplets and change the $O(3)$ universality class, associated with such a transition. To compare our results with the large-$N$ approach we notice that the latter also predicts two large length scales near the critical point, corresponding to the small gaps of a triplet and a spinless collective mode (Read and Sachdev 1990). One of the scales was found to be a (large) power of the other one and it was argued (Chubukov et al. 1994) that this is characteristic of a "dangerously irrelevant" coupling, i.e. a coupling which is irrelevant at the critical fixed point, but relevant in the phase, from which the transition is approached. Thus the overall structure of the spectrum as well as the appearance of the additional singlet mode and its effect on the critical properties seem to be quite similar in ours and the large-$N$ field theory approach. We must notice however that the construction of the spectrum of excitations (and consequently all associated details) are very different in the two approaches and comparison has not been attempted.

### III. DIMER ORDER PARAMETER.

In this section we discuss the behavior of the dimer order parameter (DOP), mainly focusing on two issues: (1.) How does the DOP reflect the structure of the spectrum found in the previous section? (2.) Does the DOP go to zero at the Néel critical point, i.e. is spontaneous dimerization possible in the ordered phase? Even though we can not give completely definite answers to these questions, we believe we can shed some light on the issues. The large-$N$ approach gives the following prediction for the DOP near the critical point (Read and Sachdev 1990): $\text{DOP} \sim \Delta \text{const.}N$, $\Delta \to 0$, where $N \gg 1$ is the parameter of the $1/N$ expansion ($N = 1$ is the physical limit), and the constant is of order unity. Therefore a very fast (even singular) vanishing of the DOP at the critical point is expected.

---

$^2$This is in contrast to the spectral weight of the triplet, which stays finite at the transition.
in this scenario. However other works (Gelfand et al. 1989, Sachdev and Bhatt 1990), which are similar in treatment to ours, have not found any substantial decrease of the DOP near the transitions point, thus leaving the issue still open.

Let us start by examining the spin-spin correlation functions, defined as: \( \rho_x = \langle S_2.S_3 \rangle \), \( \rho_y = \langle S_1.S_5 \rangle \), \( \rho = \langle S_1.S_2 \rangle \), where the spin indexes refer to the sites, as numbered in figure 1. In figure 7 we plot the variation of these quantities obtained by dimer series expansions to order nine. The strength of the inter-dimer correlations in the \( x \) direction is weaker than in the \( y \) direction in most of the disordered phase, with the exception of the regions around the two critical points. Therefore the spontaneously dimerized phase can be viewed as a system of weakly coupled two-chain ladders (the chains running in the \( y \) direction). This interpretation was pointed out by Singh et al. (1999).

The dimer order parameters in the two directions are defined as: \( D_x = \rho_x - \rho \), \( D_y = \rho_y - \rho \), and their variation is shown in figure 8. Unlike the previously reported dimer series calculations (Gelfand et al. 1989), our results show a substantial decrease of both order parameters near the Néel critical point. In fact \( D_y \) appears to be zero in a wide region around this point, up to \( J_2/J_1 \approx 0.45 \). The difference between ours and the previous results is most likely due to the longer series that we have obtained, and consequently a better treatment of the inter-dimer interactions. In figure 8 we present also our diagrammatic result for \( D_x \) which clearly shows a very small variation (\( D_y \) has similar behavior). This is quite puzzling, as the dimer series and the diagrammatic approach give very similar results for the triplet spectrum. We have identified the source of this discrepancy to be in the very small variation of the in-dimer spin-spin correlation \( \rho \) in the diagrammatic calculation. Figure 9 shows this quantity, calculated by the two methods. Indeed, a substantial difference is found in the vicinity of the critical point. An important effect, leading to an explanation of this difference was discussed by Kotov et al. (1999a). Let us elaborate more on that point. One can easily see that the correlator \( \rho \) is related to the density of quasiparticles \( N_T \) via:
\[ \rho = -\frac{3}{4} + N_T, \quad N_T = \langle t_{\alpha i}^\dagger t_{\alpha i} \rangle = 3 \sum_q v_q^2. \]  

