Signature of Néel Order in Exact Spectra of Quantum Antiferromagnets on Finite Lattices

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We show how the broken symmetries of the Néel state are embodied in the exact spectrum of the triangular Heisenberg antiferromagnet on finite lattices as small as $N = 21$ (spectra up to $N = 36$ have been computed). We present the first numerical evidence of an extensive set of low-lying levels that are below the softest magnons and collapse to the ground state in the thermodynamic limit. This set of quantum states represents the quantum counterpart of the classical Néel ground state. We develop an approach relying on the symmetry analysis and finite-size scaling and we provide new arguments in favor of an ordered ground state for the $S = \frac{1}{2}$ triangular Heisenberg model.

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In the last decades, a large amount of work has been devoted to the understanding of the quantum ground state of two-dimensional antiferromagnets. In the early seventies, Anderson launched a debate on the possible existence of a "resonating valence bond" (RVB) state which could represent an alternative to the Néel antiferromagnetic state [1]. The first candidate to be considered was the spin-$\frac{1}{2}$ Heisenberg antiferromagnet on the triangular lattice:

$$H = \sum_{\langle i,j\rangle} 2\mathbf{S}_i \cdot \mathbf{S}_j \,, \tag{1}$$

where the sum runs over first neighbor pairs. A variational RVB state was proposed to be more stable than the Néel state [2]. From the spin-wave analysis, it was later concluded that quantum fluctuations were insufficient to destabilize Néel's classical state [3]; perturbative approaches led to the same conclusion and variational ones did not weaken it [4, 5]. However, exact results of diagonalization on small periodic samples up to $N = 27$ were extrapolated and gave the opposite result [6, 7]. But in the above numerical studies, the spin-liquid hypothesis was not really explored, nor was the Néel long-range order (NLRO) assumption convincingly discarded.

Usually NLRO is checked on the finite-size scaling of the ground-state energy and magnetization [8, 9]. The magnon dispersion relation being linear in k, the leading finite-size correction to the ground-state energy per particle E_∞ is $\mathcal{O}(N^{-3/2})$ and that for the magnetization modulus per particle M_∞ is $\mathcal{O}(N^{-1/2})$ (M_∞ is defined in [10]). Figure 1 shows the values of E_N and M_N for small periodic samples. We present the results for the first calculation of the $N = 36$ sample, a calculation made possible by using all the symmetries of the Hamiltonian and the lattice. We find $\langle 2\mathbf{S}_i \cdot \mathbf{S}_j\rangle_{36} = -0.373\,582\,3(1)$ and $M_{36} = 0.400\,575(1)$. From the values, it is clear that the magnetization modulus does not extrapolate to zero in the $N \to \infty$ limit; but, it is difficult to assert that the finite-size scaling of the ground-state energy behaves as $N^{-3/2}$. Therefore, no definite conclusion can be drawn from the ground-state evaluations on small samples.

In this Letter we show how the hypothesis of NLRO implies a list of drastic conditions on the symmetries, dynamics, and the finite-size scaling of an extensive $[\mathcal{O}(N^{3/2})]$ set of low-lying levels of the spectrum. Some of these conditions are new, others go back to Anderson's seminal paper on antiferromagnets [11] or have been discussed since then [8, 9]. We shall see that this *complete* list of conditions, which determines all the quantum numbers of the $N^{3/2}$ lowest-lying levels of Eq. (1), constitutes a necessary and sufficient condition for an *à la Néel* sym-

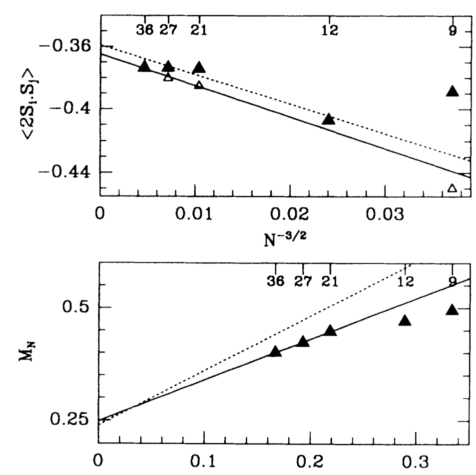

FIG. 1. Finite-size scaling analysis. Top: Energies E_N vs $N^{-3/2}$; ▲, exact diagonalization values; - - -, spin-wave results (note that the energies for $N = 21, 27, 36$ do not seem to obey a reasonable finite-size scaling law); △, energies after rotational correction (see text) for odd N; —, extrapolation of the "corrected results" for $N = 21, 27, 36$. Bottom: magnetization modulus M_N vs $N^{-1/2}$; ▲, exact diagonalization results; - - -, spin-wave results; —, extrapolation of the diagonalization results for $N = 21, 27, 36$.

