Spontaneous Plaquette Dimerization in the J_1–J_2 Heisenberg Model

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We investigate the nonmagnetic phase of the spin-half frustrated Heisenberg antiferromagnet on the square lattice using exact diagonalization (up to 36 sites) and quantum Monte Carlo techniques (up to 144 sites). The spin gap and the susceptibilities for the most important crystal symmetry breaking operators are computed. A genuine and somehow unexpected "plaquette resonating valence bond," with spontaneously broken translation symmetry and no broken rotation symmetry, comes out from our numerical simulations as the most plausible ground state for $J_2/J_1 \simeq 0.5$.

The nature of the nonmagnetic phases of a quantum antiferromagnet is a topic of great interest and has been a subject of intense theoretical investigation since Anderson's suggestion [1] about the possible connections with the mechanism of high-T_c superconductivity.

Within the Heisenberg model the simplest way in which the antiferromagnetism can be destabilized is by introducing a next-nearest-neighbor frustrating interaction leading to the so-called J_1–J_2 Hamiltonian

$$\hat{H} = J_1 \sum_{\text{n.n.}} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + J_2 \sum_{\text{n.n.n.}} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j, \qquad (1)$$

where $\hat{\mathbf{S}}_i = (\hat{S}_i^x, \hat{S}_i^y, \hat{S}_i^z)$ are spin-$\frac{1}{2}$ operators on a square lattice. J_1 and J_2 are the (positive) antiferromagnetic superexchange couplings between nearest and next-nearest-neighbor pairs of spins, respectively. In the following we will consider finite clusters of N sites with periodic boundary conditions (tilted by 45° only for $N = 32$).

Although there is a general consensus about the disappearance of the Néel order in the ground state (GS) of the present model for $0.38 \lesssim J_2/J_1 \lesssim 0.60$ [2–4], no definite conclusion has been drawn on the nature of the nonmagnetic phase yet. In particular, an open question is whether the GS of the J_1–J_2 Heisenberg model is a resonating valence bond (RVB) spin liquid with no broken symmetries, as it was originally suggested by Figueirido *et al.* [5]. The other possibility is a GS which is still SU(2) invariant, but nonetheless breaks some crystal symmetries, dimerizing in some special pattern [6–11].

In this Letter we address this point using exact diagonalization (ED) and a quantum Monte Carlo technique, the Green function Monte Carlo (GFMC), which allows the calculation of GS expectation values on fairly large system sizes ($N \leq 144$). This is extremely important to draw reasonable conclusions on the physical thermodynamic, zero temperature properties of the model.

For frustrated spin systems as well as for fermionic models, quantum Monte Carlo methods are affected by the so-called *sign problem* that can be controlled, at present, only at the price of introducing some kind of approxima-

tions, such as the fixed node (FN) one [12]. In this work we have also extensively used a recently developed technique, the Green function Monte Carlo with stochastic reconfiguration (GFMCSR), which improves systematically the accuracy of the FN approximation for GS calculations [4,13–15].

The FN method allows us to work without any sign problem by using the following simple strategy: The exact imaginary time propagator $e^{-\tau \hat{H}}$—used to filter out the GS from the best variational guess $|\psi_G\rangle$—is replaced by an approximate propagator $e^{-\tau \hat{H}_{\text{FN}}}$ such that the nodes of the propagated state $e^{-\tau \hat{H}_{\text{FN}}}|\psi_G\rangle$ do not change, due to an appropriate choice of the effective FN Hamiltonian $\hat{H}_{\text{FN}}$ (which in turn depends on $|\psi_G\rangle$). The FN approximation becomes exact if the so-called *guiding* wave function $|\psi_G\rangle$ is the exact GS. However, for frustrated spin models even the best variational wave function of the Jastrow type [4], used to guide the FN dynamic, provides rather poor results even for the GS energy expectation value [4,13,14].

The GFMCSR method allows us to release the FN approximation and to obtain results much less depending on the quality of the guiding wave function. During each short imaginary time evolution $\tau \to \tau + \Delta\tau$, where both the exact and the approximate propagations can be performed without sign problem instabilities, the FN dynamic is systematically improved by requiring that a given number p of *mixed averages* [13] of correlation functions is propagated consistently with the exact dynamic. By increasing the number of correlation functions one typically improves the accuracy of the calculation since the method becomes exact if all the independent correlation functions are included in the stochastic reconfiguration (SR) scheme.

