Influence of quantum fluctuations on zero-temperature phase transitions between collinear and noncollinear states in frustrated spin systems

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We study a square-lattice spin-$\frac{1}{2}$ Heisenberg model where frustration is introduced by competing nearest-neighbor bonds of different signs. We discuss the influence of quantum fluctuations on the nature of the zero-temperature phase transitions from phases with collinear magnetic order at small frustration to phases with noncollinear spiral order at large frustration. We use the coupled-cluster method for high orders of approximation (up to LSUB6) and an exact diagonalization of finite systems (up to 32 sites) to calculate ground-state properties. The role of quantum fluctuations is examined by comparing ferromagnetic-spiral and antiferromagnetic-spiral transitions within the same model. We find clear evidence that quantum fluctuations ''prefer'' collinear order, and that they may favor a first-order transition instead of a second-order transition when there are no quantum fluctuations.

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INTRODUCTION

While quantum fluctuations do not influence the critical properties of phase transitions at $T>0$, they play an important role at $T=0$, and can yield to quantum phase transitions, which have attracted a lot of attention in recent times (see, e.g., Ref. 1). Quantum fluctuations arise due to Heisenberg's uncertainty principle, and play a role similar to those of thermal fluctuations (for $T>0$) in classical transitions. The spin-$\frac{1}{2}$ Heisenberg model is a basic model which shows strong quantum fluctuations in the antiferromagnetic case. While the ground-state of the pure Heisenberg antiferromagnet (HAFM) on the square lattice shows Néel long-range order,[2] a competition of bonds can increase quantum fluctuations and may result in rotationally invariant paramagnetic states, suppressing the (collinear) Néel order. This was demonstrated by recent experiments on (quasi-) two-dimensional Heisenberg systems, like CaV_4O_9 (see, e.g., Refs. 3 and 4) or $SrCu_2(BO_3)_2$ (see, e.g., Refs. 5 and 6).

Besides local singlet formation, magnetic frustration is an important mechanism to drive zero-temperature transitions. In the classical Heisenberg model, strong frustration often leads to noncollinear (e.g., spiral) spin states which may or may not have counterparts in the quantum case. It is generally argued that quantum fluctuations prefer a collinear ordering. A typical example is the frustrated spin-$\frac{1}{2}$ J_1-J_2 model on the square lattice (see, e.g., Refs. 7–11). Here the classical version of the J_1-J_2 model has a continuously degenerate ground state for $J_2>J_1/2$, but quantum fluctuations can remove this degeneracy, yielding to a collinear state (''order from disorder'' phenomenon; see, e.g., Refs. 12 and 13). Moreover quantum fluctuations can shift the critical point of a collinear-noncollinear transition, so that the collinear state can survive into a region where classically it is already unstable.[14–16]

In this paper we extend our previous work,[16] where we have studied the transition from a collinear Néel order to noncollinear spiral order in a frustrated spin-$\frac{1}{2}$ HAFM, and now consider the transition from a collinear ferromagnetic order to a noncollinear spiral order within the same model. While in the classical version of the model both situations can be mapped onto each other, the quantum model behaves basically different in both cases. This is because of the different nature of the collinear state: While the quantum Néel state on two-dimensional lattices exhibits strong quantum fluctuations (the sublattice magnetization of the HAFM on the square lattice is only about 60% of its classical value), the ferromagnetic state is the same for the quantum and classical models, and there are no quantum fluctuations in this state.

We use the coupled-cluster method[17,18] (CCM) and an exact diagonalization of finite systems to calculate the ground state. The CCM is a very powerful method and, particularly high-order implementations of this method can be used to obtain a consistent description of various aspects of quantum spin systems (for an overview see, for example, Refs. 19–24). We note that another important method for spin systems, the quantum Monte Carlo method, cannot be used for frustrated spin systems since it suffers from the minus sign problem.

