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WHAT ARE THE INDICATIONS FOR A QUANTUM SPIN LIQUID IN THE J_1-J_2 ANTIFERROMAGNET ON A SQUARE LATTICE?[1]

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We examine the influence of frustration on the ground-state of the spin-$\frac{1}{2}$ J_1-J_2 Heisenberg antiferromagnet on the square lattice using exact diagonalization, variational wave functions as well as spin-wave approach. The data indicate the breakdown of the conventional collinear antiferromagnetism at a critical value of homogeneous frustration $J_2^{crit} \approx 0.4J_1$. This conclusion is obtained from (i) the analysis of the ground-state wave function, (ii) the spin-spin correlation functions, and (iii) the inspection of low-lying levels. The breakdown of the Néel order is followed by a quantum spin liquid state with enhanced exotic correlations (spin-Peierls, chiral). The analysis of the ground-state wave function of the Néel phase indicates that the Marshall-Peierls sign rule (derived for unfrustrated bipartite lattices) survives till about $J_2 \approx 0.4J_1$. In the strongly frustrated region the wave function significantly deviates from a simple Jastrow type state. In general, we find that both the self-consistent spin-wave calculations and the variational calculations tend to underestimate role of frustration in respect to the breakdown of the Néel ordering.

1. Introduction

The subject of quantum antiferromagnetism in low-dimensional systems has attracted a great interest in recent time [1] in connection with the magnetic properties of the cuprate high-temperature superconductors. However, the low-dimensional quantum spin systems are of interest in their own right as an example of a strongly interacting quantum many-body system. Though we know from the Mermin-Wagner theorem [2] that at any finite temperature the thermal fluctuations are strong enough to destroy magnetic long-range order, the role of quantum fluctuations is less understood. One specific area of research is the square-lattice spin 1/2 Heisenberg antiferromagnet with frustration. It is well-established now that for a system with nearest-neighbour interaction the ground-state is long-range ordered. But going back to Anderson's and

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Fazekas' investigations [3, 4] of the triangular lattice there is the conjecture that quantum fluctuations plus frustration may be sufficient to destroy the long-range order in two dimensions. Hence the situation on the square lattice may be changed drastically by taking into account frustrating next-nearest neighbour interaction. In connection with slightly doped cuprate superconductors the so-called $J_1 - J_2$ model on the square lattice

$$H = J_1 \sum_{i=1}^{N} (\mathbf{s}_i \mathbf{s}_{i+\hat{x}} + \mathbf{s}_i \mathbf{s}_{i+\hat{y}}) + J_2 \sum_{i=1}^{N} (\mathbf{s}_i \mathbf{s}_{i+\hat{x}+\hat{y}} + \mathbf{s}_i \mathbf{s}_{i-\hat{x}+\hat{y}}) \quad (1)$$

was proposed to simulate doping by frustrating diagonal bonds [5]. The parameter J_2/J_1 determines the frustration within the spin system. In the classical spin limit of the $J_1 - J_2$ model there are two regions of conventional antiferromagnetic long-range order : For $J_2/J_1 < 0.5$ the ordinary Néel two-sublattice antiferromagnet is the ground state of the system, whereas for $J_2/J_1 > 0.5$ a collinear antiferromagnet with four sublattices is realized. Precisely at $J_2 = J_1/2$ one finds a high degree of degeneracy of different states. For the extreme quantum case one could expect that the quantum fluctuations are able to open a window of a spin liquid phase without conventional antiferromagnetic long-range order for a finite parameter region around $J_2/J_1 \approx 0.5$ which separates the two antiferromagnetic phases for small and large J_2. However, different theoretical approaches yield controversial results. The linear spin-wave theory [6, 7], extensive exact diagonalization studies [8, 9, 10, 11, 12, 13], 1/N expansion techniques [14], bond operator techniques [15], effective action approaches [16] predict a destruction of the classical Néel order in a finite region around $J_2/J_1 \approx 0.5$, whereas modified spin-wave theories, taking into account spin-wave interactions within a Hartree-Fock scheme, do not find this intermediate phase between the Néel antiferromagnet and the four-sublattice state [17, 18, 19, 20, 21]. The aim of the present paper is to discuss several indications for the existence of the quantum spin liquid in a region around $J_2/J_1 \sim 0.5$.