metry breaking. The success of this scheme, illustrated here on the triangular Heisenberg case, clearly depends on the ability to determine all quantum numbers (symmetries) and the degeneracy of a large number of states $[\mathcal{O}(N^{5/2})]$, which implies to calculate the energies in each irreducible representation of the symmetry group of the system. We shall now deduce these five conditions from the analysis of the nature of a Néel semiclassical ground state.

In a Néel antiferromagnet, the magnetization of each sublattice is a *macroscopic vector* which breaks the SU_2 symmetry. The previous numerical calculations checked the modulus of this vector in the ground state of the Hamiltonian (an $S = 0$ or $S = \frac{1}{2}$ eigenstate depending on the even or odd value of N), but this is insufficient to insure the vectorial character of the order parameter. Remember that in an $S = 0$ state, all vectorial observables are zero (Wigner-Eckart theorem). In order to fix a macroscopic vectorial order parameter along the direction (Ω) with an uncertainty $\mathcal{O}(\varepsilon)$, it is necessary to form a wave packet of eigenstates with S values up to $\mathcal{O}(1/\varepsilon)$. The quantum counterpart of a classical Néel state, either a ground state or a magnon excitation, is necessarily a coherent superposition of an *extensive* set of eigenstates of Eq. (1) reading

$$|\text{Néel}(\Omega)\rangle = \sum_{i,S,m_S} \alpha(i,S) Y_S^{m_S}(\Omega)|i,S,m_S\rangle \;, \qquad (2)$$

where the total spin S, its z component m_S, and the spatial-symmetry type i are the labels describing the eigenstates of Eq. (1) [12]. Such a state is called a "macroscopic" state in the following.

It is clear that in the NLRO hypothesis the sublattice-magnetization modulus must be macroscopic in each of the $|i,S,m_S\rangle$ states participating in Eq. (2).

On a finite lattice, these $|i,S,m_S\rangle$ levels are nondegenerate and the sublattice magnetization in such a state $\langle\text{Néel}|\mathbf{S}_{\mathbf{k}_0}|\text{Néel}\rangle$, where $\mathbf{S}_{\mathbf{k}_0}$ is defined in [10], evolves at various values of $E_i - E_j$. Let us call ΔE the largest value of these differences. In the NLRO hypothesis, the sublattice magnetization is the slow variable of the problem and in the thermodynamic limit the energy scale ΔE must go to zero faster than the frequency of the softest mode of the system, i.e., faster than $N^{-1/2}$. Therefore, for large enough N, the energy spectrum must exhibit well separated extensive sets of energy levels respectively associated with the degenerate classical ground states and with any magnon excitations, which are the long-wavelength excitations of this slow collective macroscopic variable [13,14].

In the NLRO hypothesis, the purely angular nature of the slow collective variable implies that these sets of energy levels must map onto a rigid rotator model. Thus, we expect the main dynamics of the $|i,S,m_S\rangle$, participating either in the macroscopic ground state or in the magnon excitations, to be described by the centrifugal term $S^2/2I_N^{\perp}$, where $I_N^{\perp}$ scales faster than $N^{1/2}$. Indeed, this is verified on the triangular lattice for all samples considered ($N = 9, 12, 21, 36$). Figures 2(a) and 2(b) illustrate this behavior as well as the separation between the ground-state multiplicity and the magnon multiplicity. In the following, we shall concentrate specifically on the quantum states $|i,S,m_S\rangle$ participating in the ground-state multiplicity: we call them the *quasidegenerate joint* states (QDJ). In the QDJ spectrum, the largest frequency scale ΔE of the magnetization is thus of the order of $S_{\max}/I_N^{\perp}$, where $S_{\max}$ is the maximum value of S in Eq. (2). From numerical results, we find that $I_N^{\perp}$ scales as N (see Fig. 3 and [15]). The natural cutoff of ΔE being of order $\mathcal{O}(N^{-1/2})$, the energy of the softest magnon, the Néel ground state includes QDJ states up to $S = N^{1/2}$. Moreover, this mapping of the Heisenberg problem on a rigid rotator one implies that one must find $(2S+1)^2$ QDJ

