Simple Variational Wave Functions for Two-Dimensional Heisenberg Spin-$\frac{1}{2}$ Antiferromagnets

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We generalize a type of variational wave function introduced by Kasteleijn and Marshall, to include long-range correlations and nonbipartite lattices. We find the lowest-energy wave function in a three-parameter space for both the square- and triangular-lattice spin-$\frac{1}{2}$ Heisenberg antiferromagnets. This produces useful upper bounds on the ground-state energies of these systems. The wave functions are completely explicit, so that precise estimates of expectation values are readily obtained by Monte Carlo techniques. It appears that the antiferromagnet has long-range magnetic order on the triangular lattice, as well as on the square lattice.

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There has been a recent resurgence of interest in two-dimensional spin-$\frac{1}{2}$ quantum antiferromagnets, including a number of studies using variational wave functions of the Gutzwiller[1,2] or resonating-valence-bond[3,4] (RVB) type. Here we wish to discuss a different type of variational wave function that has the virtue of being very easy to work with numerically. This enables one to search readily a fairly large parameter space in order to find the best approximation to the ground state. In this paper we examine specifically spin-$\frac{1}{2}$ antiferromagnets on square and triangular lattices. However, we expect that the general type of variational wave function described below should be of rather general utility; another possible application that comes to mind is the magnetic phase diagram of solid ^{3}He.

For the square-lattice Heisenberg antiferromagnet, which has received much of the recent attention,[1-5] we find a wave function with energy $E \equiv \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle \simeq -0.3319$, where i and j are nearest-neighbor sites. This serves as a useful (and strict) upper bound on the ground-state energy, which is estimated to be $E_0 = -0.334 \pm 0.001$.[5] The various RVB- and Gutzwiller-type wave functions that have recently been proposed[1-4] have energies of -0.321 and higher. For the triangular-lattice case, we again obtain a wave function with an energy ($E \simeq -0.1789$) well below the proposed RVB-type wave functions,[6] but probably not as close in energy to the true ground state as for the square lattice. Previous numerical work on the square-lattice case strongly indicates that the ground state has long-range antiferromagnetic order[5,7]; it appears that this is also true for the frustrated triangular-lattice case.

We consider variational wave functions of the form

$$\psi = \sum_\alpha e^{\tilde{H}/2} |\alpha\rangle, \tag{1}$$

where the $\{\alpha\}$ form a complete orthonormal set of basis states and $\tilde{H}$, which contains the variational parameters, is an operator that is diagonal in this basis:

$$\tilde{H} |\alpha\rangle = \lambda_\alpha |\alpha\rangle. \tag{2}$$

If ψ is a true ground-state wave function, then (1) may be viewed as defining the "correct" $\tilde{H}$. In order to get a good approximation to the ground state of a given system, we want to find a best approximation to this "correct" $\tilde{H}$ using the variational method.

The expectation value of an operator A in the wave function ψ may be expressed as

$$\langle A \rangle \equiv \langle \psi | A | \psi \rangle = \frac{\sum_{\alpha\beta} e^{\lambda_\alpha^*/2} A_{\alpha\beta} e^{\lambda_\beta/2}}{\sum_\alpha |e^{\lambda_\alpha}|} = \frac{\sum_\alpha |e^{\lambda_\alpha}| \sum_\beta A_{\alpha\beta} e^{(\lambda_\beta - \lambda_\alpha)/2}}{\sum_\alpha |e^{\lambda_\alpha}|}, \tag{3}$$

where $A_{\alpha\beta} \equiv \langle \alpha | A | \beta \rangle$. The $\{\lambda_\alpha\}$ are potentially complex numbers. This expectation value is readily evaluated with standard Monte Carlo techniques; it is just the expectation value of $\sum_\beta A_{\alpha\beta} \exp[\frac{1}{2}(\lambda_\beta - \lambda_\alpha)]$ in the ensemble where state α occurs with probability proportional to $|\exp\lambda_\alpha|$. This ensemble may be viewed as the Boltzmann ensemble where the reduced Hamiltonian is the real part of $\tilde{H}$ (thus the notation). That expectation values may be so readily obtained numerically makes this type of variational wave function rather versatile and potentially quite useful for a variety of applications.

