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Ground-state properties of the $S=\frac{1}{2}$ Heisenberg antiferromagnet on a triangular lattice

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We study the $S=\frac{1}{2}$ Heisenberg antiferromagnet on a triangular lattice with both nearest-neighbor (J_1) and next-to-nearest-neighbor (J_2) couplings. We have performed a spin-wave analysis around the classical ground state. At large (J_2/J_1) the Néel state is destabilized and the system exhibits the "order from disorder" phenomenon with a threefold-degenerate ground state that spontaneously breaks the lattice rotational invariance. These results are in agreement with a Lanczos study of a 12-site lattice. For even larger values of J_2/J_1 the spin-wave calculation shows the existence of a second transition to an incommensurate spiral. Thus, the previously suggested existence of chiral order in this model should be reanalyzed.

The discovery of the remarkable magnetic properties of high-T_c superconductors has led to a reexamination of antiferromagnetic quantum spin systems mainly in two dimensions. The crucial issue is the nature of the ground state. Anderson[1] conjectured that a fully disordered resonating-valence-bond (RVB) state may be the ground state of the Heisenberg model in two dimensions. However, the spin-$\frac{1}{2}$ square-lattice Heisenberg antiferromagnet has been studied by several techniques and there is now convincing evidence that it has a Néel-ordered ground state. Even in the presence of a next-to-nearest-neighbor frustrating interaction numerical results suggest[2] that no RVB state is required to describe its properties. In different regions of parameter space the best candidates for describing the low-energy behavior of the model are *ordered* states.[2] Another prominent model candidate[1] for having novel ground states is the triangular Heisenberg antiferromagnet in the case of spin-$\frac{1}{2}$. With only nearest-neighbor couplings, it has been studied[3] by variational wave functions, exact diagonalization of small clusters, and semiclassical calculations. All these results point to a Néel-ordered ground state[4] with a staggered magnetization reduced from the classical value 0.5 to about 0.24 (although this result is not as firm as in the case of the square lattice). Then, although for a classical Ising model the triangular lattice is clearly frustrated, for a Heisenberg model it presents 120° Néel order. It is thus desirable to introduce frustration in the triangular lattice through next-nearest-neighbor interactions as was done for the square lattice. It can be argued that since the triangular lattice has a lattice magnetization more reduced from its classical value than the square lattice, then its Néel order may be destabilized more easily. It is also important to note that in addition to the RVB state there are other exotic spin configurations which, in principle, are allowed. For example, in the context of anyonic superconductivity it has been argued[5] that a uniform order parameter $\chi_i=\langle \mathbf{S}_i\cdot(\mathbf{S}_{i+\hat{x}}\times\mathbf{S}_{i+\hat{y}})\rangle$ which breaks parity (reflexions) and time-reversal acquires a nonzero vacuum expectation value. On the square lattice, numerical studies of frustrated Heisenberg models have shown no evidence of such a spin order.[2] However, it has been proposed[6] that this phenomenon occurs in the case of the triangular lattice when one considers next-to-nearest-neighbor exchange interactions, i.e., for the Hamiltonian:

$$H=J_1\sum_{\rm NN}\mathbf{S}_i\cdot\mathbf{S}_j+\alpha J_1\sum_{\rm NNN}\mathbf{S}_i\cdot\mathbf{S}_k\,, \tag{1}$$

with NN denoting the sum over nearest-neighbor pairs of sites, NNN over next-to-nearest neighbors, $\mathbf{S}_i$ are quantum spins, and $\alpha=J_2/J_1$.

In this paper, we investigate the ground-state properties of (1) using the spin-wave approach. We find that the Néel state is stable up to a critical coupling $\alpha_c\simeq\frac{1}{8}$. Beyond this value, the classical ground state has a continuous degeneracy which is lifted by the spin-wave corrections to the energy. This is the "order from disorder" phenomenon also found on the square lattice.[2] Here we find three degenerate states in the quantum problem. Beyond another critical coupling $\alpha_c'\simeq1$, an incommensurate spiral becomes the lowest-energy state. We then compare these results with an *ab initio* Lanczos study and discuss the possible existence of chiral order.