(16)

From figure 9 the dimer series estimates \( N_T \approx 0.43 \) at the critical point, to be compared with the smaller diagrammatic value \( N_T \approx 0.3 \). The Bogoliubov coefficient \( v_q \) is determined by the strength of quantum fluctuations \( B_q \) (Eq. (3)), as can be formally seen from its definition (see text following Eq.(10)). A sharp increase in \( B_q \) can be due to a strong mixing between the ground state (singlet) and a nearby low-lying singlet state. As discussed in the previous section the two-particle singlet bound state has a very low energy near the Néel critical point and therefore can influence substantially the quantum fluctuations. Technically this effect is given by the diagram for the anomalous Green’s function (which renormalizes \( B_q \)), shown in the inset of figure 9. In order to demonstrate the influence of the bound state on \( B_q \) let us consider a simplified form of the two-particle interaction \( T(k) \) and assume that this is the only term contributing to the formation of the bound state (i.e. neglect the \( U \) and \( R \) contributions). Choose \( T(k) = (J_1/4) \cos k_x \) (i.e. only the \( x \) part of the interaction), which leads to a separable kernel \( M^S(p, q) = -J_1 \cos p_x \cos q_x \) (Eq.(13)), and thus allows us to write the solution in closed form. In this simple case the diagram in figure 9 leads to the following renormalization:

\[ B_k \rightarrow B_k - \frac{J_1 \cos k_x \sum_p u_p v_p \cos p_x}{1 - (J_1/2) \sum_q (\cos^2 q_x/\omega_q)}. \]  

(17)

The expression in the numerator is the first order self-energy contribution, which more generally is: \( \Sigma_{4,A}(k) = -4 \sum_q T(q + k) u_q v_q \), while the denominator is accumulated from the resummation of the series, shown in figure 5. Eq. (17) is to be compared with the equation for the energy of the singlet bound state (Eq.(12)), which in this case is:

\[ 1 = -J_1 \sum_q \frac{\cos^2 q_x}{E_q^S - \omega_q/2 + \omega_q/2 - q}. \]  

(18)

Setting \( Q = (0, 0) \) in Eq.(18) one can see that as the energy of the bound state \( E^S_{Q=(0,0)} \) decreases, the correction to \( B_k \) increases (and in fact diverges in the limit \( E^S_{Q=(0,0)} \rightarrow 0 \)), leading also to an increase in the density \( N_T \).
Although we have not performed a fully self-consistent calculation of the effect discussed above, it is quite clear that the low-energy singlet bound state is mainly responsible for the increase of the quantum fluctuations near the Néel critical point. For comparison, in the large-N theory of Read and Sachdev (1990) there is a direct relationship between the gap in the singlet spectrum and the dimer order parameter, leading to the fast vanishing of the latter. Even though our dimer series results are consistent (within errorbar) with vanishing of the dimer order parameter, we certainly can not determine its critical behavior.

IV. SUMMARY AND OUTLOOK.

To summarize our main results:

(1.) We have calculated the one-particle triplet as well as the collective two-particle singlet excitations. Both branches of the spectrum were found to be stable (i.e. gapped) in the spontaneously dimerized phase.

(2.) The singlet bound state mode reflects the presence of spontaneous dimer order in the system. The singlet gap ($k=(0,0)$) vanishes at the Néel quantum critical point.

(3.) The spontaneous dimer order parameters ($D_x$ and $D_y$) vanish at the Néel quantum critical point. We have presented arguments that the disappearance of the dimer order is related to presence of the low-energy (ultimately gapless) singlet collective mode.

(4.) The collective singlet mode does not influence the quantum critical dynamics of the triplets, since its spectral weight vanishes exponentially fast at the critical point. Thus the $O(3)$ universality class describes the transition.

The above conclusions are consistent with the large-N field theory predictions for this model (Read and Sachdev 1990).

