Suppression of Dimer Correlations in the Two-Dimensional $J_1$-$J_2$ Heisenberg Model:

an Exact Diagonalization Study

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We present an exact diagonalization study of the ground state of the spin-half $J_1$–$J_2$ model. Dimer correlation functions and the susceptibility associated to the breaking of the translational invariance are calculated for the $4 \times 4$ and the $6 \times 6$ clusters. These results – especially when compared to the one dimensional case, where the occurrence of a dimerized phase for large enough frustration is well established – suggest either a homogeneous liquid or, possibly, a dimerized state with a rather small order parameter.

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In quantum systems, frustration, i.e., the impossibility to satisfy simultaneously every pairwise interaction, induces unconventional properties down to zero temperature ($T = 0$). In particular it plays a very important role in quantum antiferromagnets where the combined effect of competing interactions and zero-point motion can give rise to non-magnetic ground states which lack the classical Néel long-range order. To this respect the most intriguing situation appears in two spatial dimensions where at $T = 0$ both the magnetic and non-magnetic ground states are allowed by the Mermin-Wagner theorem, and quantum phase transitions from an ordered to a disordered magnetic phase can be triggered by increasing the frustration.\footnote{1}

One of the simplest frustrated spin systems in two dimensions is the so-called $J_1$–$J_2$ Heisenberg model. Here the antiferromagnetic coupling ($J_1 > 0$) between nearest neighbor ($n.n.$) spins is frustrated by the presence of a next-nearest neighbor ($n.n.n.$) superexchange interaction ($J_2 > 0$):

$$\hat{H} = J_1 \sum_{n.n.} \hat{S}_i \cdot \hat{S}_j + J_2 \sum_{n.n.n.} \hat{S}_i \cdot \hat{S}_j,$$

where $\hat{S}_i$ are spin-half operators on a periodic $N$–site square lattice. Recently, the interest in this model has been boosted by the synthesis of three vanadates compounds ($\text{Li}_2\text{VOSiO}_4$, $\text{Li}_2\text{VOGeO}_4$, and $\text{VOMoO}_4$) whose relevant magnetic interactions involve nearest and next-nearest spin-1/2 $V^{4+}$ ions on weakly coupled stacked planes.\footnote{2}

Despite the simplicity of the Hamiltonian, the zero-temperature phase diagram of the spin-half $J_1$–$J_2$ model has been much debated in the last 15 years. For $J_2/J_1 \ll 0.5$, an antiferromagnetic (AF) Néel order is expected, while in the opposite limit, $J_2/J_1 \gg 0.5$ the ground state should be in an AF collinear phase, with the spins ferromagnetically aligned in one direction and antiferromagnetically in the other, corresponding to magnetic wave vectors $Q = (\pi, 0)$ or $Q = (0, \pi)$. The most interesting part of the phase diagram is the one around the fully frustrated point $J_2/J_1 = 0.5$. In fact, although the existence of a gapped (non-magnetic) phase is at present rather likely for $0.4 \lesssim J_2/J_1 \lesssim 0.55$ its characterization is one of the most intriguing puzzles of the physics of strongly correlated systems. In particular, a debated issue is whether the ground state is a homogeneous spin liquid, as originally proposed by Anderson and Fazekas.\footnote{3} The other possibility, following Read and Sachdev’s large-$N$ expansion,\footnote{4} is a spontaneously dimerized phase which breaks the translation invariance of the Hamiltonian or – more in general – one of the lattice symmetries.\footnote{5,9,10,11}

