Low-energy singlet and triplet excitations in the spin-liquid phase of the two-dimensional $J_1 - J_2$ model

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We analyze the stability of the spontaneously dimerized phase of the frustrated Heisenberg antiferromagnet - the $J_1 - J_2$ model, in two dimensions. The lowest triplet excitation, corresponding to breaking of a singlet bond, is found to be stable in the region $0.38 \lesssim J_2/J_1 \lesssim 0.62$. In addition we find a stable low-energy collective singlet mode, which reflects the spontaneous violation of the discrete symmetry. The spontaneous dimerization vanishes at the point of second-order quantum transition into the Néel ordered phase ($\langle J_2/J_1 \rangle < 0.38$). We argue that the disappearance of dimer order is related to the vanishing of the singlet energy gap at the transition point.

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The nature of excitations in the quantum disordered phases of low-dimensional quantum antiferromagnets is a topic of fundamental importance for the physics of quantum magnetism [1]. Such phases can result from mobile frustrating interactions. Even though it has been intensively studied during the last ten years, the $J_1 - J_2$ model still holds many secrets, especially concerning its quantum disordered phase. Exact diagonalization studies [8] have shown that the excitation spectrum of the model is quite complex and that finite-size effects are large [9]. Spin-wave like expansions around the ordered phase (which occurs for small frustration) naturally can not give any information about the disordered phase at stronger frustration, and consequently non-perturbative methods are needed to analyze the latter regime.

A new insight into the disordered regime was achieved by field-theory methods [4], dimer series expansions [5] and related effective Hamiltonian approaches [6]. The above works have shown that the ground state in the disordered regime is dominated by short-range singlet (dimer) formation in a given pattern (see Fig.1.). The stability of such a configuration implies that the lattice symmetry is spontaneously broken and the ground state is four-fold degenerate. Such a route towards quantum disorder is known rigorously to take place in one dimension, where the Lieb-Schultz-Mattis (LSM) theorem guarantees that a gapped phase always breaks the translational symmetry and is doubly degenerate, whereas gapless excitations correspond to a unique ground state [7]. A generalization of the LSM theorem to higher dimensions has been suggested [10], but can not be rigorously proven.

In this Letter we study further the stability of the spontaneously dimerized phase in the $J_1 - J_2$ model and find that in the disordered phase not only the one-particle triplet excitations but also the two-particle singlet modes are stable. Both types of excitations are gapped and become gapless at the transition point to the Néel phase. The singlet collective mode is a low-energy excitation of the system and we argue that it is responsible for the decrease and ultimately the vanishing of the dimer order parameter as the transition is approached from the disordered side. Since the excitation spectrum is complex and close to the transition point the triplet is not the only low-energy excitation, it is quite possible, in our view, that the $O(3)$ non-linear sigma model in its usual formulation [11] is not the correct effective field theory for the frustrated antiferromagnet.

The Hamiltonian of the $J_1 - J_2$ model reads:

$$H = J_1 \sum_{n \neq n} S_i S_j + J_2 \sum_{n \neq n} S_i S_j,$$

where $J_1$ is the nearest-neighbor, and $J_2$ is the frustrating next-nearest-neighbor Heisenberg exchange on a square lattice (see Fig.1.). Both couplings are antiferromagnetic, i.e. $J_{1,2} > 0$ and the spins $S_i = 1/2$. In order to study the excitations and their stability in the dimer phase we first derive an effective Hamiltonian in terms of bosonic operators creating triplets $t_{i_\alpha}^\dagger$, $\alpha = x, y, z$ from the singlets formed by each pair of spins, as shown in Fig.1. Similar effective theories have been derived in Refs. [12] and we only present the result: $H = H_2 + H_3 + H_4 + H_U$,

$$H_2 = \sum_{k,\alpha} \left\{ A_k t_{k_\alpha}^\dagger t_{k_\alpha}^\dagger + \frac{B_k}{2} \left( t_{k_\alpha}^\dagger t_{-k_\alpha} + \text{h.c.} \right) \right\},$$

$$H_3 = \sum_{1+2=3} R(k_1, k_2) t_{k_\alpha}^\dagger t_{k_\beta}^\dagger t_{k_\gamma}^\dagger t_{k_\delta} + \text{h.c.},$$

$$H_4 = \sum_{1+2=3+4} T(k_1 - k_3) \times \left( \delta_{\alpha \beta} \delta_{\gamma \delta} - \delta_{\alpha \gamma} \delta_{\beta \delta} \right) t_{k_1}^\dagger t_{k_2}^\dagger t_{k_3} t_{k_4} t_{k_5} t_{k_6},$$

We also introduce an infinite repulsion on each site $(H_U)$, in order to enforce the kinematic constraint on the Hilbert space $t_{i_\alpha}^\dagger t_{i_\beta}^\dagger = 0$. 