So far we have not analyzed the region of the phase diagram near the transition to the collinear phase, $(J_2/J_1)_{\xi}^{(2)} \approx 0.62$. Based on the analysis of Section III it would be tempting to take as a starting point of the analysis a system of weakly interacting spin ladders. This description certainly appears to be well justified in the neighborhood of the
point \( J_2/J_1 = 0.5 \). The Heisenberg model on a frustrated spin ladder has been previously analyzed (Zheng et al. 1998, Kotov et al. 1999b) and a fairly good understanding of the various phases was achieved. However it is not clear how far this analogy can be used, since near \( (J_2/J_1)^{(2)} \) the strength of the inter-dimer correlations in the two spatial directions is practically the same, thus making the single ladder problem not representative of the full two-dimensional one. We also note that the quantum transition near \( (J_2/J_1)^{(2)} \) appears to be of first order (Singh et al. 1999).

Other possible types of spontaneous order, such as the plaquette one, have also been discussed in the literature. Two works have favored this order as a good candidate for a ground state in the quantum disordered regime (Zhitomirsky and Ueda 1996, Capriotti and Sorella 1999), whereas a recent work based on plaquette series expansions has found this configuration to be unstable (Singh et al. 1999), thus presenting strong evidence that plaquette order does not take place in the \( J_1 - J_2 \) model.

Finally we mention two frustrated spin systems that could be analyzed with techniques, similar to the ones used in this work.

(A.) The isotropic two-dimensional Heisenberg model on a triangular lattice is believed to be long-range ordered with a non-collinear spin arrangement, but with a very small magnetization. A small variation of the couplings however stabilizes a spontaneously dimerized phase in this system (Zheng et al. 1999). Since the quantum transition between a non-collinear and a quantum disordered phase is expected to be in the \( O(4) \) universality class (Chubukov 1991, Azaria and Delamotte 1994), the properties of singlet modes in such a system are of particular interest. A singlet mode, in addition to the triplet one, is expected to become gapless and relevant at the transition point. This scenario is very different from the one discussed in the present work.

(B.) The Heisenberg model on a Kagomé lattice was found to have very peculiar properties, such as a gap to triplet excitations (i.e. a disordered ground state), with many low-energy singlet excitations in the gap (Waldtmann et al. 1998, Mila 1998). This structure of the spectrum suggests that a simple spontaneous dimerization pattern (of the type discussed
in this work) is highly unlikely. A more probable scenario is a larger ”dominant” cluster with strong resonance between the dimers. It remains to be seen whether translational invariance is broken in this system, or it represents an example of a true spin liquid.

ACKNOWLEDGMENTS

We would like to thank S. Sachdev, R. Singh, N. Read, C. Lhuillier, F. Mila, O. Starykh, M. Vojta, C. Biagini, and A. Chernyshev for stimulating discussions on these and related topics. V.N.K. was supported by NSF grant DMR9357474 at the University of Florida and is grateful to S. Hershfield for his encouragement and interest in the work. J.O., O.S. and Z.W. acknowledge the support of the Australian Research Council.
REFERENCES