2. Outline of the methods

For the discussion of the $J_1 - J_2$ model we use three methods, the exact diagonalization of lattices up to $N = 26$ sites, Takahashi's spin wave theory and variational wave functions of Jastrow type.

(i) Exact diagonalization: To construct the ground state $|\psi\rangle$ we can expand it to any complete set $|n\rangle$ in the spin space $|\psi\rangle = \sum_n c_n \, |n\rangle$. Usually $|n\rangle$ **is chosen to be a** direct product of eigen states of s_{iz} (Ising states), which form an orthonormal complete set. The coefficients c_n are directly computed by a numerical diagonalization of the Hamiltonian matrix $\langle n|H|m\rangle$ (Lanczos algorithm, for an illustration see [22, 9]). Since the size of the matrix increases exponentially with N this method is restricted to small systems.

(ii) Variational wave function of Jastrow type: For larger systems the coefficients c_n can be calculated approximately. The coefficients c_n are represented as functions of certain parameters $P_{s,n}$ containing characteristic features of the basis states $|n\rangle$ [23], i.e. $c_n = (-1)^{q_n} f(\{P_{s,n}\})$, where q_n counts the number of up spins in a sublattice, say A.

The expression for the phase factor $(-1)^{q_n}$ is taken from the Marshall-Peierls sign rule [24, 25, 26]. For a Jastrow type wave function the standard ansatz is the exponential one [27, 28, 29, 23] $c_n \sim (-1)^{q_n} e^{f_1 P_{1,n}} e^{f_2 P_{2,n}} \ldots$. The f_α are variational parameters to minimize the energy of the variational state. The classification parameters $P_{s,n}$ count the number of "false spin couples" of the basis state $|n\rangle$ within a given separation shell s. To a certain shell s belong all pairs of sites i and j with fixed separation $|\mathbf{R}_{ij}| = a_s$. A "false spin couple" in a state $|n\rangle$ is a couple of spins l and m with a relative spin orientation opposite to that one in the Néel state, i.e. $\langle n|S_l^z S_m^z|n\rangle = -\langle N\acute{e}el|S_l^z S_m^z|N\acute{e}el\rangle$.

(iii) Spin wave theory: For low-dimensional systems Takahashi's modified spin-wave theory [30, 18, 21] is suitable. In this theory the conventional spin-wave technique is supplemented with a condition for zero sublattice magnetization, in this way fulfilling, by hand, Mermin-Wagner's theorem [2] at finite temperatures, or the same requirement for finite lattices. Formally, this is achieved through the Lagrange multiplier μ which has to be involved in the Hamiltonian, eqn. (1). The main elements of the theory are: 1) some bosonization scheme (say, Dyson-Maleev), 2) a Hartree-Fock decoupling of the quartic terms of the Hamiltonian, 3) an appropriate Bogoliubov transformation diagonalizing the resulting Hamiltonian. For the two-sublattice Néel state this procedure leads to the following equations (see, e.g. [18, 21])

$$\langle \mathbf{s_i s_j} \rangle = f'^2(\mathbf{R}_{ij}) - g^2(\mathbf{R}_{ij}) - \frac{1}{4}\delta(\mathbf{R}_{ij}) \quad , \quad f'(\mathbf{R}_{ij}) \equiv f(\mathbf{R}_{ij}) + \frac{1}{2}\delta(\mathbf{R}_{ij}) \quad , \quad (2)$$

where

$$f(\mathbf{R}_{ij}) \equiv \langle \hat{a}_i^+ \hat{a}_j \rangle = \frac{1}{N} \sum_{\mathbf{k}}{}' e^{-i\mathbf{k}\mathbf{R}_{ij}} (1 - \eta_{\mathbf{k}}{}^2)^{-1/2} - \frac{1}{2}\delta(\mathbf{R}_{ij}) \quad , \quad (3)$$

$$g(\mathbf{R}_{ij}) \equiv -\langle \hat{a}_i^+ \hat{b}_j^+ \rangle = \frac{1}{N} \sum_{\mathbf{k}}{}' e^{-i\mathbf{k}\mathbf{R}_{ij}} \eta_{\mathbf{k}} (1 - \eta_{\mathbf{k}}{}^2)^{-1/2} \quad , \quad (4)$$

$$\eta_{\mathbf{k}} = \frac{\gamma_{\mathbf{k}}}{1 + \mu - (J_2/J_1)U(1 - \Gamma_{\mathbf{k}})} \quad , \quad \gamma_{\mathbf{k}} = \frac{1}{2}(\cos k_x + \cos k_y) \quad , \quad \Gamma_{\mathbf{k}} = \cos k_x \cos k_y \quad . \quad (5)$$