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\[ H_U = U \sum_{i,\alpha,\beta} t_{\alpha i}^+ t_{\beta i}^+ t_{\beta i} t_{\alpha i}, \quad U \to \infty. \] (5)

The constraint is necessary in order to ensure that the bosonic Hamiltonian in terms of the triplet operators corresponds uniquely to the original spin Hamiltonian and no unphysical states appear in the final result. The introduction of \( H_U \) into the effective theory and its subsequent diagrammatic treatment follow closely our previous work Ref. \([10]\).

The following definitions are used in Eqs. (2-4):

\[ A_k = J_1 - J_2 \sum \xi_k \xi_k - J_2 \xi_k \xi_k, \]
\[ B_k = -J_2 \sum \xi_k \xi_k - J_2 \xi_k \xi_k, \] (6)

where \( \xi_k = \cos(k), \quad \gamma_k = \sin(k) \). The matrix elements in the quartic and cubic interaction terms are:

\[ T(k) = \frac{J_1}{4} \xi_k + \frac{J_2}{2} \xi_k + \frac{J_3}{2} \xi_k \]
\[ R(p, q) = -\frac{J_1}{4} \gamma_p \gamma_q - \frac{J_2}{2} \gamma_p \xi_q - \{ p \to q \}. \] (7)

Throughout the paper we work in the Brillouin zone of the dimerized lattice.

The spectrum is determined by the normal Green’s function \([10]\): 
\[ G_N(k, \omega) = \frac{[\omega + A_k(\omega)][\omega + \bar{A}_k(\omega)]}{\omega - A_k(\omega)} + \bar{B}_k^{-1} \] where \( A_k(\omega) = A_k + \Sigma_N(k, \omega) \) and \( \bar{B}_k = B_k + \Sigma_A(k, \omega) \). The normal self-energy \( \Sigma_N \) is given by the diagrams of Fig.2(a) where the effective scattering vertex \( \Gamma \) has to be found by solving the Bethe-Salpeter equation shown in the same figure. The first two diagrams in Fig.2(a) (proportional to \( \Gamma \) and \( T \)) can be calculated as explained in Ref. \([10]\), while the three-particle contribution (third diagram in Fig.2(a)) was discussed in more detail in Ref. \([1]\). We adopt the Brueckner approximation which is based on the smallness of the triplet density \( n_t = \sum_\alpha \langle t_{\alpha i}^+ t_{\alpha i} \rangle \ll 1 \) (Ref. \([10]\)). However for the system under consideration \( n_t \approx 0.3 \) is not very small and additional diagrams have to be summed up to ensure the reliability of the results. This mainly concerns the anomalous self-energy \( \Sigma_A \) shown in Fig.2(b). One improvement was discussed in Ref. \([2]\) and involves summation of rainbow diagrams containing \( U \). This is equivalent to introduction of the term \( \sum \alpha_\beta_\gamma (t_{\alpha i}^+ t_{\beta i} + h.c.) \) into the Hamiltonian and choosing the Lagrange multiplier \( \Lambda \) from the condition \( \langle t_{\alpha i}^+ t_{\beta i} \rangle = 0 \). The second improvement is the summation of rescattering diagrams which take into account the attraction in the singlet channel (see Fig.2(c) and the discussion of the singlet bound state below). The two effects put together give

\[ \tilde{B}_k \approx -\frac{2J_1}{3} \xi_k + 2J_1 \xi_k - \frac{2J_1 J_2}{2J_1 - 0.5} \xi_k \xi_k + \Lambda. \] (8)

The pole of \( G_N(k, \omega) \), which has to be found self-consistently, determines the renormalized one-particle spectrum \( \omega(k) \). Here we only present the results, obtained by a numerical iterative procedure. The dot-dashed line in Fig.3 is the spectrum for the particular value of \( J_2/J_1 = 0.4 \). For comparison we have also performed a dimer series expansion up to order 8, and have resummed the dimer series by using Padé approximants \([3]\). The result for the triplet excitation spectrum is plotted as a solid line in Fig.3. The spectra obtained by the two methods are in excellent agreement. One can also see that in some regions of \( k \) space \( \omega(k) \) lies in the shaded region in Fig.3, which represents the two-particle scattering continuum. The excitations can decay in that region which is reflected in the poorer convergence of the dimer series (larger error bars on the solid curve).