Anderson, P.W., 1973, Mater. Res. Bull., 8, 153.
Anderson, P.W., 1987, Science, 235, 1196.
Affleck, I., 1988, Phys. Rev. B, 37, 5186.
Azaria, P. and Delamotte, B., 1994, in Magnetic systems with competing interactions, ed. H.T. Diep, (World Scientific, Singapore), p. 51.
Capriotti, L. and Sorella, S., 1999, preprint, [cond-mat/9911161].
Chubukov, A.V., 1991, Phys. Rev. B, 44, 5362.
Chubukov, A.V. and Jolicoeur, T., 1991, Phys. Rev. B, 44, 12050.
Chubukov, A.V., Sachdev, S., and Ye, J., 1994, Phys. Rev. B, 49, 11919.
Fradkin, E., 1991, Field theories of condensed matter systems, (Addison-Wesley), p. 129.
Gelfand, M.P., Singh, R.R.P., and Huse, D.A., 1989, Phys. Rev. B, 40, 10801.
Gelfand, M.P., Singh, R.R.P., and Huse, D.A., 1990, J. Stat. Phys., 59, 1093.
Gelfand, M.P., 1990, Phys. Rev. B, 42, 8206.
Igarashi, J., 1993, J. Phys. Soc. Jpn., 62, 4449.
Kotov, V.N., Sushkov, O.P., Zheng, W.H., and Oitmaa, J, 1998, Phys. Rev. Lett., 80, 5790.
Kotov, V.N., Oitmaa, J., Sushkov, O.P., and Zheng, W.H., 1999a, Phys. Rev. B, 60, 14613.
Kotov, V.N., Sushkov, O.P., and Eder, R., 1999b, Phys. Rev. B, 59, 6266.
Kotov, V.N. and Sushkov, O.P., 1999, preprint, [cond-mat/9907178].
Lieb, E., Schultz, T., and Mattis, D., 1961, Ann. Phys. (N.Y.), 16, 407.
Manousakis, E., 1991, Rev. Mod. Phys., 63, 1.
Mila, F., 1998, Phys. Rev. Lett., 81, 2356.
Oitmaa, J. and Zheng, W.H., 1996, Phys. Rev. B, 54, 3022.
Read, N. and Sachdev, S., 1990, Phys. Rev. B, 42, 4568.
Read, N. and Sachdev, S., 1991, Phys. Rev. Lett., 66, 1773.
Sachdev, S. and Bhatt, R.N., 1990, Phys. Rev. B, 41, 9323.
Sachdev, S. and Read, N., 1991, Int. J. Mod. Phys. B, 5, 219.
Sachdev, S., 1999, *Quantum Phase Transitions*, (Cambridge University Press, Cambridge).

Sachdev, S. and Vojta, M., 1999a, preprint, cond-mat/9908008.

Sachdev, S. and Vojta, M., 1999b, preprint, cond-mat/9910231.

Schulz, H.J., Ziman, T.A.L., and Poilblanc, D., 1994, in *Magnetic systems with competing interactions*, ed. H.T. Diep, (World Scientific, Singapore), p. 120.

Schulz, H.J., Ziman, T.A.L., and Poilblanc, D., 1996, J. Phys. I (Paris), 6, 675.

Shevchenko, P.V., Kotov, V.N., and Sushkov, O.P., 1999, Phys. Rev. B, 60, 3305.

Singh, R.R.P., Zheng, W.H., Hamer, C.J., and Oitmaa, J., 1999, Phys. Rev. B, 60, 7278.

Sushkov, O.P. and Kotov, V.N., 1998, Phys. Rev. Lett., 81, 1941.

Sushkov, O.P., 1999, preprint, cond-mat/9907400.

Tranquada, J.M., Sternlieb, B.J., Axe, J.D., Nakamura, Y., and Uchida, S., 1995, Nature, 375, 561.

Tranquada, J.M., Axe, J.D., Ichikawa, N., Moodenbaugh, A.R., Nakamura, Y., and Uchida, S., 1997, Phys. Rev. Lett., 78, 338.

Vojta, M. and Sachdev, S., 1999, Phys. Rev. Lett., 83, 3916.

Waldtmann, C., Everts, H.U., Bernu, B., Sindzingre, P., Lhuillier, C., Lecheminant, P., and Pierre, L., 1998, Eur. Phys. J. B, 2, 501.

Zheng, W.H., Kotov, V.N., and Oitmaa, J., 1998, Phys. Rev. B, 57, 11439.

Zheng, W.H., McKenzie, R.H., and Singh, R.R.P., 1999, Phys. Rev. B, 59, 14367.