Typically [4,14], few correlation functions ($p \sim 10$) allow us to obtain rather accurate values of the GS energy with an error much less than 1%—for lattice sizes ($N \leq 36$) where the exact solution is available numerically—and without a sizable loss of accuracy with increasing size. Such accuracy is usually enough to reproduce some physical features that are not contained at the variational level, as has been shown in a previous study of the present model [4]. In the latter work, in fact, with a gapless

guiding wave function, it has been possible to detect a finite spin gap in the thermodynamic limit for $J_2/J_1 \gtrsim 0.4$.

We have extended the previous GFMCSR calculation, with the same guiding wave function of Ref. [4], by including in SR conditions not only the energy and all $\hat{S}_i^z \hat{S}_j^z$ independent by symmetry, but also the antiferromagnetic order parameter. The latter, as discussed in Ref. [13], though not improving the accuracy of the calculation, allows a very stable and reliable simulation for large p. The new results, extended up to $N = 144$, confirm the previous findings of a finite spin gap for $J_2/J_1 \gtrsim 0.40$ (Fig. 1).

As suggested in Refs. [8,9,11,17], in order to investigate the possible occurrence of a spontaneously dimerized GS displaying some kind of crystalline order, we have calculated the response of the system to operators breaking the most important lattice symmetries. This can be done by adding to the Hamiltonian (1) a term $-\delta \hat{O}$, where $\hat{O}$ is an operator that breaks some symmetry of $\hat{H}$. On a finite size, the GS expectation value of $\hat{O}$ vanishes by symmetry for $\delta = 0$, and the GS energy per site has corrections proportional to δ^2 as by the Hellmann-Feynman theorem $-de(\delta)/d\delta = \langle \hat{O} \rangle_\delta / N$. Therefore $e(\delta) \simeq e_0 - \chi \delta^2/2$, being χ the generalized susceptibility associated to the operator $\hat{O}$, namely, $\chi = 2\langle \psi_0 | \hat{O}(E_0 - \hat{H})^{-1} \hat{O} | \psi_0 \rangle / NJ_1$. For $N \to \infty$, if true long range order (LRO) exists in the thermodynamic GS, an infinitesimal field $\delta \sim 1/N$ must give a finite $\langle \hat{O} \rangle_\delta / N \sim \chi \delta$ implying that the finite size susceptibility $\chi = \langle \hat{O} \rangle_\delta / \delta N$ has to diverge with the system size [18]. Thus susceptibilities are a very sensitive tool for detecting the occurrence of LRO.

We have considered the response of the system to the following symmetry breaking operators:

$$\hat{O}_C = \sum_i (\hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+x} - \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+y}), \qquad (2)$$

$$\hat{O}_P = \sum_i e^{i\mathbf{Q}_0 \cdot \mathbf{r}_i} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+x}, \qquad (3)$$

with $x = (1,0)$, $y = (0,1)$, and $\mathbf{Q}_0 = (\pi, 0)$, for the rotation and the translation symmetry, respectively. Within ED and GFMC technique the susceptibility $\chi = -d^2 e(\delta)/d\delta^2 |_{\delta=0}$ can be evaluated by computing the GS energy per site in presence of the perturbation for few values of δ, and by estimating numerically the limit $\delta \to 0$ of the quantity $\chi(\delta) = -2[e(\delta) - e_0]/\delta^2$.

As we have tested in the one dimensional $J_1 - J_2$ model, the numerical study of LRO by means of $\chi(\delta)$ is very effective and reliable. Here a quantum critical point at $J_2/J_1 \simeq 0.2412$ separating a gapless spin fluid phase from a gapped dimerized GS, which is twofold degenerate, is rather well accepted [19–21]. As shown in Fig. 2, the response of the system to the perturbation $\delta \hat{O}_P$ [Eq. (3)], breaking the translation invariance with momentum $k = \pi$, is very different below and above the dimer-fluid transition point. However, it is extremely important to perform very accurate calculations at small δ to detect the divergence of the susceptibilities for large system sizes.