MODEL

We consider a spin-$\frac{1}{2}$ Heisenberg model on a square lattice with two kinds of nearest-neighbor bonds J and J', as shown in Fig. 1:

$$H=J\sum_{\langle ij\rangle_1}\mathbf{S}_i\cdot\mathbf{S}_j+J'\sum_{\langle ij\rangle_2}\mathbf{S}_i\cdot\mathbf{S}_j. \tag{1}$$

The sums over $\langle ij\rangle_1$, and $\langle ij\rangle_2$ represent sums over the nearest-neighbor bonds, shown in Fig. 1 by dashed and solid lines, respectively. Each square-lattice plaquette consists of three J bonds and one J' bond. A model with such a zigzag pattern has been treated with various methods.[16,25,26]

In this paper we consider only cases in which J and J' have *different* signs (i.e., one bond is ferromagnetic, while the other is antiferromagnetic) so that the plaquettes are frustrated. The case with antiferromagnetic J bonds (i.e., $J>0$ and hence $J'<0$) was studied previously using linear spin-wave theory,[26] exact diagonalization, and the coupled-cluster

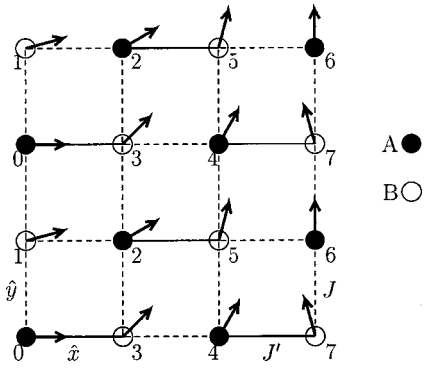

FIG. 1. Illustration of the classical spiral state for the square-lattice Heisenberg model of Eq. (1), with two kinds of regularly distributed nearest-neighbor exchange bonds: J (dashed lines) and J' (solid lines). The spin orientations at A and B lattice sites are defined by the angles $\theta_n = n\Phi$ where $n = 0,1,2,\ldots,$ and Φ is the characteristic angle of the spiral state. The state is shown for $\Phi = \pi/12$ and $n = 0,1,\ldots,7$, and refers to the ferromagnetic case ($J<0$) with a $J' > |J|/3$. For the antiferromagnetic case ($J>0$ and $J' < -J/3$) all spins on the B sublattice are reversed.

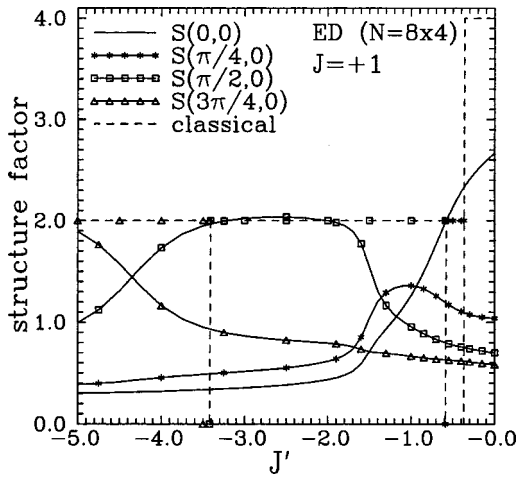

FIG. 2. Ground-state structure factor $S(\mathbf{k}) \propto \Sigma_{i,j\in A} \mathbf{e}^{i(\mathbf{R}_j - \mathbf{R}_i)\cdot\mathbf{k}} \times \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$ (i.e., the summation is taken over one sublattice) for a 8×4 lattice (with antiferromagnetic $J=+1$) for the quantum and classical cases for various spiral vectors $\mathbf{k}$.

method.[16] In this paper we therefore focus our attention mainly on the ferromagnetic case (i.e., $J<0$ and $J'>0$), but compare the obtained results with those of the antiferromagnetic case.

CLASSICAL GROUND STATE

We consider the ground state of the classical version of model (1), i.e., the spins $\mathbf{S}_i$ are assumed to be classical vectors. For $|J'|<|J|/3$ (and J and J' having different signs) the ground state of Eq. (1) is collinear (i.e., ferromagnetic or antiferromagnetic depending on the sign of J). At the critical point $J'_c = -J/3$, a second-order transition takes place from the collinear state to a noncollinear state of spiral nature (see Fig. 1), with a characteristic pitch angle $\Phi = \pm|\Phi_{\rm cl}|$ given by

$$|\Phi_{\rm cl}| = \begin{cases} 0, & |J'| < \dfrac{|J|}{3}, \\ \arccos\left(\dfrac{1}{2}\sqrt{1+\dfrac{1}{|J'|}}\right), & |J'| \geqslant \dfrac{|J|}{3}. \end{cases} \tag{2}$$

Note that for $\Phi=0$ this is the collinear state.