The Bose operators $\hat{a}_i$, $\hat{b}_i$ come from the Dyson-Maleev transformation and live on A and B sublattices, respectively. The prime over sums means that $\mathbf{k}$ vectors run in the small Brillouin zone, and $\delta(\mathbf{R}_{ij})$ is the Kronecker function. The renormalization factor U in eqn.(5), renormalizing J_2/J_1, is a result of a Hartree-Fock decoupling. U is determined by the short-ranged correlators f, g:

$$U = f/g \quad , \quad f = f(\hat{\mathbf{x}} + \hat{\mathbf{y}}) \quad , \quad g = g(\hat{\mathbf{x}}) = g(\hat{\mathbf{y}}) \quad . \quad (6)$$

The relevant fields f, g, and μ can be obtained from the equations

$$1 = \frac{1}{N} \sum_{\mathbf{k}}{}' (1 - \eta_{\mathbf{k}}^2)^{-1/2} \quad , \quad (7)$$

$$g = \frac{1}{N} \sum_{\mathbf{k}}{}' \gamma_{\mathbf{k}} \eta_{\mathbf{k}} (1 - \eta_{\mathbf{k}}^2)^{-1/2} \quad , \quad (8)$$

$$f = \frac{1}{N}\sum_{\mathbf{k}}{}' \Gamma_{\mathbf{k}}(1-\eta_{\mathbf{k}}^2)^{-1/2} \tag{9}$$

Eqn.(7) is a direct result of Takahashi's condition for zero sublattice magnetization relevant for finite lattices. Finally, we add the expression for the ground-state energy per site

$$E/N = -2J_1 g^2 + 2J_2 f^2 \tag{10}$$

Alternatively, the theory of Hirsch and Tang [31] does not take into account the renormalization, i.e. $U = 1$. In this particular case the Lagrange multiplier μ is determined by eqn.(7) only, and then f and g can directly be calculated from eqns.(8,9).

Now, let us explicitely write down the ground-state wave function connected with the theory sketched above. In terms of the two-boson parameterization adopted above, the spin-wave ansatz reads

$$\psi_{sw} \sim \exp\left(\sum_{\mathbf{k}}{}' w_{\mathbf{k}}\ \hat{a}^+_{\mathbf{k}}\hat{b}^+_{-\mathbf{k}}\right)\ |N\acute{e}el\rangle \quad . \tag{11}$$

Here $|N\acute{e}el\rangle$ is the classical Néel state. The weight factors $w_{\mathbf{k}}$ are defined by $w_{\mathbf{k}} = v_{\mathbf{k}}/u_{\mathbf{k}}$, $v_{\mathbf{k}}$ and $u_{\mathbf{k}}$ being the well-known Bogoliubov coefficients

$$2v^2_{\mathbf{k}} = (1-\eta^2_{\mathbf{k}})^{-1/2} - 1 \quad , \qquad 2u^2_{\mathbf{k}} = (1-\eta^2_{\mathbf{k}})^{-1/2} + 1 \quad . \tag{12}$$

A natural way to generalize the spin-wave ansatz, eqn.(11), is produced by the substitution

$$\sum_{\mathbf{k}}{}' w_{\mathbf{k}}\hat{a}^+_{\mathbf{k}}\hat{b}^+_{-\mathbf{k}} \longrightarrow -\sum_{\substack{i,j\\ i\in A}} w(\mathbf{R}_{ij}) s^+_i s^-_j \quad , \tag{13}$$

where the pairing function $w(\mathbf{R}_{ij})$ is defined by

$$w(\mathbf{R}_{ij}) = \frac{2}{N}\sum_{\mathbf{k}}{}' w_{\mathbf{k}} \cos \mathbf{k}\mathbf{R}_{ij} \quad . \tag{14}$$

The vector $\mathbf{R}_{ij}$ in eqns.(13,14) connects sites from different sublattices. In this way, by use of eqn.(13), we come to the following spin ansatz

$$\psi_s \sim \exp\left(-\sum_{\substack{i,j\\ i\in A}} w(\mathbf{R}_{ij}) s^+_i s^-_j\right)\ |N\acute{e}el\rangle \quad . \tag{15}$$