The quantum systems we will consider here are Heisenberg spin-$\frac{1}{2}$ antiferromagnets with Hamiltonian

$$H = \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \tag{4}$$

where the sum runs over all nearest-neighbor pairs of sites on a two-dimensional square or triangular lattice or a one-dimensional linear chain, and $\mathbf{S}_i$ is the spin operator at site i. Let us consider the square-lattice case first. Marshall[8] has proven that the ground-state wave function ψ_0 of the spin-$\frac{1}{2}$ Heisenberg antiferromagnet on any bipartite lattice (such as the square lattice or linear chain) may be chosen to be real in the basis of the eigenstates of the individual S_i^z. In addition, ψ_0 changes sign upon exchange of any nearest-neighbor pair of antiparallel spins. Thus for the square lattice, using this $\{S_i^z\}$ basis, we can choose the relative signs (thus phases) of the wave function to be precisely correct by having the

imaginary part of $\tilde{H}$ be

$$\tfrac{1}{2}\,\mathrm{Im}\tilde{H}=\pi\textstyle\sum_i^B(\tfrac{1}{2}-S_i^z), \tag{5}$$

where the sum runs over only those spins on the B sublattice. (All nearest neighbors of each spin on the B sublattice are on the A sublattice, and vice versa.) With this choice we have $\langle S_i^y\rangle=0$ for all sites, $\langle S_i^x\rangle\geq 0$ for sites on the A sublattice, and $\langle S_i^x\rangle\leq 0$ for sites on the B sublattice. The classical Néel state, $|\langle S_i^x\rangle|=\frac{1}{2}$, with spins oriented along the x axis, is then obtained if $\mathrm{Re}\tilde{H}=0$.

In general, the term in $\tilde{H}$ linear in the S_i^z corresponds to the classical spin configuration. The wave function given by

$$\tfrac{1}{2}\tilde{H}_c=\textstyle\sum_i h_i S_i^z \tag{6}$$

is just the product of individual spin states where $\exp[\frac{1}{2}h_i-(-\frac{1}{2}h_i)]=\exp(h_i)$ is the ratio of the amplitudes of the $S_i^z=\frac{1}{2}$ and $S_i^z=-\frac{1}{2}$ eigenstates for spin i. To orient a particular spin i to have angular momentum $+\frac{1}{2}$ along an arbitrary axis with polar coordinates (θ,ϕ), we choose

$$h_i=\log[\cot(\theta/2)]-i\phi. \tag{7}$$

Our choice (5) for the square lattice corresponds to $(\theta,\phi)_A=(\pi/2,0)$ on the A sublattice and $(\theta,\phi)_B=(\pi/2,\pi)$ on the B sublattice. We note that a vanishing real part of $\tilde{H}_c$ is possible only when the classical spins all lie in one plane.

Quantum corrections to the "classical" wave function (6) may be introduced, for example, by the addition of nearest-neighbor two-spin correlations,

$$\tilde{H}_2=-K_1\textstyle\sum_{\langle ij\rangle}S_i^zS_j^z, \tag{8}$$

where, as Marshall showed,[8] K_1 is real. The wave function given by (5) and $\mathrm{Re}\tilde{H}=\tilde{H}_2$ was examined by Kasteleijn[9] for the linear chain and Marshall[8] for a variety of bipartite lattices. The lowest energy for the square lattice is obtained for $K_1\simeq 1.1$ and is $E\simeq -0.322$. Marshall's[8] approximations to the energy of this wave function gave an underestimate of -0.328. Kasteleijn[9] and Marshall[8] failed to realize that this wave function has a staggered magnetization along the x axis of $\simeq 84\%$ of the classical value; this appears to have been first noted by Thouless.[10] Note that K_1 is small enough that the $\{S_i^z\}$ do not order; this is true for all the wave functions we discuss here.

We have generalized (8) to allow *all* two-spin interactions:

$$\mathrm{Re}\tilde{H}=-\textstyle\sum_{ij}K(r_{ij})S_i^zS_j^z, \tag{9}$$

where r_{ij} is the Euclidean distance between sites i and j measured in units of the nearest-neighbor lattice spacing. We have searched the three-parameter space

$$K(1)=K_1,\quad K(r)=k/r^{\sigma},\quad r>1, \tag{10}$$

for the square lattice and find the lowest-energy wave function at $K_1\simeq 2.6$, $k\simeq 1.9$, $\sigma\simeq 0.7$ with $E\simeq -0.3319$, and a staggered magnetization along the x axis of $\simeq 71\%$ of the classical value. These numbers should be compared with the series estimates for the ground state[5] of -0.334 ± 0.001 and 63%, respectively. Thus it appears that we have found a wave function whose energy is roughly 1% above the true ground-state energy. The overestimate of the staggered magnetization is to be expected, since this variational wave function is based on the classical wave function given by (5). Measurements were done on 10×10, 20×20, and 40×40 lattices with periodic boundary conditions. The 10×10 lattice showed only an $\simeq 0.05\%$ finite-size correction to the energy; thus quite accurate estimates can be made on rather small lattices. We performed our simulations on fifty independent lattices in parallel, first equilibrating for T Monte Carlo steps (MCS) per spin and then measuring every MCS for T MCS. The heat-bath algorithm was used with single-spin flip updates here. For 20×20 lattices and $T=256$, for example, the energy measurement had statistical error (1 standard deviation) of one part in 10^4. In principle, we could perform a more thorough search by allowing all the $K(r)$ to vary, calculating $dE/dK(r)$, and going downhill in this parameter space to find the minimum. It might be useful to see how much lower one could drop the energy with this procedure.