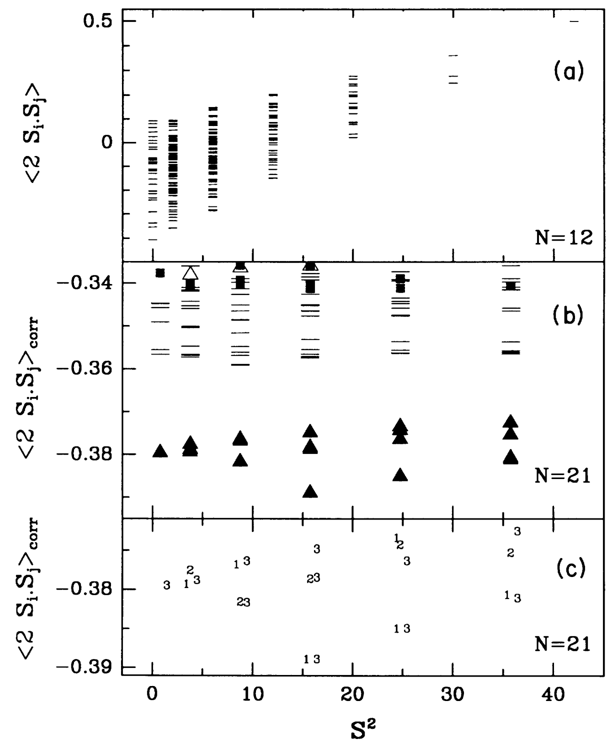

FIG. 2. Energy spectrum of Eq. (1) vs $\mathbf{S}^2 = S(S+1)$. (a) complete spectrum for $N = 12$; (b) partial spectrum for $N = 21$. The effective rotational correction proportional to $S(S+1)/I^{\perp}$ has been subtracted from the raw results. ▲, QDJ states; △, outsiders with the same spatial symmetries as the QDJ states (extra copies, see text); −, magnons ($k \neq 0, \pm k_0$); ■, C_3 rotational noninvariant states ($k = 0, \pm k_0$), the first states that could support chirality (in this system, both the extra copies and the rotationally noninvariant states present a nonvanishing gap with the ground state). (c) Enlargement of (b) for the QDJ states. The numbers 1, 2, and 3 refer to the irreducible representations Γ_1, Γ_2, and Γ_3 and with respective degeneracies $2S+1$, $2S+1$, and $2(2S+1)$.

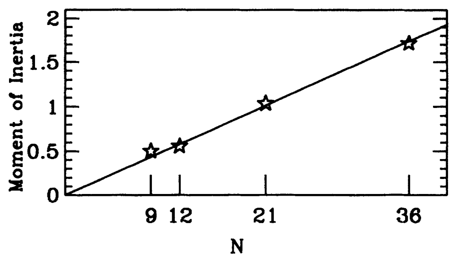

FIG. 3. Moment of inertia, I_N, vs sample size, N. For a given N, I_N is the slope of the straight line obtained by a mean-square fit to the $S(S+1)$ dependence of the QDJ energies (see Fig. 2).

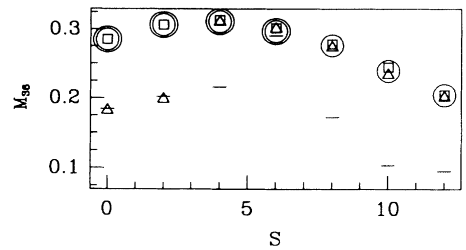

FIG. 4. Order-parameter modulus M_N vs total spin S in the Γ_1-IR for the $N=36$ spin sample. For each value of S, □, △, and − stand for the ground state, first-, and second-excited states. The double-circled values point to QDJ states actually participating to the Néel macroscopic state [Eq. (2)]. The single-circled values point to levels that will in larger samples collaborate to the Néel state. The uncircled results correspond to outsiders belonging to excited states, whatever the size of the sample.

states for each S value. The first factor of $(2S+1)$ stands for the rotational degeneracy of the S state, while the second one comes from the various values of the projection of the external angular momentum on the intrinsic top axes. In the triangular lattice, we expect this rigid rotator to have a cylindrical symmetry in the thermodynamic limit.