The main reason for the lack of agreement on this delicate issue is due to the fact that most of the existing results have been obtained with approximated numerical and analytical techniques based on reference states explicitly breaking some symmetry of the Hamiltonian,\footnote{12} so that it is very difficult to put on a solid ground any conclusion on the actual ground-state correlations. In this context, the unbiased results provided by the exact diagonalization (ED) of small clusters can give a valuable insight into the $T = 0$ properties of the model.\footnote{13} In particular, on the $4 \times 4$ cluster, Poilblanc and collaborators\footnote{13} showed that the dimer correlations are enhanced in the parameter range $0.5 < J_2/J_1 < 0.6$, concluding that the dimer state is a good candidate for describing the physics of the $J_1$–$J_2$ model in the highly-frustrated region. This claim is questionable because of the smallness of the $4 \times 4$ lattice, where it is very difficult to extract the important long-range behavior of the correlation functions. In this paper, we enlarge the cluster up to $N = 36$ sites and present a more systematic analysis based on \textit{i}) the calculation of the dimer susceptibility in the presence of an explicit symmetry-breaking term and \textit{ii}) the calculation of a static dimer correlation function. In both cases, the direct comparison with the one-dimensional chain, where the dimerization is well established for large enough frustration,\footnote{14} casts doubts on the existence of a dimerized two-dimensional ground state in the non-magnetic regime.
The occurrence of some kind of dimerized order can be detected by calculating the response of the system to operators breaking the lattice symmetries. This can be done by adding to the Hamiltonian a term $-\delta \hat{O}$, where $\hat{O} = \sum_i \delta \hat{O}_i$ is an (extensive) hermitian operator that breaks some symmetry of $\mathcal{H}$ and $\delta$ is a (small) parameter. In fact, if true long-range order in the dimer correlations exists in the thermodynamic ground state, the finite-size susceptibility $\chi_O = \langle \delta \hat{O} | \langle \delta \hat{O} \rangle \rangle / \langle \delta \hat{O} \rangle / N J_1$ has to diverge with the system size. In particular, it can be shown that it is bounded from below by the system volume squared, $\chi_O > \text{const} \times p^4 N^2$, where $p = \sqrt{\langle \psi_0 | \hat{O}^2 | \psi_0 \rangle / N^2}$ is the order parameter. A spontaneously broken translation symmetry can be detected by studying the response of the system to the perturbation $\delta \hat{O}_T$, with

$$\hat{O}_T = \sum_j e^{iQ \cdot R_j} \hat{S}_j \cdot \hat{S}_{j+x},$$

where $R_j$ indicates the coordinates of the site $j$ and $x = (1,0)$. This operator preserves the SU(2) symmetry of the $J_1-J_2$ Hamiltonian but breaks the translational invariance with momentum $Q = (\pi,0)$ and the $x \leftrightarrow y$ symmetry (or equivalently the $\pi/2$ rotation symmetry).

By using a numerical technique, like the Lanczos method, the susceptibility $\chi_T = \langle \delta \hat{O}_T | \langle \delta \hat{O}_T \rangle \rangle / \langle \delta \hat{O}_T \rangle / N J_1$ can be calculated with only energy measurements by computing the ground-state energy per site (in unit of $J_1$) in presence of the perturbation, $e(\delta)$, for few values of $\delta$ and by estimating numerically the limit $\chi_T = \lim_{\delta \to 0} \chi_T(\delta) = -2(\delta e(\delta) - e_0) / \delta^2$. Notice that, in the presence of the perturbation, Eq. (2), the Lanczos calculation is rather demanding. In fact, the symmetrized Hilbert space is increased by a factor of four with respect of the standard calculation on the $J_1-J_2$ model, and the resulting dimension of the ground-state subspace is 63.117.760 for $N = 6 \times 6$.

In Fig. 1 we present the ED results for the susceptibility associated to the operator $\hat{O}_T$. For comparison, the data for the one-dimensional $J_1-J_2$ model are also shown. In the latter case, it is well known that a transition form a quasi-ordered state, with power-law spin-spin correlation functions, to a dimerized phase occurs at $J_2/J_1 \approx 0.241$. The response of the system to the perturbation is very different and above the critical point [Fig. 1(a)-(b)]. This is particularly evident by performing the size-scaling of $\chi_T / N$ which diverges for $J_2/J_1 = 0.4$, while saturates to a constant for $J_2/J_1 = 0.1$ consistently with the known scaling dimension of the spin-Peierls perturbation in the Luttinger regime, $X = 1/2$ [Fig. 1(b)]. In contrast in the two-dimensional case, for both $J_2/J_1 = 0.5$ and $J_2/J_1 = 0.55$, $\chi_T / N$ decreases by increasing the lattice size [Fig. 1(c)-(d)] suggesting that a divergence of this quantity in the thermodynamic limit is unlikely, even if not ruled out by our small-size calculations. Indeed, Sushkov et al. found a divergence of the same susceptibility $\chi_T$, by using a series expansion, that are free from size effects but are strongly biased by the approximate approach: the limited number of coefficients known in the infinite series and the difficulty of reconstructing a possible translational invariant state starting from a dimerized reference one. Note that two-dimensional systems behave very differently from chains of comparable linear size, where $\chi_T / N$ always increases with $N$ in the dimerized phase [Fig. 1(a)-(b)]. This points toward the absence of dimer order parameter in the strong frustration regime.