In two dimensions, among the dimerized phases proposed in the literature, the so-called *columnar* and *plaquette* RVB [6–11] are the states which have obtained the most convincing numerical evidences. Both the columnar and plaquette RVB break the translation invariance but only the latter preserves the rotation symmetry. As also suggested in a recent paper by Singh *et al.* [11], the appearance of a columnar state can be tested by using as order parameter the operator $\hat{O}_C$ defined in Eq. (2). As shown in Fig. 3, the ED results for $N = 16$ and $N = 36$ indicate that the susceptibility associated with this kind of symmetry breaking, χ_C, decreases with the system size. Using the GFMCSR, described before, we have extended the calculation up to $N = 64$. The GFMCSR calculations, which reproduce pretty well the ED data, rule out clearly the columnar dimer order.

The above result is in disagreement with the conclusions of several series expansion studies [7,10,11]. However, as stated in Ref. [11], the series for χ_C is very irregular
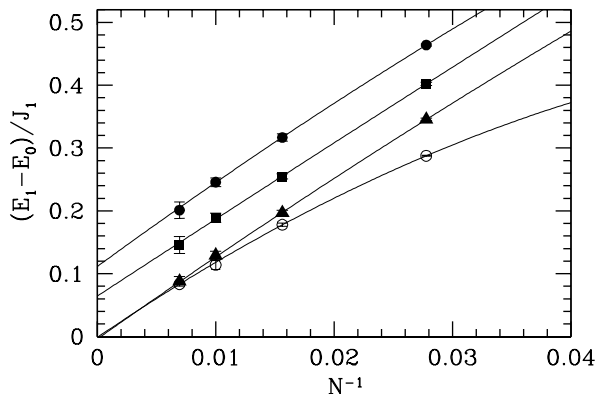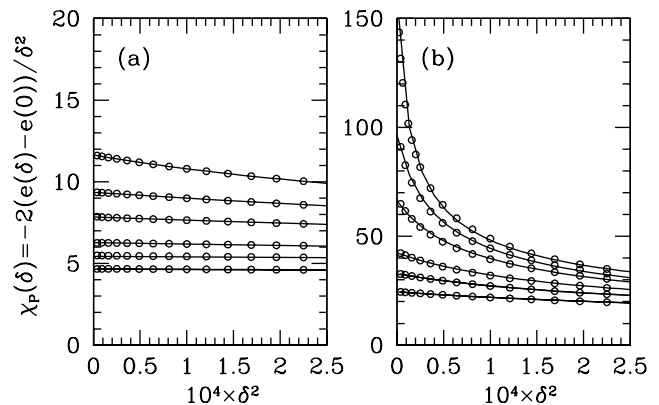

FIG. 1. Size scaling of the energy gap to the first $S = 1$ spin excitation obtained with the GFMCSR technique for $J_2/J_1 = 0.38$ (full triangles), 0.45 (full squares), and 0.50 (full circles). Data for the unfrustrated ($J_2 = 0$) Heisenberg model taken from Ref. [16] are also shown for comparison (empty circles). Lines are weighted quadratic fits of the data.

FIG. 2. ED results for the $J_1 - J_2$ chain: $\chi_P(\delta)$ associated with the operator $\hat{O}_P$ (breaking the translational invariance) for $J_2/J_1 = 0.2$ (a) and $J_2/J_1 = 0.4$ (b). Data are shown for $N = 12, 14, 16, 20, 24,$ and 30 for increasing values of $\chi_P(\delta)$.
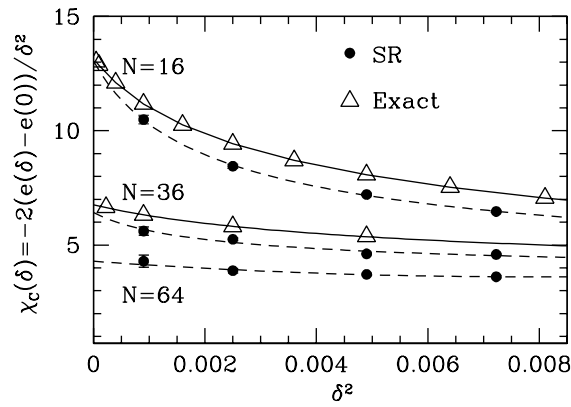
FIG. 3.   Exact and GFMCSR calculations of $\chi_C(\delta)$ associated with $\hat{O}_C$ (columnar dimerization) for $J_2/J_1 = 0.5$.

and does not allow a meaningful extrapolation to the exact result. In our calculation instead, even the ED results for $N \leq 36$ are already conclusive.

