Quantum phase transition in square- and triangular-lattice spin-$\frac{1}{2}$ antiferromagnets

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We use the coupled-cluster method to study ground-state properties of anisotropic $S=\frac{1}{2}$ antiferromagnets on square and triangular lattices, with the inclusion of arbitrarily long-ranged two-spin correlations. We detect the singularities of various quantities associated with the quantum phase transitions and also compute their critical exponents. The two-spin correlation coefficients for the triangular lattice are found to exhibit an interesting oscillatory behavior in their signs, knowledge of which could assist the implementation of quantum Monte Carlo simulations.

The ground-state magnetic ordering of two-dimensional quantum antiferromagnets with or without frustration has attracted much theoretical interest. While various calculations[1] including extensive quantum Monte Carlo simulations[2] strongly support the existence of a Néel ordering for the square-lattice $S=\frac{1}{2}$ Heisenberg antiferromagnet with a reduced magnetic moment of about 68% of its classical value, the three-sublattice ordering for the corresponding triangular case is much less clear. For example, the conventional spin-wave results,[3] variational wave function calculations[4] and recent finite-cluster studies[5] support a magnetically ordered phase, whereas a high-order series expansion by Singh and Huse[6] suggests that the system may be at its critical point for antiferromagnetism. One noticeable difficulty in dealing with frustrated antiferromagnets is the unavailability of the otherwise usually very accurate and decisive quantum Monte Carlo simulations, due to the ''minus sign problem'' inherent in the unknown nodal surface of the ground-state wave function.

Recently, the so-called coupled-cluster method (CCM) has been successfully applied to collinear magnets in order to study the quantum phase transitions.[7–9] In this paper we extend these studies to include the triangular lattice $S=\frac{1}{2}$ antiferromagnet, a noncollinear and frustrated magnet. By computing such ground-state properties as the energy, the sublattice magnetization, and the anisotropy susceptibility as functions of the anisotropy parameter, we are able to study possible quantum phase transitions in the anisotropic models and compute the critical exponents, even if the critical anisotropy may lie outside the radius of convergence of series expansion methods. Furthermore, the systematic and fully microscopic parametrization of the ground-state wave function in terms of spin correlations in the CCM enables us to probe its nodal structure by investigating the sign oscillations of the spin-spin correlation coefficients.

Since detailed descriptions of the fundamentals of the CCM are available in the literature,[10] we only highlight the essential ingredients of its application in the present context. The CCM ansatz for the ground ket state is given by

$$|\Psi\rangle=e^{S}|\Phi\rangle, \quad S=\sum s_I C_I^+ , \tag{1}$$

where the correlation operator S is decomposed wholly in terms of mutually commuting creation operators $\{C_I^+\}\equiv\{(C_I^-)^\dagger\}$ for distinct multispin excitations with respect to the reference state $|\Phi\rangle$, i.e., $\langle\Phi|C_I^-|\Phi\rangle=0$. By taking the inner product of the Schrödinger equation in the form of $e^{-S}He^{S}|\Phi\rangle=E_g|\Phi\rangle$, with both the reference state itself and the set of multispin excitation states, i.e., $\{C_I^+|\Phi\rangle\}$, we thus obtain respectively the ground-state energy

$$E_g=\langle\Phi|e^{-S}He^{S}|\Phi\rangle \tag{2}$$

and the coupled set of nonlinear equations

$$0=\langle\Phi|C_I^- e^{-S}He^{S}|\Phi\rangle, \tag{3}$$

by which the correlation coefficients $\{s_I\}$ can be determined. It can be seen that this parametrization leads to a workable scheme since the similarity-transformed Hamiltonian, $e^{-S}He^{S}$, which can be expressed as the nested commutator expansion $H+[H,S]+(1/2!)[[H,S],S]+\cdots$, terminates at a finite order due to the fact that the correlation operator S contains mutually commuting multiconfigurational creation operators only, as long as H is a finite-order multinomial of the elementary single-spin operators, as is the case for almost all models of interest. All other ground-state quantities can also be evaluated via a similar and elegant CCM parametrization of the ground bra state.[10]