In order to investigate the possible occurrence of a spin-Peierls phase we now analyze the dimer-dimer corre-

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**FIG. 1:** ED calculation of the susceptibility associated to the operator $\hat{O}_T$ of Eq. (2). Left panels: results for the one-dimensional chain. Bottom: $\chi_T(\delta)/N$ vs $\delta^2$ for (from below) 4, 6, and 8 sites. $J_2/J_1 = 0.1$ (empty symbols), 0.4 (full symbols). Top: size scaling of $\chi_T/N$. Right panels: same quantities for the two-dimensional case for $J_2/J_1 = 0.5$ (empty symbols) and 0.55 (full symbols).

**FIG. 2:** ED results of the dimer-dimer correlation functions for the 32-site one-dimensional chain with $J_2/J_1 = 0.1$ (empty dots), 0.4 (full dots), and 0.5 (stars). Notice however that at the exactly solvable point $J_2/J_1 = 0.5$, $|\Delta_{x+x}(r)| = 1/64$, independent on the distance for any non-overlapping singlets.
FIG. 3: ED calculation of the dimer-dimer correlations among pairs parallel to the y axis as a function of the Manhattan distance \(d\) for \(N = 6 \times 6\). \(J_2/J_1 = 0.2\) (empty dots), 0.45 (empty squares), 0.55 (empty triangles), 0.6 (full triangles), 0.65 (full squares), and 10 (full dots). The stars (crosses) indicate the results for the lowest \(d\)-wave (s-wave) excited state for \(J_2/J_1 = 0.65\) (\(J_2/J_1 = 10\)). The large circles in the top panel are the values for \(J_1 = 0\).

Correlation functions of the unperturbed (\(\delta = 0\)) ground state:

\[
\Delta_{\mu,\nu}(r - r') = \langle \hat{S}_r^z \hat{S}_{r + \mu}^z \hat{S}_{r + \nu}^z \hat{S}_{r + \mu + \nu}^z \rangle - \langle \hat{S}_r^z \hat{S}_{r + \mu}^z \rangle \langle \hat{S}_{r + \nu}^z \hat{S}_{r + \mu + \nu}^z \rangle,
\]

where \(\mu\) and \(\nu\) are two unit vectors connecting the nearest-neighbor sites on the lattice. In presence of a broken spatial symmetry, the latter has to converge to a finite value for large distance, displaying also the typical staggered pattern shown in Fig. 4 for the one-dimensional model. In this case, the different decay of the dimer correlations below and above the transition can be also recognized: oscillatory power-law in the Luttinger regime and constant amplitude oscillations in the dimerized phase. In Fig. 4 we present a systematic study of the dimer-dimer correlations on the \(6 \times 6\) cluster as a function of \(J_2/J_1\). The lower panel is for \(J_2/J_1 = 0.2\) (AF Néel phase), the middle one is for \(0.5 \leq J_2/J_1 \leq 0.65\) and the panel on the top is for \(J_2/J_1 = 10\) (AF collinear phase).