Having established that the columnar susceptibility is bounded, it is now important to study the response of the J_1-J_2 model to a small field coupled to the perturbation $\hat{O}_P$ of Eq. (3), breaking the translation invariance of the Hamiltonian. The evaluation of χ_P, with a reasonable accuracy, is a much more difficult task. In fact, in this case the ED values of the susceptibility for $N = 16$ and $N = 32$ increase with the size, and much more effort is then required to distinguish if this behavior corresponds to a spontaneous symmetry breaking in the thermodynamic limit. As is shown in Fig. 4(a), the FN technique, starting from a guiding wave function without dimer order, is not able to reproduce the actual response of the system to $\hat{O}_P$, even on small sizes. The GFMCSR technique allows us to get an estimate of the susceptibility which is a factor of 3 more accurate, but not satisfactory enough. In order to improve this estimate, we have attempted to include in the SR conditions many other, reasonably simple, correlation functions (such as the spin-spin correlation func-
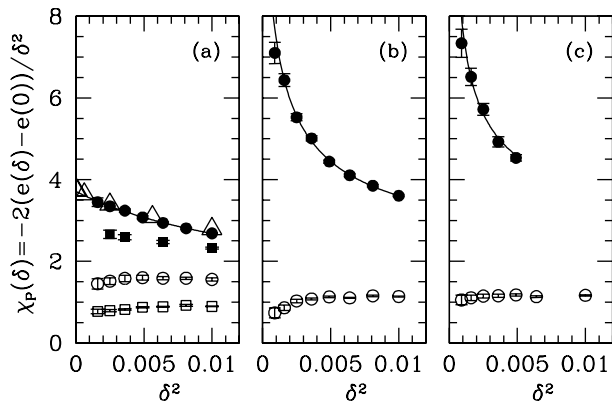
tions $\hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j$ for $|r_i - r_j| > \sqrt{2}$), but without obtaining a sizable change of the estimate of χ_P. In fact, the most effective SR conditions are those obtained with operators more directly related to the Hamiltonian [13,14].

After many unsuccessful attempts, we have realized that it is much simpler and straightforward to improve the accuracy of the guiding wave function itself. In fact, it is reasonable to expect that both the FN and the GFMCSR will perform more efficiently with a better $|\psi_G\rangle$, i.e., with an improved initial guess of the GS wave function. This can be obtained by applying a generalized Lanczos operator $(1 + \alpha\hat{H})$ to the variational wave function $|\psi_G\rangle$, where α is a variational parameter. This defines the so-called *one Lanczos step* (LS) wave function, which has been particularly successful for the *t-J* model [22].

In the present model by using the LS wave function, a clear improvement on the variational estimate of the GS energy is obtained. More importantly, as shown in Fig. 4(a), the LS wave function allows a much better estimate of the susceptibility. Remarkably, on all the finite sizes where ED is possible, the GFMCSR estimate of this important quantity is basically exact within a few error bars (see also Fig. 5). This calculation was obtained by including in the SR conditions the energy, the spin-spin correlation functions up to next-nearest-neighbors, distinguishing also $\hat{S}_i^z\hat{S}_j^z$ and $(\hat{S}_i^x\hat{S}_j^x + \hat{S}_i^y\hat{S}_j^y)$ ($p = 4$). The mixed averages of these correlation functions can be computed over both the wave function $|\psi_G\rangle$ and the LS wave function $(1 + \alpha\hat{H})|\psi_G\rangle$ during the same Monte Carlo simulation. Thus with a LS wave function one can also easily double the number of constraints that are effective to improve the accuracy of the method ($p = 8$). In this case we have tested that it is irrelevant to add further long range correlation functions in the SR conditions even for large size.