We first consider the square lattice case[8] to illustrate the approximation scheme adopted throughout this paper and the main features of the calculations. The Hamiltonian under consideration describes the so-called $S=\frac{1}{2}$ XXZ antiferromagnet:

$$H=\sum_{(ij)}\left[-\sigma_i^z\sigma_j^z-\frac{\lambda}{2}(\sigma_i^+\sigma_j^+ +\sigma_i^-\sigma_j^-)\right], \tag{4}$$

where the summation runs over all nearest-neighbor bonds and λ denotes the anisotropy parameter. Note that we have performed a spin rotation on one of the two sublattices, namely, the up-down transformation, in which the $S=\frac{1}{2}$ Pauli matrices on the rotated sublattice obey a simple transformation as $\sigma^x\rightarrow-\sigma^x, \sigma^y\rightarrow\sigma^y$, and $\sigma^z\rightarrow-\sigma^z$. Here $\sigma^\pm(\equiv\sigma^x\pm i\sigma^y)$ are the corresponding creation and destruc-

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tion operators in the rotated spin coordinates. Such a transformation not only makes transparent the physical meaning of the correlation operator S, but also facilitates later discussions on the Marshall-Peierls sign theorem.

The Hamiltonian in Eq. (4) at $\lambda=0$ describes the usual Ising antiferromagnet with a Néel-ordered ground state or the fully aligned ferromagnetic configuration in the rotated spin coordinates, whereas $\lambda=1$ recovers the Heisenberg antiferromagnet. We thus choose the Néel state as the reference state. To implement the CCM in practice, the S operator has to be truncated. Here we employ the so-called full SUB2 approximation scheme[8] where all the two-spin correlations are retained while higher-order correlations are set to zero. This truncated S operator therefore has the following form:

$$S \rightarrow S_2 = \sum_{\mathbf{r}\mathbf{r}'} B_{\mathbf{r}\mathbf{r}'} \sigma_{\mathbf{r}}^{+} \sigma_{\mathbf{r}'}^{+}, \tag{5}$$

where the correlation coefficient $B_{\mathbf{r}\mathbf{r}'}$ depends only on the difference of its indices, i.e., $B_{\mathbf{r}\mathbf{r}'} = B_{\mathbf{r}-\mathbf{r}'}$, due to the lattice translational symmetries. Here the summations run over all lattice sites such that the vector $\mathbf{r}-\mathbf{r}'$ connects sites on different sublattices. This comes about because of the conserved z component of the total spin. With the prescription given by Eqs. (2) and (3), it is straightforward to compute the ground-state energy and the coupled set of nonlinear equations which, in turn, can be decoupled by performing a Fourier transform. We thus obtain

$$\frac{E_g}{N} = -2(1+16\lambda B_1) \tag{6}$$

and

$$\left(\frac{\lambda}{64} + \frac{B_1}{4} + 2\lambda B_1^2\right)\gamma_{\mathbf{q}} - \left(\frac{1}{4} + 4\lambda B_1\right) B_{\mathbf{q}} + \lambda\, \gamma_{\mathbf{q}} B_{\mathbf{q}}^2 = 0, \tag{7}$$

where $B_1 (\equiv \frac{2}{N} \Sigma_{\mathbf{q}} \gamma_{\mathbf{q}} B_{\mathbf{q}})$ denotes the correlation coefficient of a pair of nearest-neighbor spins. Here $\gamma_{\mathbf{q}} = \frac{1}{4} \Sigma_{\boldsymbol{\rho}} \exp(i\mathbf{q}\cdot\mathbf{r}_{\boldsymbol{\rho}})$, with the sum running over all four nearest-neighbor lattice vectors $\mathbf{r}_{\boldsymbol{\rho}}$ and $\mathbf{q}$, a vector in the momentum space, runs over the first magnetic Brillouin zone in the thermodynamic limit $N \rightarrow \infty$. N denotes the number of spins.