It is easy to see that the wave functions, eqns.(11,15), coincide on the physical subspace. It is natural to introduce also the symmetrized variant of eqn.(15)

$$\psi_s^{sym} \sim \exp\left(-\sum_{\substack{i,j\\ i\in A}} w(\mathbf{R}_{ij}) \left[s^+_i s^-_j + s^-_i s^+_j\right]\right) \left(|N\acute{e}el_1\rangle + |N\acute{e}el_2\rangle\right) \quad . \tag{16}$$

We notice that this state is another representation of a Jastrow state, however, the pairing function $w(\mathbf{R}_{ij})$ comes from the spin wave theory and there are no individual

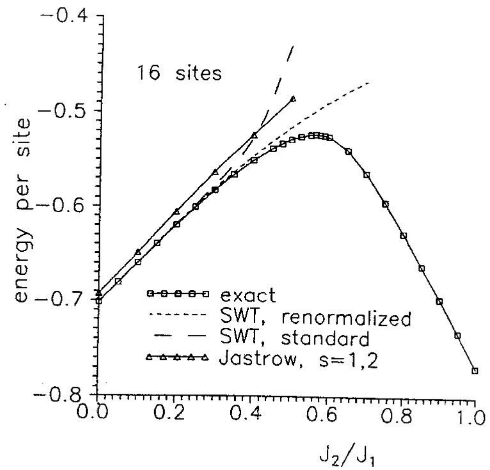

Fig.1. Ground-state energy versus J_2/J_1 for $N = 16$ calculated by exact diagonalization, by standard spin-wave theory without renormalization (i.e. $U = 1$, Hirsch-Tang theory), by spin-wave theory with renormalization ($U \neq 1$, Hartree-Fock decoupling of quartic terms) and by variational Jastrow-wave function with two parameters $P_{1,n}$ (nearest-neighbour shell) and $P_{2,n}$ (next-nearest-neighbour shell)

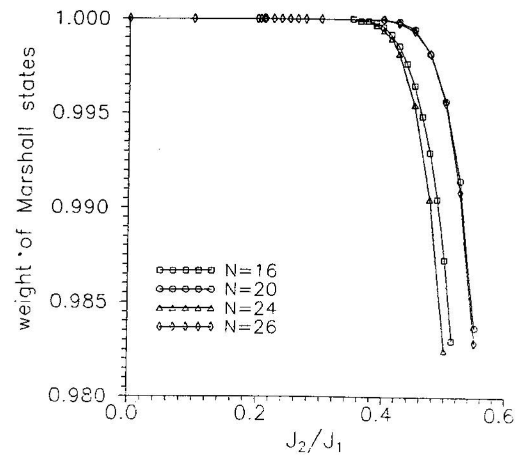

Fig. 2. The weight $\sum_n c_n^2$ of basis states $|n\rangle$ fulfilling the Marshall-Peierls sign rule (i.e. the sum runs over states with $sign(c_n) = (-1)^{q_n}$ only) for square lattices of $N = 16, 20, 24, 26$ sites.

variational parameters f_α for the different shells of paired spins. The factor U, arising from the Hartree-Fock decoupling, can be either taken form eqns. (7,8,9) or, more

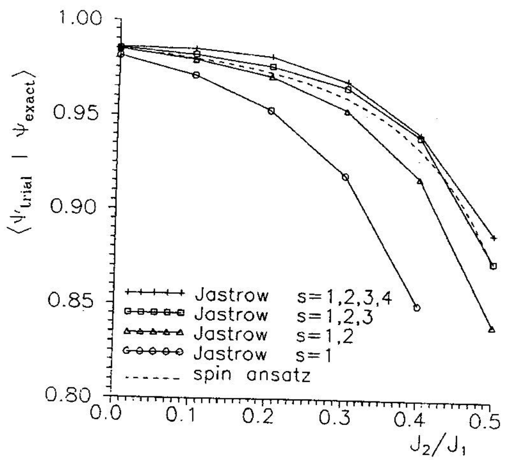

Fig. 3. Overlap of various variational Jastrow functions with different classification parameters $P_{s,n}$ and of the spin ansatz, eqn. (16), with the exact ground state for $N = 16$. For the spin ansatz (16) the parameters U and μ were not taken from eqns. (7,8,9) but are varied to get the best energy.

appropriate, can be used as a free variational parameter in the wave function (16).