The SU_2 symmetry breaking of the Néel antiferromagnet implies thus strong constraints on the low-lying energy spectrum of the Heisenberg Hamiltonian. It requires (i) the existence of an *extensive* set of QDJ levels scaling towards the absolute ground state faster than the first magnon mode [Figs. 2(a) and 2(b)], (ii) exhibiting the dynamics and degeneracy of a top for total spins up to $S=\mathcal{O}(N^{1/2})$ (Figs. 2 and 3), (iii) having the same macroscopic value of the sublattice magnetization or, equivalently, the same value M_N of the order of unity (Fig. 4). *With these first three properties, any combination of the QDJ states with a smooth distribution of α's breaks the $SU(2)$ symmetry in the thermodynamic limit, giving a rigid state with a macroscopic magnetization.*

The last step to totally determine the QDJ states is to specify their i labels, that is the spatial quantum numbers which constitute with S and m_S the complete set of quantum numbers of the problem [12]. The complete analysis is rather technical and will be published elsewhere, and we outline here the main ideas. The symmetry group of the lattice is the semidirect product of the group of translations T_0 (with generators $\vec{u}_1$, $\vec{u}_2$, $\vec{u}_1\cdot\vec{u}_2=-1/2$) by the dihedral group D_6. The symmetry group of the Néel order parameter is the semidirect product of the group of translations on each sublattice T_1 $(2\vec{u}_1+\vec{u}_2, \vec{u}_2-\vec{u}_1)$ by the dihedral group D_3 $(\equiv C_{3v})$. In the NLRO hypothesis, the fourth condition is that the QDJ states must be invariant under C_{3v}. The three irreducible representations (IR) of the spatial group of the lattice which have this invariance are the following: Γ_1, the $k=0$, even under inversion, invariant under C_{3v} IR; Γ_2, the $k=0$, odd under inversion, invariant under C_{3v} IR; Γ_3, the $k=\pm k_0$ IR. The other IRs of this spatial group, which effectively appear higher in the spectrum, are necessarily excluded from the QDJ. The last condition is deduced from the symmetries of the classical Néel state which permute the three sublattices and simultaneously rotate the spins (these symmetries conserve the total spin); for each value of S, the number of replicas of each allowed IR in the QDJ states must be $n_{\Gamma_1}=(a+3b+2c)/6$, $n_{\Gamma_2}=(a-3b+2c)/6$, and $n_{\Gamma_3}=(a-c)/3$, where $a=2S+1$, $b=\cos(S\pi)$, and $c=\sin[\frac{2\pi}{3}(2S+1)]/\sin(\frac{2\pi}{3})$. Extra copies of these IR should not mix with the QDJ states. *These two last conditions, which determine the i label of the QDJ states, are exactly verified in all our samples up to $S\sim\mathcal{O}(\sqrt{N})$* [see Fig 2(c)].

If any of these five criteria are not fulfilled, then one cannot ascertain that the thermodynamic ground state is Néel like: so these criteria are necessary conditions for that kind of symmetry breaking. Reversely if these criteria are fulfilled, the structure factor of the system will have the precise features which are observed in Néel antiferromagnets.

Now, one can achieve the finite-size scaling analysis of E_N and M_N. Note that for odd N, we have to extrapolate the ground-state energy to "$S=0$," because the $S(S+1)/2I_NN$ contribution is not small enough to be neglected. After this correction, the $N^{-3/2}$ scaling law appears much more convincingly (Fig. 1). Our final extrapolations include only the $N=21,27,36$ results. We find $E_\infty=\langle 2\mathbf{S}_i\cdot\mathbf{S}_j\rangle=-0.365$ and $M_\infty=0.25$. These numbers by themselves do not call for long comments. Their accuracy could be estimated to be of the order of a few units in the last digit and E_∞ is compatible with previous other results [3–5]. Despite the dispersion of the results for the magnetization modulus M_∞ obtained by the different methods [3, 4, 16] (from 20% to 50% of the classical value), most of them converge, now, to a nonzero value. Our estimation of M_∞, definitely a nonzero value, is about 50% of the classical value, that is approximately the same estimate as the spin-wave theory [3].

We have shown in this work how exact diagonalizations on relatively small samples lead to a clear-cut illustration of the symmetry-breaking scheme in Heisenberg quantum antiferromagnets with an ordered ground state. The cornerstone of our approach is the identification of the macroscopic symmetry-breaking state [Eq. (2)] and of the nature and dynamics of the QDJ states. The scaling laws of the whole set of low-lying levels (QDJ and magnons) is a new evidence in favor of an *à la Néel* symmetry breaking for the triangular Heisenberg problem in the thermodynamic limit and fully justifies the spin-wave theories. This method can be generalized to other kinds of tensorial long-range order.

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