Clearly Eq. (7) imposes a quantitative self-consistency condition on the solution. This is in contrast to the conventional spin-wave theory or large-S expansion, where the consistency of the assumption of an ordered phase can only be ascertained qualitatively. It has been shown[7] that the full SUB2 approximation yields -2.603 for the ground-state energy per spin at the isotropic Heisenberg point ($\lambda=1$) with a sublattice magnetization of about 83% of its classical value. This energy should be compared with the corresponding classical energy of -2.0 and the quantum Monte Carlo result[2] of -2.677. Perhaps the most interesting feature in this approximation is the appearance of a terminating point $\lambda_c \approx 1.252$, beyond which no physical solution can be obtained. Although the CCM based on the Néel reference state is bound to break down in the region of the anisotropy parameter space where the true ground-state wave function possesses a different symmetry from that of the Néel order-

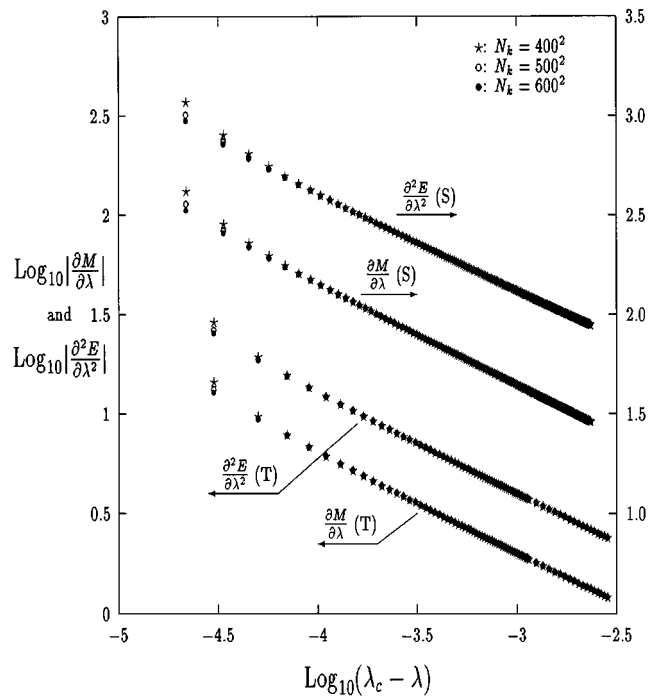

FIG. 1. Log-log plots of the second derivative of the ground-state energy and the first derivative of the sublattice magnetization with respect to the anisotropy parameter versus the deviation of the anisotropy from the corresponding critical anisotropy, for both the square-lattice (S, right scale) and triangular-lattice (T, left scale) antiferromagnets.

ing, it is clearly demonstrated in Ref. 7 that the terminating point corresponds to the critical point of a phase transition. The nature of this critical point can be elucidated by studying the singular behavior of various quantities. To this end, we discretize the magnetic Brillouin zone in solving Eq. (7), and use a large number N_k of points to obtain accurate numerical values of various quantities near the critical point.

Specifically, we calculate the anisotropy susceptibility defined as

$$\chi_a \equiv \frac{\partial^2 (E_g/N)}{\partial \lambda^2}. \tag{8}$$

Because Eq. (7) is explicitly given, the correlation coefficients and their derivatives with respect to the anisotropy parameter λ can be readily computed by taking derivatives of both sides of the equation and solving the equations so obtained. In Fig. 1 we show the log-log plot of the anisotropy susceptibility versus the deviation of the anisotropy away from the critical value, $\lambda_c - \lambda$. Clearly, in the region where both the deviation is sufficiently small and the convergence has been achieved by increasing N_k, χ_a shows a power-law singularity of the form: $\chi_a \sim (\lambda_c - \lambda)^{\mu}$ as $\lambda \rightarrow \lambda_c$. The slope readily yields the critical exponent $\mu = -\frac{1}{2}$. Equivalently, the ground-state energy has a $\frac{3}{2}$ power-law singularity at λ_c. In the same figure, we also present similar numerical results for the first derivative of the sublattice magnetization M with respect to the anisotropy. These clearly reveal a $\frac{1}{2}$ power-law singularity at λ_c, $M \sim M_c + k(\lambda_c - \lambda)^{1/2}$. It should be noted that while these singularities can also be

proved analytically, since the quadratic Eq. (7) can be solved explicitly, it is useful to be able also to determine these critical exponents numerically because an analytic solution is no longer available for the triangular lattice case discussed below.