To map out the phase diagram of the model we have studied the instability of the spectrum as \( J_2/J_1 \) varies. At the critical value \( (J_2/J_1)_{c1} = 0.38 \) the gap at \( k = (0, \pi) \) (which corresponds to the Néel ordering wave-vector \((\pi, \pi)\) of the original (non-dimerized) lattice) vanishes, signaling a transition to the Néel ordered phase. For larger frustration the gap grows almost linearly with \( J_2/J_1 \), as shown in Fig.4(a). At \( (J_2/J_1)_{c2} = 0.62 \) an instability towards a collinear phase takes place and the gap at \((0, 0)\) of the original lattice) goes to zero. The transition at \( (J_2/J_1)_{c2} \) appears to be of first order and is quite similar to the transition which occurs in the Heisenberg ladder with frustration \([3]\). The number of triplets increases sharply near this transition point and the excitation spectrum is a complex mixture of one- and many-triplet bound states \([12]\). In what follows we will concentrate on the vicinity of the second-order transition into the Néel phase.

Let us now investigate the spectrum of collective excitations - in particular we have studied the two-particle bound state with \( S = 0 \). This state has the form: \[ |\Psi_Q\rangle = \sum_{p, q} \langle \Psi(q, Q) t_{\alpha p}^+ \xi_{Q/2 + q}^+ \xi_{Q/2}^+ \rangle \approx 1 \] where \( Q \) and \( q \) are, respectively, the total and relative momenta of the two quasiparticles. The wave function \( \Psi \) satisfies the integral equation \( (E_S(Q) - \omega Q^{-2} - \omega_{Q/2}^{-2}) \Psi(q, Q) = 0 \).

Since we have to take \( U \to \infty \), the following condition must be imposed via a Lagrange multiplier:

\[ \sum_q \Psi(q, Q) = 0. \] (9)

The numerical solution of Eq.(9) is presented in Fig.3 with a long dashed line. Notice that the singlet bound state has a very low energy and exists below the two-particle continuum for all wave-vectors. The singlet is gapped everywhere in the disordered phase, however its energy at \( k = (0, 0) \) approaches zero near the transition point to the Néel phase as shown in Fig.4(a). We emphasize that the appearance of a low energy sin-
glet in the spectrum is quite unusual and represents a characteristic feature of frustrated spin systems.

Next we calculate the dimer order parameters in the two spatial directions, defined as (see Fig.1 for notations): $D_x = \langle S_2 S_3 \rangle - \langle S_1 S_2 \rangle$, $D_y = \langle S_1 S_3 \rangle - \langle S_1 S_2 \rangle$. Dimer series expansions to order 9 have been carried out for these quantities and the results are presented in Fig.4(b). Both order parameters appear to approach zero at the transition point to the Néel phase, even though the error bars are also quite large. Our result is very different from former works [3] where no substantial decrease of the order parameter was found. We attribute the difference to the longer series we have generated. We have also calculated the dimer order parameter by using the diagrammatic approach. The results shown in Fig.4(b) suggest that the dimer order is overestimated in this way, since it shows no appreciable decrease as frustration decreases. However we envisage the following physical mechanism which would explain the discrepancy between the two calculations. As discussed earlier, when the transition point $(J_2/J_1)_{c1} = 0.38$ is approached, the energy of the singlet state decreases and ultimately goes to zero. Since the ground state is also a singlet, a strong mixing between the bound state singlet and the ground state must occur near the transition, which effectively leads to the increase of quantum fluctuations. Technically this effect can be taken into account by exact calculation of the diagram shown in Fig.2(b). This diagram represents a contribution to the anomalous Green’s function and thus effectively to the strength of quantum fluctuations. The exact two-particle scattering amplitude $M$ which appears in Fig.2(b) satisfies the equation shown in Fig.2(c). Notice that the bound state equation (1) is the pole of $M$, and it is quite easy to prove that as the bound state energy decreases, the contribution of the diagram in Fig.2(b) increases substantially. The numerical implementation of this procedure is however quite difficult. The renormalization of $B_5$ given by Eq.(6) is a step in this direction, but does not fully take into account the effect. Nevertheless it is clear that the vanishing of the singlet gap at the transition is intimately related to the increased quantum fluctuations and therefore the vanishing of the dimer order parameter.

