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Variational calculations for the square-lattice quantum antiferromagnet

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We study the spin-$\frac{1}{2}$ Heisenberg antiferromagnet on a square lattice using variational Monte Carlo calculations. We construct a variational wave function which gives accurate ground-state energy and is consistent with sum rules for the dynamical structure function. The spin-wave velocity and the spin-stiffness constant can be calculated using sum rules. We obtain $Z_c \equiv c/c_0 = 1.22 \pm 0.02$, where c_0 is the classical spin-wave velocity. This and other ground-state quantities are in good agreement with spin-wave theory and Monte Carlo calculations.

The suggestion that the superconductivity mechanism in the copper-oxide materials may be understood by studying the two-dimensional Hubbard model in its strong-coupling limit[1] has recently received significant attention. The spin-$\frac{1}{2}$ antiferromagnetic (AF) Heisenberg model can be obtained from Hubbard models in the strong-coupling limit and at half filling and can describe the dynamics of the spin degrees of freedom in the undoped La_2CuO_4.[2] Even though the spin-$\frac{1}{2}$ AF Heisenberg model (AFHM): $H = J\sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$ lacks an exact solution for the square lattice which is the case of our interest, several systematic approaches, including analytical, semianalytical, and purely numerical techniques, support the picture suggested by spin-wave theory (SWT).[3] SWT assumes the existence of AF long-range order (LRO) in the ground state and treats the zero-point motion of small quantum fluctuations about the classical Néel state perturbatively.

To make contact with the phenomenological properties of the copper oxides, and further establish whether the spin-$\frac{1}{2}$ AFHM is a part of the microscopic Hamiltonian relevant for the physics of the materials, we need accurate calculations of its ground-state and excited-state properties. For instance, neutron scattering[4] and more recently high-energy neutron scattering[5] and Raman scattering experiments[6] performed on the undoped La_2CuO_4, determine the antiferromagnetic coupling J and the spin-wave velocity c. Linear SWT gives a value $c_0 = \sqrt{2}Ja$ for the spin-wave velocity while including the next correction one finds that $c/c_0 \simeq 1.158$.[3] In this paper we perform a variational Monte Carlo (VMC) calculation to determine among other ground-state quantities the spin-wave velocity accurately.

In a recent paper[7] the following ground-state wave function was derived using paired-magnon analysis of the spin-$\frac{1}{2}$ Heisenberg antiferromagnet:

$$|\psi_0\rangle = \Lambda \exp\left[-\tfrac{1}{2}\sum_{i<j} u_{ij}\sigma_i^z\sigma_j^z\right]|\phi\rangle, \tag{1a}$$

$$u_{ij} \equiv \frac{1}{N}\sum_{\mathbf{k}}\left[\left(\frac{1+\gamma(k)}{1-\gamma(k)}\right)^{1/2} - 1\right] e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)}, \tag{1b}$$

where $\gamma(\mathbf{k}) = \frac{1}{2}[\cos(k_x) + \cos(k_y)]$ and

$$|\phi\rangle \equiv \frac{1}{\sqrt{2^N}}\sum_c (-1)^{L(c)}|c\rangle. \tag{1c}$$

Here the sum is over all possible spin configurations c of the lattice and $L(c)$ is the number of down spins in one sublattice contained in the configuration c. Therefore the state $|\phi\rangle$ is

$$|\phi\rangle = \prod_{\mathbf{R}\in A}|\mathbf{R},+\rangle \prod_{\mathbf{R}\in B}|\mathbf{R},-\rangle$$

and the states $|\mathbf{R},+\rangle$ and $|\mathbf{R},-\rangle$ are the eigenstates of $\hat{S}^x_{\mathbf{R}}$. A and B represent the two sublattices. It can be easily verified that the state (1c) has zero staggered magnetization in the z and y directions but has full staggered magnetization in the x direction. In fact, if we rotate the Néel state around the y axis by $\pi/2$ we obtain the state (1c). A very similar wave function can be obtained[3] by taking into account the zero-point motion of spin waves in the spin-wave approximation with the aid of the Holstein-Primakoff transformation.[8]