The spins $\mathbf{S}_A$ and $\mathbf{S}_B$, belonging to the A and B sublattices, respectively, can be expressed in terms of the spiral $\mathbf{k}$ vector[16] with $\mathbf{k}=(2\Phi,0)$ (see Fig. 1). We note that this spiral state is incommensurate in the x direction. We also note that for the classical model the antiferromagnetic case can be transformed into the ferromagnetic case by the simultaneous substitution $J \to -J$, $J' \to -J'$ and $\mathbf{S}_{i\in B} \to -\mathbf{S}_{i\in B}$. Hence the physics for both cases is classically the same.

CALCULATION OF THE QUANTUM GROUND STATE

To calculate the quantum ground state of Hamiltonian (1) we use the CCM. Details concerning the treatment of model (1) with the CCM are given in Ref. 16. We use the CCM for high orders of approximation up to LSUB6 (using 1638 fundamental configurations).

We further exactly diagonalize finite lattices of rectangular shape ($L_x \times L_y = 4\times4$, 6×4, and 8×4) using periodic boundary conditions. The longer side L_x of the rectangle corresponds to the direction of the J' bonds, and so we can diminish the influence of the boundary conditions by an increase of L_x.

COLLINEAR-NONCOLLINEAR TRANSITION

While classically we always have a second-order phase transition from collinear order to noncollinear order at $J'_c = -J/3$, for the quantum case we obtain a different behavior for the ferromagnetic and the antiferromagnetic case.

Using the CCM, for the antiferromagnetic case ($J=+1$) we find indications for a shift of this critical point to a value $J'_c \approx -1.35$ (see Fig. 4). On the other hand, for the ferromagnetic case ($J=1$) we do not find such a shift (see Fig. 4). The exact diagonalization (ED) data of the structure factor $S(\mathbf{k})$ (see Figs. 2 and 3) agree to these findings. For $J=+1$ (see Fig. 2) the collinear Néel order [$\mathbf{k}=(0,0)$] becomes unstable against the noncollinear spiral order [$\mathbf{k}=(\pi/4,0)$] in the classical model for $J' \lesssim -0.36$, but in the quantum case only for $J' \lesssim -0.95$. The situation for the ferromagnetic case ($J=-1$) is again different. Here the results of the structure factor (see Fig. 3) show that the transition from $\mathbf{k}=(0,0)$ (collinear ferromagnetic order) to $\mathbf{k}=(\pi/4,0)$ (spiral order) takes place at nearly the same value of $J' \approx 0.36$ for both the classical case and the quantum case.

Taking the deviation of the on-site magnetic moment $\langle S_i \rangle$ from its classical value $\langle S_i \rangle_{\rm cl} = \frac{1}{2}$ as an indication of the degree of quantum fluctuations, we can compare the strength of quantum fluctuations near the collinear-noncollinear transitions for both antiferromagnetic and ferromagnetic cases. As reported in Ref. 16 for $J=+1$, the quantum fluctuations are particularly strong near the antiferromagnetic-spiral transition, leading to an on-site magnetic moment less than 20% of

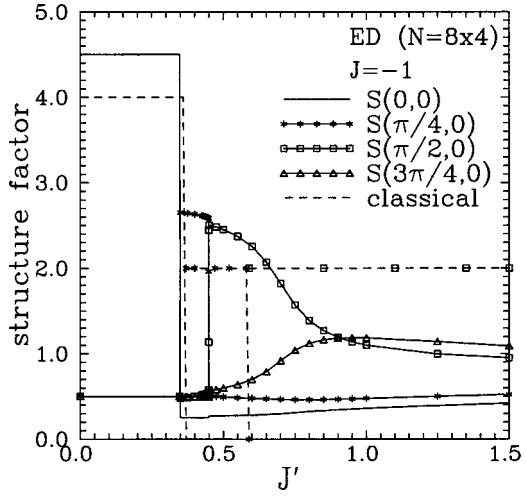

FIG. 3. Ground-state structure factor $S(\mathbf{k})$ (see Fig. 2) for a 8×4 lattice (with ferromagnetic $J=-1$) for the quantum and classical cases for various spiral vectors $\mathbf{k}$.