Finally we present exact diagonalization data on a small cluster which provide further evidence that the ground state is spontaneously dimerized. Since there are four ways the system can choose the dimer pattern in Fig.1, the ground state is expected to be four-fold degenerate. Indeed, three singlet states are clearly seen above the ground state (See Table.I) even though they have non-zero excitation energy on a finite cluster (an exponentially small splitting is expected between them for large cluster size). One also expects that upon introduction of explicit dimerization $\delta$ into the system (i.e. coupling $1+\delta$ on the bond [12, 34], etc. (see Fig.1.) and $1-\delta$ on the bonds [23], [15], etc.), the degeneracy of the ground state will be lifted and therefore the three zero energy singlets, related to the spontaneous dimerization, should disappear. On a finite cluster the energies of these singlets should grow as $\delta \times$ (system size). Indeed one can see from Table.I that the singlet energies decrease with $\delta$, which proves their symmetry breaking origin.

In summary, we have explicitly calculated, for the first time, the lowest triplet and collective singlet excitations of the two-dimensional $J_1 − J_2$ model. Both branches of the spectrum were found to be stable and gapped in the quantum disordered phase and become gapless at the boundary with the Néel ordered phase. We emphasize that a low-lying singlet around $k = (0, 0)$ is present only in a spontaneously dimerized system. In contrast, for systems with explicit, exchange driven dimerization, the binding energy for such a singlet it strictly zero. Thus the low-energy singlet around the Brillouin zone center should be, in principle, observable in quasi two-dimensional systems exhibiting a quantum critical point, separating a dimerized (spin-phonon interaction driven) and a Néel phase [4]. In addition, we have found that the dimer order parameter tends to zero at the transition point and have argued that the disappearance of spontaneous dimer order is tied to the vanishing of the singlet gap. Let us mention that the structure of the spectrum, found in the present work, supports the picture of Read and Sachdev [1], based on their analysis of the $Sp(N)$ field theory. In particular, both in our and in the field-theory approach, a singlet mode associated with the spontaneous dimer order is present.

The example of the $J_1 − J_2$ model provides further support for the idea, put forward some time ago [1], that the Lieb-Schultz-Mattis theorem can be extended to higher dimensions, and the gapped states of quantum systems necessarily break the discrete symmetries of the lattice. In this connection it would be very interesting to investigate if similar conclusions can be reached for systems with finite doping, e.g. the $t − J$ model away from half filling. The recent discovery of stripes in the high-$T_c$ materials [13], as well as the analysis of the $Sp(2N) t − J$ model for large $N$ [10], strongly suggest that the disordered ground states at finite doping may be unstable towards configurations which break the lattice symmetries. Further analysis of these possibilities is highly desirable.

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FIG. 1. Schematic representation of the system. The ovals represent two spins coupled into a singlet.

FIG. 2. (a) Lowest order diagrams for the normal self-energy $\Sigma_N$. (b) Contribution to the anomalous self-energy $\Sigma_A$. (c) The Bethe-Salpeter equation for the two-particle scattering amplitude $M$.

FIG. 3. Excitation spectrum for $J_2 = 0.4J_1$. The dot-dashed and solid lines are the diagrammatic and dimer series results for the triplet dispersion, respectively. The long dashed line is the two-particle singlet excitation. The shaded area represents the two-particle scattering continuum.

FIG. 4. (a) Triplet and singlet energy gaps in the disordered phase. Solid lines represent the dimer series results for the triplet gaps. The dotted and dashed lines are the diagrammatic calculations of the singlet and triplet gaps, respectively. (b) The dimer order parameters $D_x$ and $D_y$ (see text for definitions). Dashed lines are the diagrammatic results and solid lines are the dimer series results.

**TABLE I.** Exact diagonalization results on $N=4\times4$ cluster for the energies of the lowest five excited states at $k = (0,0)$. Frustration is fixed at $J_2/J_1 = 0.5$ and $\delta$ is the explicit dimerization. "s" and "t" denote singlet and triplet levels.

| $\delta$ | 0 | 0.1 | 0.2 | 0.3 |
|----------|---|-----|-----|-----|
| $E_1$    | 0.319 (s) | 0.655 (s) | 0.908 (t) | 0.864 (t) |
| $E_2$    | 0.830 (s) | 0.864 (s) | 0.924 (s) | 1.130 (s) |
| $E_3$    | 0.971 (s) | 1.001 (t) | 1.327 (s) | 1.814 (s) |
| $E_4$    | 1.128 (t) | 1.144 (s) | 1.506 (s) | 2.001 (s) |
| $E_5$    | 1.135 (s) | 1.425 (t) | 1.684 (t) | 2.028 (t) |
Fig. 2.
Fig. 3.
Fig. 4.