The dimer correlations do not seem to be much affected entering the non-magnetic region (\(J_2/J_1 \geq 0.4\)) and the large distance value of the correlations for \(J_2/J_1 \approx 0.5\) is comparable to the one in the AF Néel phase, where the dimer correlations decay to zero with a power law. By increasing further the frustration, the dimer-dimer correlations increase reaching a maximum around \(J_2/J_1 \approx 0.65\) and then converging for large \(J_2/J_1\) to a non-zero value, which is positive for parallel dimers and negative for orthogonal ones (not shown). Notice, however, that for large enough frustration, \(\Delta_{y,y}(r)\) does not show a staggered behavior, which is inconsistent with the pattern predicted on the basis of a standard spin-Peierls state. On the other hand, the broken translation symmetry of the collinear phase, also expected in the large-\(J_2\) region, cannot be detected by the dimer-dimer correlation functions plotted in Fig. 4. Instead, the fact that \(\Delta_{\mu,\nu}(r)\) has different signs for parallel and orthogonal dimers is the signature of the breaking of the \(\pi/2\) rotation symmetry, which also characterizes the AF collinear phase. In fact, for \(J_2 \gg J_1\), the system decouples in two unfrustrated Heisenberg models and \(\Delta_{\mu,\nu}(r - r') \approx \langle \hat{S}_r^z \hat{S}_{r + \mu}^z \rangle \langle \hat{S}_{r + \nu}^z \hat{S}_{r + \mu + \nu}^z \rangle\) with the two pairs belonging to opposite sublattices and \(\langle \hat{S}_r^z \hat{S}_{r + \nu}^z \rangle\) being the ground-state spin correlations of the Heisenberg model on a \(N/2\)-site cluster (see the top panel of Fig. 5). Hence, in this limit, \(\Delta_{\mu,\nu}(r - r') \approx s_{\mu,\nu} |\langle \hat{S}_r^z \hat{S}_{r + \nu}^z \rangle |\langle \hat{S}_{r + \mu}^z \hat{S}_{r + \mu + \nu}^z \rangle|\) with \(s_{\mu,\nu} = 1\) and \(-1\) for parallel and orthogonal dimers, respectively. Interestingly enough, for \(J_2 \gg J_1\), the large distance value of the dimer correlations will approach in the thermodynamic limit \(\Delta_{\mu,\nu}(r) \approx (m_1)^4/9\), where \(m_1 \approx 0.307\) is the antiferromagnetic order parameter for the spin-half Heisenberg model.

The non-monotonic behavior of the dimer correlations, displaying a maximum around \(J_2/J_1 \approx 0.65\) is likely to be a finite-size effect related to the finite-size gap between the two lowest singlet states. These states cross on the \(6 \times 6\) for \(J_2/J_1 \approx 0.65\), the ground-state symmetry changing from s-wave (below) to \(d\)-wave (above). In fact, as we have checked in the AF collinear phase, the value of the dimer-dimer correlations on the s-wave singlet (crosses in the top panel of Fig. 4) is larger than the value on the \(d\)-wave one, the two values converging to the same only when they become degenerate, i.e., at the level crossing on finite-sizes (stars in the middle panel of Fig. 5) and in all the AF collinear phase in the thermodynamic limit.

Following Ref. 52, it is possible to obtain a finite-size estimate of the dimer order parameter from the long-distance behavior of the dimer-dimer correlations shown in Fig. 5:

\[
D_d^2 = 9 \lim_{|r| \to \infty} |\Delta_{y,y}(r - y) - 2\Delta_{y,y}(r) + \Delta_{y,y}(r + y)|,
\]

where the factor 9 is required to take into account the three spin component of the order parameter. The behavior of \(D_d\) as a function of \(J_2/J_1\) is reported in Fig. 5(a) for the \(4 \times 4\) and \(6 \times 6\) lattices. A remarkable decrease can be appreciated by increasing the lattice size, even in the strongly frustrated regime. Moreover, the maximum order parameter on the \(6 \times 6\) lattice is considerably smaller than the corresponding one for the dimerized chain, i.e., \(D_d = 0.75\) for the Majumdar-Ghosh case (\(J_2/J_1 = 0.5\)). These facts should suggest that a sizable dimer order parameter does not survive in the thermodynamic limit, leaving the possibility of a disordered spin liquid (or, at most, of a ground state with a very small dimer order).