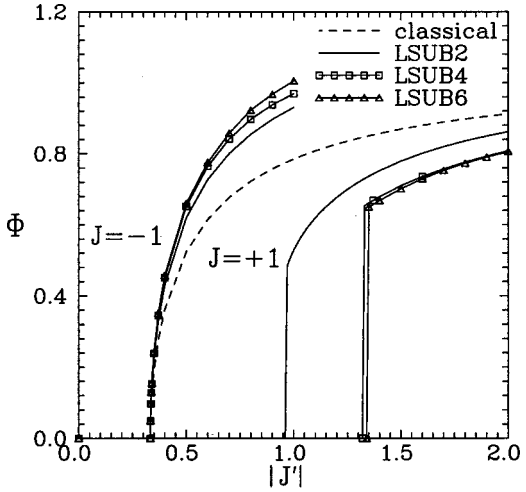

FIG. 4. Pitch angle Φ vs $|J'|$ for the quantum and classical cases. While Φ is classically the same for the ferromagnetic case ($J=-1$, $J'>0$) and for the antiferromagnetic case ($J=+1$, $J'<0$), the quantum pitch angle is different for both cases. The curves left of the classical (dashed) curve belong to $J=-1$, and those right of it belong to $J=+1$. Note that for the ferromagnetic case ($J=-1$) for $J'>1$ the pitch angle Φ becomes meaningless, since the spiral order is already destroyed in this region (cf. Fig. 5).

its classical value. On the other hand, it can be seen from Fig. 5 that the on-site magnetic moment takes its classical value $\frac{1}{2}$ up to $J'\approx0.36$ for $J=-1$, and therefore virtually *no* quantum fluctuations occur at the ferromagnetic-spiral transition. Hence the shift of the critical J_c' in the antiferromagnetic case can clearly be attributed to the strong quantum fluctuations. In general, our findings are consistent with the statement that quantum fluctuations (which we have in the antiferromagnetic case only) prefer a collinear ordering, so that in this case the quantum collinear state can survive into a frustrated, region where classically the collinear state is already unstable.

We further note an agreement between the CCM and ED results beyond the critical J_c'. By examining the structure factors (see Figs. 2 and 3), we find that for the antiferromagnetic (ferromagnetic) case the transitions to a spiral state with a greater $\mathbf{k}$ vector (i.e., with a greater pitch angle Φ) always occur in the quantum model at a greater (smaller) *absolute* value of J' than the corresponding classical transitions. This agrees with the CCM results of the pitch angle (see Fig. 4), where we have $\Phi_{\rm qm}<\Phi_{\rm cl}$ ($\Phi_{\rm qm}>\Phi_{\rm cl}$).

The discussion given above corresponds to our finding concerning the order of the transition. Clearly in the ferromagnetic case ($J'=-1$), both the classical and quantum models show a second-order phase transition (see Figs. 4 and 5). On the other hand, it was discussed in Ref. 16 that the collinear-noncollinear transition in the antiferromagnetic case ($J=+1$) is probably a first-order phase transition for the quantum model (cf. Fig. 4), as opposed to the second-order transition in the classical case.

FORMATION OF LOCAL SINGLETS

For sufficiently strong antiferromagnetic J' bonds, system (1) is characterized by a tendency to singlet pairing of the two spins coupled by a J' bond, and hence the long-range magnetic (collinear or noncollinear) order is destroyed. We obtain clear indications of a second-order phase transition to a quantum paramagnetic dimerized phase at a certain critical value of $J'=J_s'$. For $J'>J_s'$ the on-site magnetic moment $\langle S_i\rangle$ becomes zero. For the ferromagnetic case ($J=-1$) we find $J_s'\approx1$ using the extrapolated CCM-LSUBn results (see Fig. 5). We may also consider the inflection points of $\langle S_i\rangle$ versus J' for the LSUBn approximations, assuming that the true curve will have a negative curvature up to the critical point. We find the corresponding inflection points at $J'\approx1.2\,(n=2)$, $J'\approx0.76\,(n=4)$ and $J'\approx0.74\,(n=6)$, indicating a critical J_s' even slightly smaller than $J_s'\approx1$.