Variational wave functions of similar form have been studied earlier by Hulthen[9] and Kastelijn[10] for one dimension and for one, two, and three dimensions by Marshall[11] and Bartkowski.[12] More recently the same form was studied by Huse and Elser[13] using the VMC approach. They took $u(1) = u_1$ and $u(r) = a/r^b$ for $r > 1$, where $r = |\mathbf{R}_i - \mathbf{R}_j|$ and treated u_1, a, and b as variational parameters. The energy per site obtained in this approach[13] is $\sim -0.664J$ for $u_1 \sim 0.65$, $a \sim 0.475$, and $b \sim 0.7$. Similar VMC studies were carried out by Horsch and Linden[14] where, using only $u(1)$ as a variational parameter [and $u(r > 1) = 0$], they found $-0.644J$ for the ground-state energy in our units. Notice that our u is not a function of the distance r between two points on the lattice but rather a function of the two components x and y of the vector $\mathbf{R}_{ij}$. The form (1) has long-distance behavior consistent with the existence of long-wavelength spin-wave excitations. From Eq. (1) we find that

$$u(r \to \infty) = \sqrt{2}/\pi r. \tag{2}$$

We notice that the tails of the wave function of Ref. 13

and that of Eq. (1) are quite different. The reason for this is the energy is not sensitive to the exact tail of the wave function. We have calculated the expectation value of the Heisenberg Hamiltonian with the wave function (1) using MC integration and restricting the sum in $|\phi\rangle$ [Eq. (1c)] over configurations having zero total S_z only. We found almost the same energy [slightly better (lower)] with that of Ref. 13. The advantage of the wave function (1) is its simple physical origin and the fact that we obtain the same ground-state energy with no free parameter. In this paper, we improve the wave function (1) further using sum rules of the dynamical structure function. We also propose a method to calculate the spin-wave velocity accurately.

Let us use a representation in which the eigenstates of the spin-$\frac{1}{2}$ AFHM are expressed as $|\Psi\rangle=\sum_c \psi(c)\times(-1)^{L(c)}|c\rangle$. The configuration $|c\rangle$ is labeled by the location of one kind of spin (say the down spins) on the lattice, i.e., $|c\rangle=|i_1,i_2,\ldots,i_r\rangle$ and the function $\psi(i_1,i_2,\ldots,i_r)$ gives the amplitude of that configuration in the wave function. The phase $(-1)^{L(c)}$ has been defined earlier and is separated from the amplitude ψ in order to have a non-negative ψ for any ground-state configuration.[11] In this representation, it is straightforward to show that the eigenvalue problem reduces to a difference equation, for the amplitude $\psi(i_1,i_2,\ldots,i_r)$, identical to the many-particle (the "particles" correspond to the down spins) Schrödinger equation on a two-dimensional (2D) lattice. In this quantum Boson lattice-gas the particles have "mass" $m=2$ (in the rest of this paper we work in units of J and lattice-spacing a) and interact via a pair potential V_{ij} having an infinite on-site repulsion, $V_{ij}=1$ if ij are nearest neighbors, otherwise $V_{ij}=0$. This representation was first used by Matsubara and Matsuda[15] to treat liquid helium using spin variables. It is a useful representation because our knowledge about the system of Bose-particles can be applied to the magnetic system also. For example, it is known that the ground state of this system has a broken symmetry (condensate) which in the magnetic language corresponds to AF-LRO. The elementary excitations in the Bose system are density fluctuations (phonons in the long-wavelength limit) which in the magnetic system correspond to spin waves. Chester and Reatto have shown[16] that the zero-point motion of the long-wavelength modes of the Bose system (zero sound) gives rise to a long-range tail in the Jastrow wave function. For a 2D system, we obtain

$$u(r\rightarrow\infty)=mc/4\rho_0\pi r\,, \tag{3}$$

where for the spin-$\frac{1}{2}$ system c is the spin-wave velocity, $m=2$. The ground state of the Heisenberg antiferromagnet has zero total S_z and the number of down spins is exactly half the total number of sites giving $\rho_0=\frac{1}{2}$. Comparing the tails (2) and (3) we find $c=\sqrt{2}$ which is the value found by linear SWT. Next, we discuss how to improve the wave function (1) and calculate the spin-wave velocity using sum rules.

We define the dynamic structure factor $S(\mathbf{q},\omega)$ for the AFHM as

$$S(\mathbf{q},\omega)=\sum_n |\langle n|S_q^z|0\rangle|^2\delta(\omega-\omega_{n0})\,, \tag{4}$$

where $|n\rangle$ is the complete set of the eigenstates of the Hamiltonian with $|0\rangle$ being the ground state and $S_{\mathbf{q}}^z=1/\sqrt{N}\sum_i S_i^z e^{i\mathbf{q}\cdot\mathbf{r}_i}$. The first two sum rules are

$$S(\mathbf{q})=\int_0^\infty S(\mathbf{q},\omega)d\omega=\langle 0|S^z_{-q}S^z_q|0\rangle\,, \tag{5}$$

$$\int_0^\infty \omega S(\mathbf{q},\omega)d\omega=\tfrac{1}{2}\langle 0|[S_{-q},[H,S_q]]|0\rangle = f\sum_\delta[1-\cos(\mathbf{q}\cdot\boldsymbol{\delta})]\,, \tag{6}$$

where

$$f=-\frac{J}{4}\langle 0|(S_i^+S_{i+\delta}^-+S_i^-S_{i+\delta}^+)|0\rangle\,. \tag{7}$$