Te perform the spin-wave study we first need to know the lowest-energy classical spin configurations of the Hamiltonian (1) as a function of the parameter α. The classical ground state is known to be of the form[7]

$$\mathbf{S}_i=\mathbf{u}\cos(\mathbf{Q}\cdot\mathbf{R}_i)+\mathbf{v}\sin(\mathbf{Q}\cdot\mathbf{R}_i)\,. \tag{2}$$

In this formula $\mathbf{u}$ and $\mathbf{v}$ are two orthogonal unit vectors, $\mathbf{R}_i$ is the position of site i in real space. The wave vector $\mathbf{Q}$ has to minimize $J(k)$, the Fourier transform of the exchange coupling,

$$J(k)=\sum_{i,j}J_{ij}\cos[\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)]\,. \tag{3}$$

In our case, we have

$$J(k)=\cos(k_x)+2\cos\left(\frac{k_x}{2}\right)\cos\left(\frac{k_y\sqrt{3}}{2}\right)+\alpha\left[\cos(k_y\sqrt{3})+2\cos\left(\frac{3k_x}{2}\right)\cos\left(\frac{k_y\sqrt{3}}{2}\right)\right], \tag{4}$$

where we set $J_1=1$. We have obtained the various minima of the function $J(k)$ inside the first Brillouin zone of

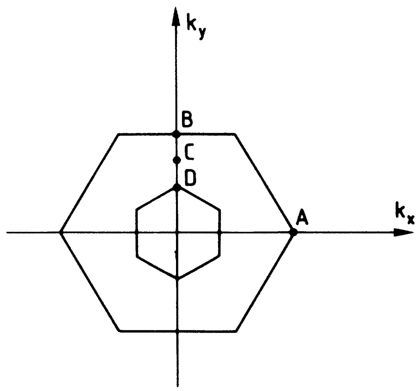

FIG. 1. The Brillouin zone of the triangular lattice with the various minima of $J(k)$ when α is varied. The small hexagon inside is the Brillouin zone for a Néel sublattice.

the triangular lattice as α is varied.

(i) When $\alpha<\frac{1}{8}$, the minima are located at the corners of the hexagonal Brillouin zone such as point A in Fig. 1. There are six such points. However, we should not consider as distinct the points which are related by a reciprocal lattice vector. We are thus left with only two points, say A on Fig. 1, with coordinates $\mathbf{Q}_0=(4\pi/3,\ 0)$ and $-\mathbf{Q}_0$, but it is easily seen in Eq. (2) that they are on the same orbit of the broken $O(3)$ group of spatial rotations. There is thus a unique ground state which is the Néel state with 120° structure (not counting the rotational degeneracy). The energy of this state is $E_1(\alpha)=\alpha-\frac{1}{2}$.

(ii) For $\frac{1}{8}<\alpha<1$, the minima of $J(k)$ occur now at the centers of the faces such as point B in Fig. 1. These wave vectors have the peculiarity that they are half of a reciprocal lattice vector. For example, point B has $\mathbf{Q}_1=(0,\ 2\pi/\sqrt{3})$ and thus half the length of the vector joining the origin with the next zone along the k_y axis. This situation has been analyzed by Villain.[8] Although the spiral configuration given by Eq. (1) always corresponds to the minimum of the energy, it can happen that there are states not of this form degenerate with it. This is the case when the wave vector of the spiral is a peculiar rational fraction[8] of a reciprocal lattice vector. In our case one has indeed a continuum of classical states with the same energy as the spiral with vector $\mathbf{Q}_1$. They can be parametrized by an angle θ and are shown in Fig. 2. One also has to add the configurations obtained from those of Fig. 2 by any discrete lattice symmetry operation (when different). Their energy is $E_2(\alpha)=-\frac{1}{3}(1+\alpha)$ independent of θ.