We now describe our results for the triangular-lattice case. Following Singh and Huse,[6] we write the Hamiltonian as

$$H=\sum_{i\to j}\left[-\frac{1}{2}\sigma_i^z\sigma_j^z-\frac{\lambda}{2}(\sigma_i^x\sigma_j^x-2\sigma_i^y\sigma_j^y)+\frac{\sqrt{3}\lambda}{2}(\sigma_i^z\sigma_j^x-\sigma_i^x\sigma_j^z)\right], \tag{9}$$

where $\{\sigma_i^x,\sigma_i^y,\sigma_i^z\}$, the $S=\frac{1}{2}$ Pauli matrices at site i, are defined with respect to their local quantization axis, which is chosen to orient along the spin direction at site i of the classical ground state. Such a configuration, in the global spin coordinates, has three sublattices with spins on sublattice A pointing along the z direction, and spins on sublattices B and C being, respectively, rotated $-120°$ and $120°$ away from the z axis in the xz plane. It is easy to show that the Hamiltonian with $\lambda=1$ describes the conventional Heisenberg antiferromagnet, whereas the case $\lambda=0$ has as its ground state the fully aligned ferromagnetic configuration in the above local spin coordinates. We choose this configuration as our reference state in applying the CCM. Note that the summation in the Hamiltonian runs over all nearest-neighbor bonds but with directionality indicated by $i\to j$ which goes from A to B, B to C, and C to A.

We still restrict ourselves to the full SUB2 approximation $S\to S_1+S_2$, where the two-spin correlation operator S_2 retains the form given in Eq. (5) with the only difference being that now $\mathbf{r}-\mathbf{r}'$ can connect sites on the same sublattice because the reference state is no longer an eigenstate of the z component of the total spin, and for the same reason we must also include a single spin-flip operator S_1. We have, after considerable algebra, obtained the coupled set of equations analogous to Eq. (7) for the square-lattice case. We only report our findings here, and the details of the calculations will be presented elsewhere. Although the SUB2 equations may permit other solutions, we restrict ourselves henceforth to the so-called symmetric and coplanar solution in which $S_1=0$, and the two-spin correlation coefficients $B_{\mathbf{rr}'}$ for a given separation $\mathbf{r}-\mathbf{r}'$ depend only on whether the two spins are on the same or different triangular sublattices A, B, and C.

At the isotropic Heisenberg point, the SUB2 ground-state energy per spin is -2.015 which should be compared with the classical energy of -1.5 and -2.21 ± 0.01 obtained by Singh and Huse from a series expansion. We note that the full SUB2 approximation for the triangular lattice captures fewer quantum corrections to the classical energy than for the square lattice. This essentially reflects the significance of three-spin correlations not retained in the SUB2 approximation which are only present in the triangular lattice. As in the square lattice, we also observe a terminating point, now at a value $\lambda_c=1.335\,25$. In Fig. 1 we also show our numerical results for the critical exponents for the anisotropy susceptibility and the first derivative of the sublattice magnetization. Clearly, at the level of the full SUB2 approximation, the ground-state energy and sublattice magnetization have, respectively, $\frac{3}{2}$ and $\frac{1}{2}$ power-law singularities, which are identical to those of the square-lattice case. This strongly indicates that both phase transitions belong to the same universality class, and further supports the existence of three-sublattice ordering in the frustrated triangular antiferromagnet. This is fully consistent with the result of the sublattice magnetization computed within the full SUB2 approximation, which we find to be around 85% of its classical value in the Heisenberg case.