Zhitomirsky, M.E. and Ueda, K., 1996, Phys. Rev. B, 54, 9007.
FIGURES

FIG. 1. (a) Square-lattice $J_1$-$J_2$ model. The ovals represent spins bound into singlets, forming the columnar dimer configuration. (b) Néel ordered state with wave-vector $Q_{AF} = (\pi, \pi)$. (c) Collinear state with $Q_{COL} = (\pi, 0)$.

FIG. 2. Phase diagram of the model, as determined by high-order series expansions. Ising expansions to order 10 (9) for the magnetization $M$ were used in the Néel (collinear) phase. The quantum disordered phase is characterized by a gap in the triplet spectrum $\omega(k)$, determined by the dimer series expansion to order 8. The series were summed by using standard techniques.

FIG. 3. Triplet excitation spectrum for two values of frustration in the quantum disordered phase. Notice that we work in the Brillouin zone of the dimerized lattice, where $Q_{AF} = (0, \pi)$ and $Q_{COL} = (0, 0)$.

FIG. 4. Excitation spectrum, including the two-particle singlet collective mode, for a value of frustration close to the transition point into the Néel phase. The solid and the dot-dashed lines are the triplet spectrum calculated by the dimer series expansion and diagrammatically, respectively. The long-dashed line represents the spectrum of the singlet bound state, and the shaded region is the two-particle scattering continuum.

FIG. 5. Equation for the two-particle scattering amplitude (filled square), whose poles determine the energies of the bound states.

FIG. 6. Bound state singlet (dashed line) and triplet (solid line through filled squares) gaps, calculated diagrammatically. Opens squares represent the calculated singlet binding energy, and the solid line is the fit, based on the asymptotic formula Eq.(15).

FIG. 7. Nearest-neighbor spin-spin correlation functions inside a dimer ($\rho$) and in the $x$ ($\rho_x$) and $y$ ($\rho_y$) directions between dimers (see text for definitions). All results are obtained by the dimer series expansion.
FIG. 8. The dimer order parameters $D_x$ and $D_y$ in the two directions, calculated by the dimer series expansion (points, connected by solid lines). For comparison, the diagrammatic result for $D_x$ is also plotted (dashed line).

FIG. 9. The in-dimer spin-spin correlation function $\rho$ (see text for definition). Comparison is made between the dimer series and the diagrammatic result, which does not take into account self-consistently the diagram, shown in the inset. The filled square in the diagram represents the effective two-particle interaction from figure 5.
FIG.1., Kotov et al.
Square-lattice $J_1$--$J_2$ model

$M(Q_{AP})$  $\omega(Q_{AP})$  $\omega(Q_{COL})$  $M(Q_{COL})$

Neel order  Quantum disorder  Collinear order

FIG.2., Kotov et al.
FIG. 3., Kotov et al.
FIG. 4., Kotov et al.
BETHE-SALPETER EQN. FOR THE EFFECTIVE TWO-PARTICLE INTERACTION VERTEX

\[ \begin{array}{c}
\begin{array}{c}
\begin{array}{c}
\quad = \quad \begin{array}{c}
\text{bare vertex}\end{array} \quad + \quad \begin{array}{c}
\text{bethe-salpeter eqn.}
\end{array}
\end{array}
\end{array}
\end{array} \]

BARE VERTEX

\[ \begin{array}{c}
\begin{array}{c}
\begin{array}{c}
\quad = \quad \begin{array}{c}
\text{bare vertex}\end{array} \quad + \quad \begin{array}{c}
\text{bare vertex}\end{array} \quad + \quad \begin{array}{c}
\text{bare vertex}\end{array}
\end{array}
\end{array}
\end{array} \]

FIG.5., Kotov et al.
FIG. 6., Kotov et al.
FIG. 7., Kotov et al.
FIG. 8., Kotov et al.
CRITICAL POINT

DIMER SERIES

IN-DIMER SPIN-SPIN CORRELATIONS

\[ J_2/J_1 \]

Neel CRITICAL POINT

\[ 0.4 \quad 0.6 \]

\[ -0.2 \quad -0.6 \]

FIG.9., Kotov et al.