(iii) When $\alpha>1$, the minima move away from the boundary of the Brillouin zone and are on the lines connecting the origin to the middle of the faces such as point C in Fig. 1. In the case of point C, the wave vector is $\mathbf{Q}_2=(0,Q_y)$ where

$$\cos\left[Q_y\frac{\sqrt{3}}{2}\right]=-\frac{1}{2}-\frac{1}{2\alpha}. \tag{5}$$

The complete picture, of course, has sixfold symmetry. The previous relation with the reciprocal lattice vectors is lost so no new degeneracy occurs. Since $\mathbf{Q}$ and $-\mathbf{Q}$ are in the same orbit we deduce that there are three states in this phase. Their energy is $E_3(\alpha)=-\frac{1}{2}\alpha-1/6\alpha$. They are incommensurate spirals for a generic α.

When α becomes very large, the vectors are approaching points such as D in Fig. 1. These are the corners of the Brillouin zone for the Néel sublattices of the triangular lattice. This fact has a very simple interpretation: For $\alpha\to\infty$, the three sublattices are essentially decoupled and it is energetically favorable to put them in the 120° Néel configuration. In this situation the nearest-neighbor term in the energy does not contribute anymore and one has an energy which is $E_\infty(\alpha)=-\frac{1}{2}\alpha$. This spin configuration is below the continuum found in (ii) as soon as $\alpha>2$. But the spiral is always lower in energy as soon as $1/\alpha$ is nonzero, as seen in Fig. 3. Such states are only approached asymptotically when α goes to infinity. They are irrelevant for any finite α. The various energies are plotted in Fig. 3.

Let us now describe the semiclassical analysis of Hamiltonian (1) using as a starting point the ground states found above. We have performed a spin-wave expansion using the standard Holstein-Primakov formalism.[9] We have retained the first nontrivial quantum corrections, i.e., first order in the $1/S$ expansion. If we study a spin configuration which can be written as a spiral, such as Eq. (2), the energy per site computed in the spin-wave approximation takes a compact form:

$$E_0=3S(S+1)J(Q_0)+3S\frac{\sqrt{2}}{2N}\sum_k\{[J(k)-J(Q_0)][J(Q_0+k)+J(Q_0-k)-2J(Q_0)]\}^{1/2}. \tag{6}$$

The staggered magnetization is given by

$$\langle S^z\rangle=S+\frac{1}{2}-\frac{1}{2N}\sum_k[1-\tanh^2(2\theta_k)]^{-1/2}, \tag{7}$$

where

$$\tanh(2\theta_k)=\frac{A(k)}{A(k)+J(k)-J(Q_0)}, \tag{8}$$

and

$$A(k)=\tfrac{1}{4}[J(Q_0+k)+J(Q_0-k)]-\tfrac{1}{2}J(k), \tag{9}$$

where N is the number of sites. Phases (i) and (iii) are readily treated by the use of these formulas with Q_0 the

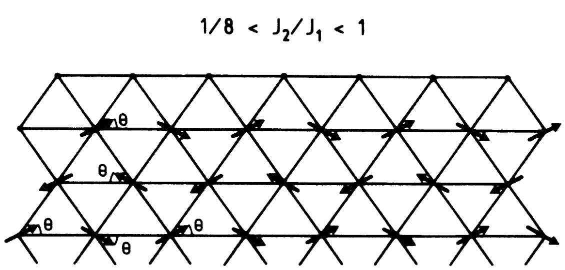

FIG. 2. The degenerate classical ground states when $\alpha=J_2/J_1$ is between $\frac{1}{8}$ and 1.

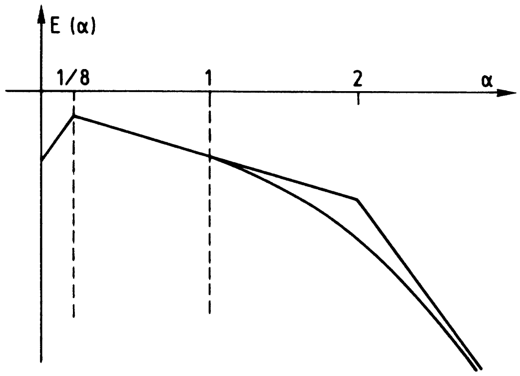

FIG. 3. Classical energy of the ground state vs α. The spiral is always lower in energy when $\alpha > 1$.