3. Results

A. Energy

First we present the energy in dependence on the frustration parameter J_2/J_1 for a lattice of $N = 16$ (Fig.1). The maximum of the energy indicates the region of strongest frustration and coincides according to the Hellmann-Feynman theorem [32] with the point where the next-nearest neighbour correlation $\langle \mathbf{s}_i \mathbf{s}_{i\pm\hat{x}\pm\hat{y}} \rangle$ vanishes. It can be shown that the critical ratio J_2/J_1 where the antiferromagnetic long-range order breaks down (for the order parameter see eqn. (17)) is left from the maximum of the energy [33]. We find the maximum of the exact energy at $J_2/J_1 = 0.56$. It is evident that both the spin-wave theory and the Jastrow wave function describe the energy for low frustration very well but fail in the most interesting region of strong frustration.

B. Ground state wave function

In the limit of $J_2 = 0$ the model fulfills Lieb's and Mattis' [25] criterion for bipartite lattices and consequently the Marshall-Peierls phase rule [24, 25, 26] $sign(c_n) = (-1)^{q_n}$ (see section (2)) is fulfilled. Including the frustrating J_2 the lattice is no more bipartite and the rule may be violated. Recently, we have argued [26] that the rule may survive even for large frustration. We present in Fig.2 some results for the exact ground state wave function for square lattices of $N = 16, 20, 24, 26$ sites with periodical boundary conditions. It is evident that the Marshall-Peierls phase rule gives an excellent description of the phase relationship till $J_2/J_1 \sim 0.4$. Just above this value starts a rearrangement of the phases indicating a significant change in the ground state. The difference between $N = 16$ and $N = 24$ on the one hand and $N = 20$ and $N = 26$ on the other hand is connected with the different symmetries of these lattices for finite J_2 (see [8, 13, 34]). Next we consider in Fig.3 the overlap between several variational Jastrow states including the spin ansatz (16) and the exact wave function for $N = 16$. We find that these wave functions work quite well for small frustration even for a short-range pairing. For strong frustration $J_2/J_1 > 0.4$ the quality of the description is worse and the long-range pairing becomes more important.

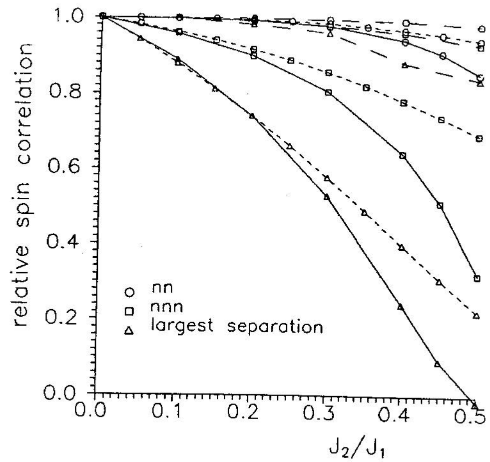

Fig. 4. Pair correlation function $\langle \mathbf{s}_i \mathbf{s}_j \rangle$ scaled by its value for $J_2 = 0$ for nearest-neighbour (nn), next-nearest-neighbour (nnn) and largest separation for the given finite lattice. The solid lines represent the exact results for $N = 24$ (average over both possible configurations $N = 4x6$ and $N = 6x4$), the short-dashed line the results of the Hartree-Fock spin wave theory for $N = 64$ and the long-dashed line the results for the variational Jastrow state with two parameters (shells $s = 1, 2$).

C. Pair correlation

The antiferromagnetic order parameter for finite systems is the square of sublattice magnetization [8, 13]

$$M_s^2 = \frac{1}{N^2} \sum_{i,j=1}^{N} (-1)^{i+j} \langle \mathbf{s}_i \mathbf{s}_j \rangle . \qquad (17)$$

Obviously, this order parameter is based on the pair correlation function $\langle \mathbf{s}_i \mathbf{s}_j \rangle$ multiplied by a staggered factor. For small systems the short-range correlations contribute

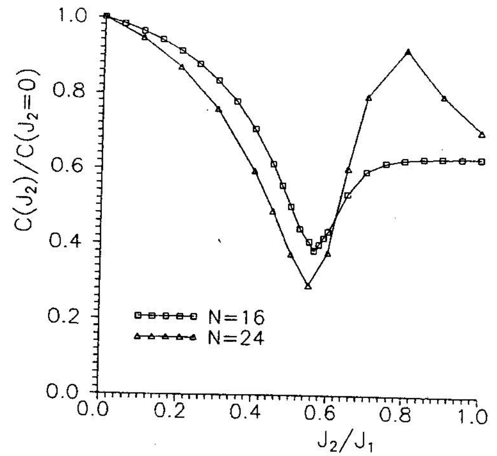

Fig. 5. Total correlation C (18) scaled by its value of the unfrustrated model for $N = 16$ and $N = 24$ (exact diagonalization results).