The ED data give a similar approximation of J_s': For $J'\approx1$, finite-size effects in spin-spin correlations disappear al-

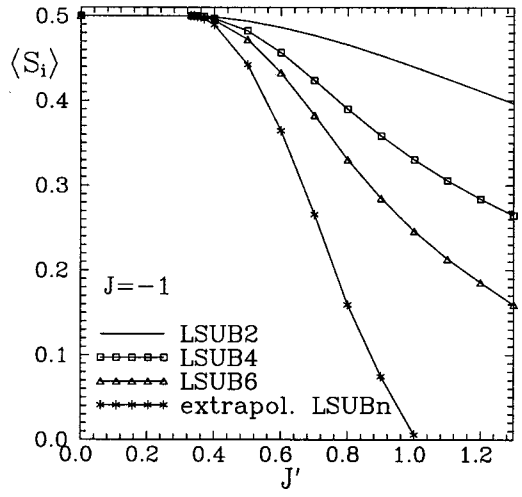

FIG. 5. On-site magnetic moment vs J' for the ferromagnetic case ($J=-1$) calculated within the CCM-LSUBn approximations and extrapolated to $n=\infty$ (the extrapolation is done as described in Ref. 16).

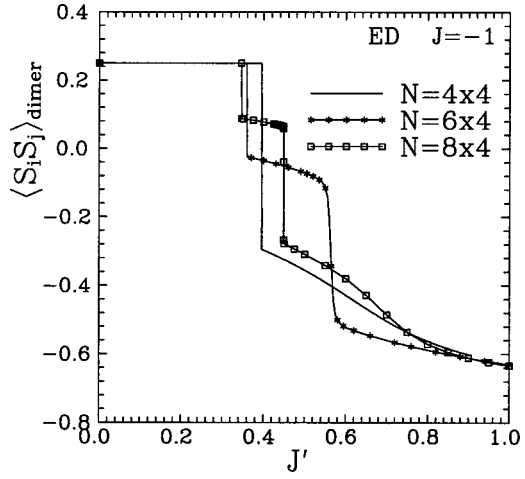

FIG. 6. Nearest-neighbor spin-spin correlation of the two spins connected via a J' bond vs J' for the ferromagnetic case using exact diagonalization (ED) data.

most completely (for illustration, see Fig. 6). This indicates that spin-spin correlations are short ranged, with a length scale smaller than the size L_x.

We note that for the antiferromagnetic case ($J=+1$) the strength of antiferromagnetic J' needed for breaking Néel order by formation of local singlets is much larger ($J'_s \approx 3$; see Ref. 16). The lower critical $J'_s \approx 1$ in the ferromagnetic case is due to frustration which assists local singlet formation (cf. Refs. 27 and 28).

SUMMARY

Using CCM and ED techniques, we have studied the influence of quantum fluctuations on zero-temperature transitions between collinear ordered and noncollinear ordered states in a frustrated spin-$\frac{1}{2}$ square-lattice Heisenberg model with two kinds of nearest-neighbor exchange bonds. The frustration drives a second-order transition between collinear (antiferromagnetic or ferromagnetic) and noncollinear (spiral) states in the classical model. For the quantum model the CCM provides a consistent description of collinear, noncollinear, and disordered phases, while some other standard techniques (e.g., the quantum Monte Carlo technique) are not applicable. We find a strong influence of quantum fluctuations on the nature of the collinear-noncollinear transition, and quantum fluctuations (which favor collinear ordering) may change the second-order classical transition to a first-order quantum transition. If quantum fluctuations are suppressed in the collinear phase of the quantum model, the transition to the spiral phase is similar for the quantum and classical models.

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