wave vector of the spiral. Phase (ii) requires a special study since the configurations shown in Fig. 2 are, in general, not of a spiral form. We have performed the spin-wave expansion around an arbitrary configuration characterized by an angle θ. The Hamiltonian has to be expressed in terms of spin operators each quantized in the direction of the classical orientation of the spin:

$$H=\sum_{i,j} J_{ij}[\cos(\phi_i-\phi_j)(S_i^z S_j^z+S_i^x S_j^x) +\sin(\phi_i-\phi_j)(S_i^x S_j^z-S_i^z S_j^x)+S_i^y S_j^y]\,. \quad (10)$$

Here the angle ϕ_i is between a fiducial quantization axis O_z and the spin direction at site i. At lowest order, the sine term disappears and we find that the cosine is constant along each of the three privileged directions of the triangular lattice, this being true for both the NN and NNN part of the Hamiltonian. Expressing the spin operators in terms of Holstein-Primakov bosons leads to lengthy expressions that will be published elsewhere. We have numerically obtained the ground-state energy as a function of $\cos(2\theta)$. The zero-point fluctuations of the spin waves lift the degeneracy in θ and we find that the true minima are given by $\theta=0$, $\pi/2$ (as well as π, $3\pi/2 \cdots$ which are redundant). Taking into account the lattice discrete symmetry, we obtain *three* degenerate states that are selected by the spin-waves. Looking at Fig. 2 and taking $\theta=0$, one obtains a state which is made from rows of spins ferromagnetically arranged in the horizontal direction but with 180° antiferromagnetic orientation in the perpendicular direction. The two other states are obtained by taking the ferromagnetic direction to be at 60° or 120° with the horizontal.[10] This is all what one gets from the discrete symmetries.

This selection of states among a continuum is the "order from disorder" phenomenon previously emphasized[11] by Villain and Henley in the context of the square lattice *XY* model and by Oguchi, Nishimore, and Taguchi in the case of the fcc antiferromagnet.[12] Quantum fluctuations select some special states as ground states out of an otherwise infinitely degenerate manifold of classical states. Equation (5) shows that the three incommensurate spiral states of phase (iii) are obtained with continuity from the three states of phase (ii). This suggests that the quantum transition that happens around $\alpha \simeq 1$ is of second order. This is to be contrasted with the transition for $\alpha \simeq \frac{1}{8}$, which is due to a crossing of levels and thus presumably of first order (unless there is a new nonclassical phase in between for small S). The staggered magnetization in the spin-wave approximation also has been studied. We find that the critical spin at which the magnetization vanishes diverges at the two points $\alpha=\frac{1}{8}$ and $\alpha=1$, where the classical ground state changes. This is very close to what happens on the square lattice.[13,14] There are thus two regions of width $\simeq 0.05$ in J_2 centered around these points where the system is possibly disordered.

We have also analyzed the triangular lattice with $J_2 \neq 0$ using a Lanczos method on a 12-site lattice. The reason for using such a small lattice is that to respect the discrete symmetries of the bulk limit only lattices with a special number of sites N and geometry are allowed[3] as it occurs on the square lattice. In the present model, at large J_2 the system decouples into three sublattices; in addition, we need N to a multiple of 3 to avoid spurious finite-size effects. If we also require that the total spin is zero, then we need N even. It can be shown that only $N=12$ satisfies these requirements for lattices accessible with present day computers. In Fig. 4 we show the ground-state energy of this model as a function of J_2 ($J_1=1$). Also shown are two excited states at $J_2=0$ which become almost degenerate with the ground state for $J_2 \geq (0.1-0.2)$, in excellent agreement with the spin-wave prediction. The three states shown in Fig. 4 have zero momentum and they differ in the quantum number under rotations of the lattice in 120°.