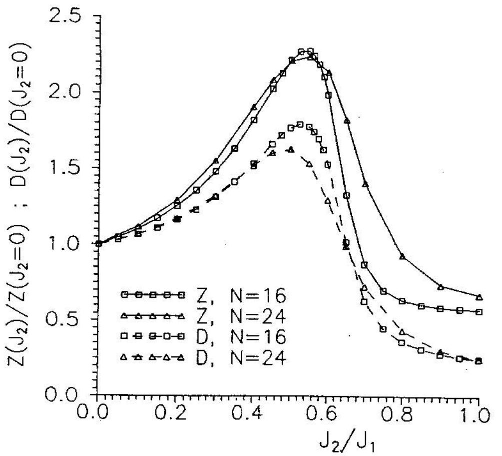

Fig. 6. Exotic order parameters D (19) and Z (20) scaled by its value of the unfrustrated model for $N = 16$ and $N = 24$ (exact diagonalization results). For $N = 24$ the results represent the average over both possible configurations $N = 4x6$ and $N = 6x4$.

substantially to M_s^2, whereas for large systems the prefactor N^{-2} in (17) cancels the short-range correlators and only the long-range correlations are important. Hence we discuss the influence of the frustration on $\langle s_i s_j \rangle$ for different separations $\mathbf{R}_{ij}$. In Fig.4 we present the numerical exact data for $N = 24$ and the Hartree-Fock spin-wave results (eqn. (2), variational U) as well as the results for a variational Jastrow state with

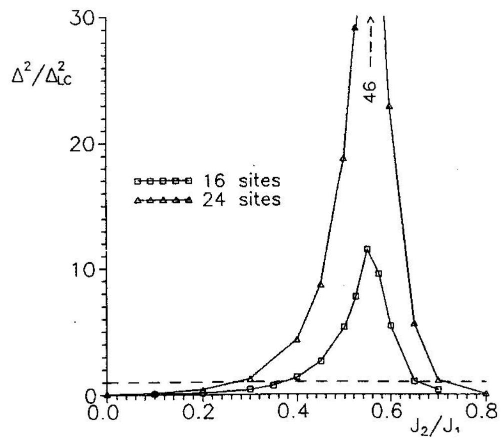

Fig. 7. Mean square deviation Δ^2 of the low-lying eigen values of (1) from the levels of the effective model (22) scaled by the corresponding value Δ^2_{LC} of the linear chain for $N = 16$ and $N = 24$ (exact diagonalization results).

two parameters (shells $s = 1, 2$), both for $N = 64$. It is evident that the frustration supresses the long-range correlations much more than the short-range correlations. In particular, for the exact solution ($N = 24$) the correlation function for the largest separation ($\mathbf{R}_{ij} = (2, 3)$) goes to zero for $J_2/J_1 < 0.5$. In principle, the same tendency is obtained from the spin-wave approach, however, the latter seems to underestimate the role of frustration.

Furthermore, let us define a measure of total correlation

$$C = \sum_{\substack{i,j=1 \\ i \neq j}}^{N} |\langle \mathbf{s}_i \mathbf{s}_j \rangle| \qquad (18)$$

which sums the absolute values of all pair correlators. We present the exact diagonalization results for total correlation scaled by its value for the unfrustrated system in Fig.5 for $N = 16$ and $N = 24$. For small and for large J_2 the correlation is strong and conventional antiferromagnetic long-range order (two sublattices for small J_2 and four sublattices for large J_2, cf. [8, 11, 13]) is realized. The region with weak total correlations is around the point of maximum energy $J_2/J_1 \sim 0.56$ indicating the lack of collinaer long-range order in this area.