Finally, in analogy with the definition of \(D_d\), it is possible to define a dimer order parameter related to a ro-
tational symmetry breaking:

\[ D_o^2 = 18 \lim_{|r| \to \infty} |\langle \Delta_{y,y}(r) \rangle - \langle \Delta_{y,x}(r) \rangle| . \]

Again, the behavior of \( D_o \) as a function of \( J_2/J_1 \) is reported in Fig. 4(b) for the \( 4 \times 4 \) and \( 6 \times 6 \) lattices. A very sharp increase of \( D_o \) around \( J_2/J_1 \sim 0.6 \) confirms that a rotational symmetry breaking is plausible for large frustration, where the collinear phase is settled down.

For smaller values of \( J_2/J_1 \), instead, the ground state is rotational invariant, as previously found.

In conclusion, we have presented an exact diagonalization study of dimer susceptibility and dimer correlations of the spin-half \( J_1-J_2 \) model. Although we are aware that important finite-size effects occur near phase transitions, our analysis does not provide evidence in favor of a spin-Peierls ground state. By contrast, a homogeneous spin-liquid ground state may be realized in the regime of strong frustration. This possibility has been recently supported by a variational calculation showing that a spin-liquid projected BCS wave function provides an almost exact variational ansatz of the ground-state in the non-magnetic phase of this frustrated system. Work is in progress to corroborate these conclusions and to clarify the nature of the excitation spectrum of this unconventional state of matter.

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1. S. Sachdev, Science 288, 475 (2000).
2. P. Chandra and B. Doucot, Phys. Rev. B 38, 9335 (1988).
3. V. N. Kotov, J. Oitmaa, O.P. Sushkov, and Z. Weihong, Phil. Mag. B 80, 1483 (2000).
4. P. Fazekas and P.W. Anderson, Philos. Mag. 30, 423 (1974).
5. F. Figueiredo, A. Karlhede, S. Kivelson, S. Sondhi, M. Rocek, and D.S. Rokhsar, Phys. Rev. B 41, 4619 (1989).
6. R.P. Singh, Z. Weihong, C.J. Hamer, and J. Oitmaa, Phys. Rev. B 60, 7278 (1999).
7. O.P. Sushkov, J. Oitmaa, and Z. Weihong, Phys. Rev. B 63, 104420 (2001); Phys. Rev. B 66, 054401 (2002).
8. M.S.L. du Croo de Jongh, J.M.J. van Leeuwen, and W. Saarloos, Phys. Rev. B 62, 14844 (2000).
9. N. Read and S. Sachdev, Phys. Rev. B 8, 456 (2000).
10. P. Carretta, R. Melzi, N. Papinutto, and P. Millet, Phys. Rev. Lett. 88, 47601 (2002).
11. L. Capriotti and S. Sorella, Phys. Rev. Lett. 84, 3173 (2000).
12. H.J. Schulz, T.A.L. Ziman, and D. Poilblanc, J. Phys. I (France) 6, 675 (1996).
13. D. Poilblanc, E. Gagliano, S. Bacci, and R. Dagotto, Phys. Rev. B 43, 10970 (1991).
14. G. Castilla, S. Chakravarty, and V.J. Emery, Phys. Rev. Lett. 75, 631 (1995), and references therein.
15. G. Santoro, S. Sorella, L. Guidoni, A. Parola, and E. Tosatti, Phys. Rev. Lett. 83, 3065 (1999).
16. L. Capriotti, Int. J. Mod. Phys. B 15, 1799 (2001).
17. The scaling dimension \( X \) of an operator is related to its susceptibility by the relation \( \chi \propto N^{2(1-X)} \).
18. We have verified that the spin-isotropic correlation functions, \( \langle \mathbf{S}_r \cdot \mathbf{S}_{r+\mu} \rangle \) and \( \langle \mathbf{S}_r \cdot \mathbf{S}_{r+\mu} \rangle \), give the same qualitative behavior.
19. The broken translation symmetry of the collinear phase is clearly shown by the peaks of the magnetic structure factor at \( Q = (\pi,0) \) and \( Q = (0,\pi) \).
20. M. Calandra and S. Sorella, Phys. Rev. B 57, 11446 (1998).
21. S.R. White and I. Affleck, Phys. Rev. B 54, 9862 (1996).
22. L. Capriotti, F. Becca, A. Parola, and S. Sorella, Phys. Rev. Lett. 87, 097201 (2001).