What about chiral order in this model? Our study shows that in the analysis of Baskaran[6] two important states that compete with the chiral state were not considered. These are the Néel-ordered state and the triply degenerate states we found [phase (ii)]. The numerical

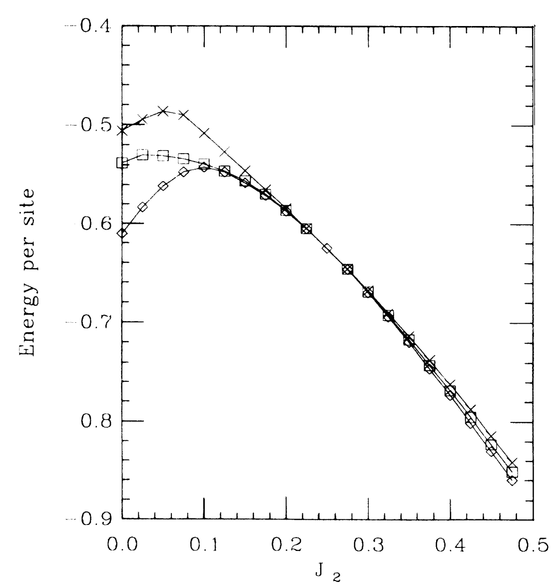

FIG. 4. Lanczos energies of the first three states of our model as a function of J_2 ($J=1$) on a 12 site lattice. The degeneracy due to the "order from disorder" phenomenon appears very clearly.

results strongly suggest that phase (ii) is realized in the $S=\frac{1}{2}$ model and thus the chiral state may exist only in a narrow region near $\alpha_c \sim \frac{1}{8}$. To analyze this possibility we studied the square of the chiral order parameter as that used on the square lattice.[2] The order of magnitude of the result is similar to that found for the square lattice (although we observe a little enhancement precisely in the intermediate region $\alpha_c \sim \frac{1}{8}$). Then, although our numerical results are not as robust as in the case of the square lattice, we believe that the competition of other states is strong enough to constrain the possible range of existence of the chiral state to a narrow regin in parameter space. More work is necessary to clarify if the chiral state is stable in that narrow region (other ordered states may exist there, as happens on the square lattice).

From the previous analysis we conclude that the triangular lattice with NNN interactions has a very rich phase diagram including a Néel phase, a phase with spin-wave selection of nontrivial ground states and a phase with incommensurate long-range order. The spin-wave approximation breaks down only in two narrow windows centered at the points where the nature of the classical ground state changes. These two regions may correspond to new nonclassical ground states. This semiclassical picture is confirmed by our numerical study. Finally, we remark that the existence of chiral order in this model needs further revision.

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[1]P. W. Anderson, Mater. Res. Bull. **8**, 153 (1973).

[2]E. Dagotto and A. Moreo, Phys. Rev. Lett. **63**, 2148 (1989), and references therein.

[3]D. A. Huse and V. Elser, Phys. Rev. Lett. **60**, 2531 (1988); H. Nishimori and H. Nakanishi, J. Phys. Soc. Jpn. **57**, 626 (1988); S. Fujiki, Can. J. Phys. **65**, 489 (1987).

[4]Th. Jolicoeur and J. C. Le Guillou, Phys. Rev. B **40**, 2727 (1989); H. Nishimori and S. Miyaki, Prog. Theor. Phys. **73**, 18 (1985).

[5]X. G. Wen, F. Wilczek, and A. Zee, Phys. Rev. B **39**, 11413 (1989); V. Kalmeyer and R. Laughlin, Phys. Rev. Lett. **59**, 2095 (1987).

[6]G. Baskaran, Phys. Rev. Lett. **63**, 2425 (1989).

[7]J. Villain, J. Phys. Chem. Solids **11**, 303 (1959); A. Yoshimori, J. Phys. Soc. Jpn. **14**, 807 (1959).

[8]J. Villain, J. Phys. (Paris) **38**, 385 (1977).

[9]L. Walker, in *Magnetism,* edited by G. T. Rado and H. Suhl (Academic, New York, 1963).

[10]P. Chandra and P. Coleman arrived independently to similar conclusions (private communication).

[11]J. Villain, R. Bidaux, J. P. Carton, and R. Conte, J. Phys. (Paris) **41**, 1263 (1980); C. L. Henley, Phys. Rev. Lett. **62**, 2056 (1989).

[12]T. Oguchi, H. Nishimori, and Y. Taguchi, J. Phys. Soc. Jpn. **54**, 4494 (1985).

[13]P. Chandra and B. Doucot, Phys. Rev. B **38**, 9335 (1988).

[14]A. Moreo, E. Dagotto, T. Jolicoeur, and J. Riera (unpublished).