FIG. 4. $\chi_P(\delta)$ associated to $\hat{O}_P$ (plaquette dimerization) for $J_2/J_1 = 0.5$, $N = 32$ (a), $N = 64$ (b), and $N = 100$ (c). FN (empty squares), GFMCSR (full squares), FN with LS (empty circles), GFMCSR with LS (full circles), and exact (empty triangles).
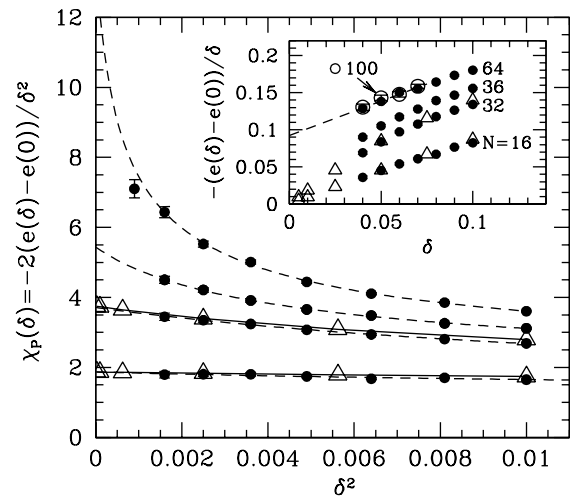

FIG. 5.   Exact (empty triangles) and GFMCSR (circles) calculations of $\chi_P(\delta)$ (plaquette dimerization) for $J_2/J_1 = 0.5$ and (from the bottom) $N = 16$, 32, 36, and 64. Inset: numerical determination of the order parameter (see text). Lines are guides for the eye.

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By increasing the size, the response of the system is very strongly enhanced, in very close analogy to the one dimensional model in the dimerized phase [see Fig. 2(b)]. This is obtained only with the GFMCSR technique, since as shown in Fig. 4, the combination of FN and Lanczos step alone is not capable to detect these strongly enhanced correlations. For $N = 100$ the GFMCSR increases by more than 1 order of magnitude the response of the system to the dimerizing field. This effect is particularly striking, considering that the starting guiding wave function is spin wave like [23], i.e., gapless, Néel ordered, and without any dimer LRO. This suggests that all our systematic approximations are able to remove almost completely even a very strong bias present at the variational level.

We believe that the numerical results we have presented here give a very robust indication of a spontaneous dimerization with broken translation symmetry but without broken rotation symmetry (as discussed before), i.e., a plaquette RVB. This kind of state can be thought of as a collection of rotation invariant valence bond states

$$\left| \begin{array}{c} \square \end{array} \right\rangle = \left| \begin{array}{c} \end{array} \right\rangle + \left| \begin{array}{c} \end{array} \right\rangle ,$$

where $|\circ\!\!-\!\!\circ\rangle = |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$. Such plaquettes cover only one-half of the possible elementary plaquettes of the lattice since two plaquettes cannot have a common side. In this way one necessarily has to break translation invariance and the resulting GS is fourfold degenerate in the thermodynamic limit, in agreement with the Haldane's hedgehog argument described in Ref. [24].

In the past, among several attempts to guess the nature of the nonmagnetic phase of this model, the description closest to ours was that proposed by Zithomirski and Ueda [9]. Amazingly, part of their conclusions were based on an unfortunate mistake in the series expansion [11].

The quantitative estimate of the order parameter can be obtained by taking *first* the thermodynamic limit $N \rightarrow \infty$ of the order parameter $O_P(\delta) = \langle \hat{O} \rangle_\delta / N$ at fixed field δ, and then letting $\delta \rightarrow 0$, $\lim_{\delta \rightarrow 0} O_P(\delta) = O_P$ being the value of the order parameter. In order to estimate $O_P(\delta)$ at fixed size we have used the Hellmann-Feynmann theorem with a finite difference estimate of $-de(\delta)/d\delta \sim [e(0) - e(\delta)]/\delta$. As shown in the inset of Fig. 5, the finite size effects of this quantity seem to saturate above the $N = 64$ lattice size for $\delta \geq 0.04$, allowing a rather convincing estimate of the dimer order parameter as $O_P \sim 0.1$, being

sizably nonzero. The sharp crossover of the size effects for $N \geq 64$ is due to the presence of a finite triplet gap in the excitation spectrum (Fig. 1), implying, typically, a finite characteristic length. The value of the order parameter O_P is rather large considering that $J_2/J_1 = 0.5$ is very close to the transition point for the onset of spontaneous dimerization $J_2/J_1 \simeq 0.40$. This is an interesting and measurable physical property that can be, in principle, investigated experimentally.

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