D. Exotic correlations

Instead of the conventional collinear magnetic ordering more exotic noncollinear order parameters may be relevant in the region of strong frustration $J_2 \approx 0.5 J_1$. Most favoured candidates are the spin-Peierls (or dimer) order parameter D [8, 35, 11, 13] and the vector-chiral order parameter Z [36, 11, 13]. The first one is defined as

$$D = 2\left\langle\left[\frac{1}{N}\sum_i(-1)^{i_x}\mathbf{s}_i\mathbf{s}_{i+\hat{x}}\right]^2\right\rangle \quad , \tag{19}$$

where i_x is given by the lattice vector $\mathbf{R}_i = (i_x, i_y)$. This quantity describes a columnwise arrangement of spin dimers (singlet states of two spins). The vector-chiral order parameter Z measures the long-range phase coherence of the handiness of triangular plaquettes and is given by

$$Z = \left\langle\frac{1}{3}\left[\frac{1}{2N}\sum_i(\mathbf{Z}_{i,i+\hat{x},i+\hat{x}+\hat{y}} - \mathbf{Z}_{i,i+\hat{x}+\hat{y},i+\hat{y}})\right]^2\right\rangle \tag{20}$$

$$\mathbf{Z}_{ijl} = 8(\mathbf{s}_i\times\mathbf{s}_j + \mathbf{s}_j\times\mathbf{s}_l + \mathbf{s}_l\times\mathbf{s}_i) \tag{21}$$

Both parameters are presented in Fig.6 for $N = 16$ and $N = 24$ (exact diagonalization data). The maxima of both parameters coincide with the maximum in the energy (Fig.1) and the miminum of the total pair correlation (Fig.5). Of course, it remains unclear whether these data for small systems really indicate the existence of exotic long-range order in the thermodynamic limit (cf. [13]).

E. Inspection of low-lying levels

Recently Bernu et al. [37] and Azaria et al. [38] suggested to investigate the arrangement of the low-lying levels belonging to different quantum numbers S of the square of the total spin $\mathbf{S}^2$. These levels should be described by an effective Hamiltonian of the form

$$H_{eff} = E_0 + \frac{K}{N}\mathbf{S}^2 \tag{22}$$

in order to realize a symmetry broken antiferromagnetic Néel state in the thermodynamic limit. We calculated the mean square deviation Δ^2 between the lowest levels with $S = 0, 1, 2, 3$ and the corresponding levels of the effective model (22) in dependence on J_2/J_1 for $N = 16$ and $N = 24$. The results for Δ^2 are drawn in Fig.7, where we have scaled Δ^2 by the respective value of the linear chain (the same number of sites) with nearest-neighbour exchange. For the linear chain it is well-known (Bethe-Hulthen solution) that the quantum fluctuations are strong enough to destroy the long-range order. As expected, the deviation Δ^2 for the square lattice is less than that one for linear chain (i.e. $\Delta^2/\Delta^2_{LC} < 1$) for small and large J_2 but in the strong-frustation area we have $\Delta^2/\Delta^2_{LC} \gg 1$, which is another indication for the breakdown of conventional LRO.

4. Summary

In this paper we present a number of arguments for the breakdown of the conventional collinear antiferromagnetic long-range order in the ground state of the $J_1 - J_2$

model on the square lattice at critical frustration $J_2^{crit}/J_1 \approx 0.4$. These are based on the investigation of the energy, the ground-state wave function, the spin correlation and the arrangement of the low-lying levels by means of exact diagonalization, spin-wave theory and variational Monte-Carlo calculations. The arguments are summarized as follows: (i) The spin-wave theory and the variational wave functions work very well in the region of small frustration where the ground state is long-range ordered but fail in the strong frustration area, which indicates a serious change in the ground state towards a new quality not well described by these theories. (ii) For $J_2 > 0.4J_1$ the phase relationship of the ground state is not precisely described by that one for bipartite lattice. (iii) The frustration particularly supresses the long-distance spin-spin correlation. The short-range correlators are less influenced. (iv) The total strength of pair correlation shows well-pronounced minimum in the region of strong frustration. (v) There are indications of enhanced exotic correlations in this region which are not compatible with collinear antiferromagnetic long-range order. (vi) The arrangement of low-lying levels belonging to different quantum numbers of the square of total spin changes significantly at $J_2 \approx 0.4J_1$ whichs signals of a lack of antiferromagnetic symmetry breaking for strong frustration in the thermodynamic limit.

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