Finally, we turn our discussion to the structure of the ground-state wave functions of both systems in connection with the possibility of performing quantum Monte Carlo simulations. For the nonfrustrated antiferromagnets, the essential ingredient is provided by the Marshall-Peierls sign theorem which concerns the phase relations of the projection coefficients of the ground-state wave function onto a complete set of spin configurations.[11] This theorem, when applied to the square Heisenberg antiferromagnet in particular, states that all of the coefficients, when expressed in the spin-rotated coordinates introduced in Eq. (4), are positive. Equivalently, the ground-state wave function has only one nodal region, a connected region via the Hamiltonian in the spin configuration space where the wave function has the same sign. This feature is at the heart of straightforward applications of quantum Monte Carlo simulations.[2]

Let us now consider the CCM parametrization of the ground ket state. By expanding the exponential operator in Eq. (1), we can easily show that each spin-spin correlation coefficient is a projection coefficient of the ground-state wave function onto the corresponding elementary excitation configuration which flips a pair of spins with respect to the

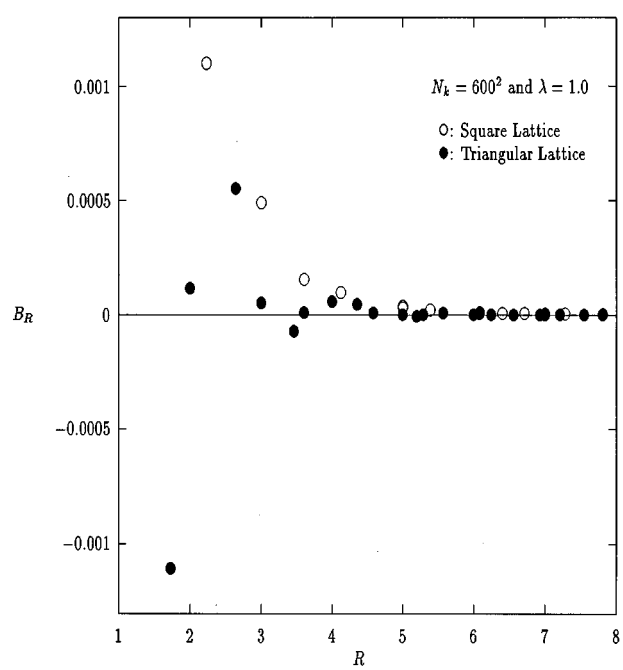

FIG. 2. Spin-spin correlation coefficient B_R as a function of the lattice distance R for both the square- and triangular-lattice Heisenberg antiferromagnets ($\lambda=1$). Note that we have omitted the nearest-neighbor correlation coefficient B_1 to enlarge the small oscillatory behavior.

Néel state. Although it is not obvious that the CCM in SUB2 approximation will satisfy the Marshall sign theorem for the square lattice, the numerical values of these coefficients at the isotropic Heisenberg point plotted in Fig. 2 clearly show that this is the case. By contrast, the corresponding coefficients for the frustrated triangular Heisenberg antiferromagnet are found to have an oscillatory behavior in their signs, as is also shown in Fig. 2. There has been some recent work[12] in which it is argued that the Marshall sign theorem may survive weak frustrations in certain models. However, our present finding is in favor of the breakdown of the Marshall sign theorem for the triangular Heisenberg antiferromagnet. More interestingly, perhaps, we note that the fixed-node Monte Carlo method[13] and its extension[14] for attacking both continuum and lattice fermion problems require a reliable trial wave function in terms of which the true wave function can be well approximated, especially in terms of its nodal structure. The oscillatory behavior observed here in the full SUB2 approximation for the frustrated triangular Heisenberg antiferromagnet may represent a reasonable description of the nodal structure of the exact wave function, since we expect corrections from higher-order spin correlations to be small.

To summarize, in this paper we have extended the domain of application of the CCM to include frustrated antiferromagnets. We study the magnetic order-disorder phase transitions of both square- and triangular-lattice antiferromagnets by employing the full SUB2 approximation, and find that at this level the criticalities of the phase transitions in both systems fall into the same universality class, although the precise nature of the phase transitions and more realistic estimates of the critical anisotropies need to be further investigated beyond the current level of approximation by incorporating higher-order spin correlations. There already exists, however, a marked difference in the structure of their ground-state wave functions in terms of the respective absence or presence of sign oscillations in their spin-spin correlation coefficients. Such a comparison may shed some light on the hitherto hidden rule that governs the nodal surface of the ground-state wave function of the triangular antiferromagnet, and thus help to circumvent the minus sign problem which bedevils Monte Carlo simulations for this system.

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