The long-range correlations in (10) suppress long-wavelength fluctuations of the z component of the uniform magnetization. This suppression is presumably present in the true ground state and appears to be important in the attaining of a low energy: Letting only the three nearest-neighbor interactions, $K(1)$, $K(\sqrt{2})$, and $K(2)$, vary independently, with all other $K(r)=0$, we were only able to lower the energy to $\simeq -0.3275$.

If we wish to extend (9) while keeping the spontaneous staggered magnetization entirely along the x axis (which is natural with this wave function) then the wave function should be invariant under a global flip of S^z. Thus the real part of the "correct" $\tilde{H}$ contains only even-spin couplings. From the above results it appears that the four-spin and higher-order couplings do not play a very important role in the attainment of a low energy. How accurate a wave function can one obtain with only two-spin interactions in $\tilde{H}$? This question could, in principle, be answered for the linear-chain antiferromagnet where many properties of the ground state are exactly or accurately known.[11]

We have examined the linear-chain (one-dimensional) antiferromagnet with only nearest-neighbor interactions and the wave function given by (1), (5), and (9) with $K(r)=kr^{-\sigma}$ for all r. In this two-parameter (k,σ) space both the energy and the staggered magnetization decrease upon going to large k and small σ; the staggered magnetization apparently vanishes for $\sigma\rightarrow 0$ at fixed $k\sigma$. The minimum energy of $\langle\mathbf{S}_i\cdot\mathbf{S}_{i+1}\rangle\simeq -0.4424$

occurs at $k\sigma \simeq 4$ and $0.1 \lesssim \sigma \lesssim 0.2$ where the staggered magnetization is less than 20% of its classical value. The exact ground-state energy for the linear chain is $E_0 \simeq -0.443\,15$, while the Gutzwiller wave function has energy -0.4421.[11] The spin-spin correlation function $\langle \mathbf{S}_i \cdot \mathbf{S}_{i+j} \rangle$ for our optimized wave function is between the estimated exact ground-state values and those of the Gutzwiller wave function[11] for all $j \leq 8$. Note that in this strong-coupling regime of large k and small σ, we performed spin-exchange Monte Carlo calculations to speed up convergence of our measurement greatly. Here we estimated the staggered magnetization from the long-distance spin-spin correlation function, since $\langle \mathbf{S}_i \rangle$ appears to have strong finite-size effects that lead to an underestimate.

We now describe our results for the triangular-lattice case.

The classical ground state of the triangular-lattice antiferromagnet has three sublattices, A, B, and C, with spins on each sublattice at an angle of $2\pi/3$ to those on the other two sublattices. Nearest-neighbor pairs of spins are on different sublattices, while each spin is on the same sublattice as all of its second neighbors at distance $\sqrt{3}$ nearest-neighbor spacings. Let us consider a particular classical ground state with the spins on the A sublattice oriented along the $+x$ axis, and those on the B and C sublattices rotated $2\pi/3$ away from the $+x$ axis in the x-y plane. As discussed above and by Miyashita,[12] this classical state can be written in our form (1) with

$$\tfrac{1}{2}\tilde{H}_c = \tfrac{2}{3}\pi i \left[\left(\textstyle\sum_i^B S_i^z \right) - \left(\textstyle\sum_i^C S_i^z \right) \right], \tag{11}$$

where the sums run over spins on the B and C sublattices, respectively.

Miyashita[12] considered the wave function with $\tilde{H} = \tilde{H}_c + \tilde{H}_2$ [from (8) and (11)], thus introducing only nearest-neighbor correlations in $\tilde{H}$. This wave function is optimized at $K \simeq 1.2$ with energy $E \simeq -0.169$ and a sublattice magnetization $|\langle \mathbf{S}_i \rangle|$ of about 85% of the classical value. The apparently best available estimate of the true ground-state energy is $E_0 = -0.183 \pm 0.003$ from diagonalization of small clusters by Nishimori and Nakanishi[13] and Fujiki.[14] There does not appear to be a credible numerical estimate of the sublattice magnetization available; preliminary results of a perturbation-series approach[15] similar to that performed for the square lattice[5] suggest that the ground-state sublattice magnetization is in the vicinity of 50% to 60% of the classical value.

For the square lattice, as well as any other bipartite lattice, the amplitudes of the classical Néel-state wave function in our basis actually have the same relative phases (signs) as the true ground-state wave function; only the magnitudes differ. For the triangular lattice, which is *not* bipartite and on which the antiferromagnet is therefore frustrated, this is not the case and so we want to introduce additional variational parameters that adjust the imaginary part of $\tilde{H}$. Thus the "correct" $\tilde{H}$ that gives a ground-state wave function on the triangular lattice with sublattice magnetizations in the x-y plane will include, in addition to the imaginary single-spin term in (11), imaginary odd-spin terms. We assume that our wave function is invariant under the symmetry operations of the classical ground state given by (11). These symmetries include (i) real-space rotation by angle $2\pi/3$ about a lattice point, (ii) real-space reflection about a row of lattice points, (iii) a real-space translation by a nearest-neighbor vector and a simultaneous rotation in spin-space by angle $2\pi/3$ about the z axis, and (iv) a real-space rotation by angle $\pi/3$ about a point on the A sublattice and a simultaneous reflection in spin space across the x-z plane. We have chosen to include only the shortest-range three-spin term that appears. This term couples the nearest neighbors along a "dog leg":

$$\tfrac{1}{2}\tilde{H}_3 = iL \textstyle\sum_{ijk} \gamma_{ijk} S_i^z S_j^z S_k^z, \tag{12}$$

where the sum runs over all distinct triplets of three sites i,j,k, where both i and k are nearest neighbors of j, and sites i and k are second neighbors to one another, separated by distance $\sqrt{3}$. The sign factor $\gamma_{ijk} = \gamma_{kji} = \pm 1$ is invariant under rigid translations or rotations in real space by angle $2\pi/3$ of the three-spin cluster i,j,k, but changes sign under rotations by $\pi/3$ or π, as dictated by the above symmetries. If site j is on the A sublattice and sites i and k are on the B sublattice, then we choose $\gamma_{ijk} = +1$; the remaining γ_{ijk} follow. If we add just this second parameter L, thus letting $\tilde{H} = \tilde{H}_c + \tilde{H}_2 + \tilde{H}_3$ [(8), (11), and (12)], the variational wave function takes advantage of it: The minimum energy in the (K_1, L) plane is at $K_1 \simeq 1.45$, $L \simeq 0.25$, with $E \simeq -0.176$ and a sublattice magnetization of approximately 76% of the classical value. This three-spin term (12) is the only connected three-spin term permitted by the above symmetries of the wave function.

Finally, we have optimized a three-parameter variational wave function given by (9) with $K(r) = kr^{-\sigma}$ for all r, and $i\mathrm{Im}\tilde{H} = \tilde{H}_c + \tilde{H}_3$. The minimum energy occurs at $K \simeq 2.5$, $\sigma \simeq 1.8$, and $L \simeq 0.25$, where $E \simeq -0.1789$ and the sublattice magnetization is approximately 68% of the classical value. We feel this last result yields a useful (strict) upper bound on the triangular-lattice ground-state energy that can be readily improved by inclusion of more terms and more free parameters in $\tilde{H}$.

Various RVB-type wave functions have been proposed for the ground-state of the triangular-lattice spin-$\frac{1}{2}$ antiferromagnet.[6] These wave functions all have energy in the range -0.158 ± 0.005, which is clearly well above the true ground-state energy. In order to see how poor these RVB-type wave functions really are, it is worth noting that the classical energy is -0.125. Thus the nontrivial quantum part of the ground-state energy is approximately -0.06, and these RVB wave functions only

find a little more than half of this. Presumably, much improved RVB-type variational wave functions for this triangular-lattice system can be obtained by procedures similar to those used in Ref. 4.

Finally, we note that the qualitative behavior of the sublattice magnetization as our variational wave functions are improved is very similar for the square and triangular lattices. Thus similar three-parameter wave functions yield sublattice magnetizations of roughly 70% of the classical value in both cases. This should be contrasted to the linear chain where we know that the ground state has no sublattice magnetization,[11] and the variational wave function with only two parameters discussed above apparently tries to tell us this by going to very strong coupling and a sublattice magnetization of less than 20%. In view of this, and the preliminary series results,[15] it appears that the ground state of the triangular-lattice spin-$\frac{1}{2}$ antiferromagnet has long-range order with a sublattice magnetization not very much less than that of the square-lattice case.

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