We are going to show that $2f$ is the spin-stiffness constant ρ_s. We have obtained a third sum rule analogous to the compressibility sum rule in the case of quantum fluids which in the spin system is translated to "magnetic susceptibility sum rule:"

$$\frac{1}{2\varepsilon''}=\lim_{q\rightarrow 0}\int_0^\infty\frac{S(\mathbf{q},\omega)}{\omega}d\omega=\lim_{q\rightarrow 0}\sum_{n\neq 0}\frac{|\langle n|S_q^z|0\rangle|^2}{E_n-E_0}\,. \tag{8}$$

Imagine that we can study the variation of the ground-state energy per site $\varepsilon(M)$ as a function of the magnetization $M=1/N\langle 0|\sum_i S_i^z|0\rangle$. Here ε'' is the second derivative of $\varepsilon(M)$ with respect to M. We note that the magnetization corresponds to the particle density in the Bose system and $\varepsilon(M)$ to the ground-state energy as a function of density. The susceptibility per site χ for a field perpendicular to the staggered magnetization (in our case the staggered magnetization is along the [11] direction of the x-y plane) is given as $\chi=1/\varepsilon''$. We use units where $g\mu_B=1$ where μ_B is the Bohr magneton and g the g factor of the electron. The proof of (8) and more details will be published elsewhere.

In the long-wavelength limit ($q\rightarrow 0$) and for $\mathbf{q}$ along the [10] direction we find that

$$\lim_{q\rightarrow 0}\int_0^\infty \omega S(\mathbf{q},\omega)d\omega=fq^2$$

with $f\simeq 0.125J$. In this limit, assuming that the single-magnon excitation exhausts the sum rules and using the sum rules (5) and (6), we find

$$\omega_q=\frac{\int_0^\infty\omega S(\mathbf{q},\omega)d\omega}{S(\mathbf{q})}\,. \tag{9}$$

Hence, the spin-wave velocity is given by

$$c=f/s_1\,, \tag{10}$$

where s_1 is the slope of $S(q)$ in the long-wavelength limit.

Again assuming that the single-magnon excitation exhausts the third sum rules (6) and (8) we obtain

$$c=\sqrt{2f\varepsilon''}\,. \tag{11}$$

Notice that using $\varepsilon''=\chi^{-1}$, this equation becomes $c^2=2f/\chi$ and we therefore find that the spin-stiffness constant[17] $\rho_s=2f$. We calculate ε'' by restricting ourselves to a subspace with well-defined S^z_{tot}, i.e., total z component of the magnetization. The constant f can be calculated using (7) and therefore the spin-wave velocity can be

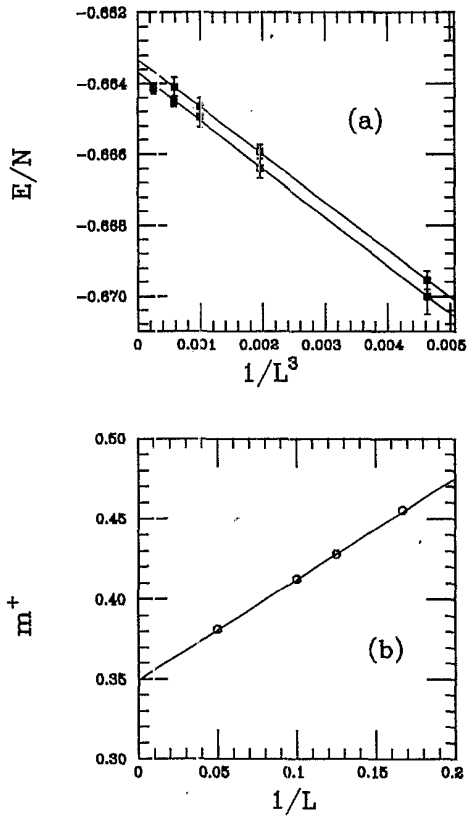

FIG. 1. (a) The energy per site as a function of L^{-3} for an L^2 lattice. The parameter-free wave function (1) gives the upper line and the improved wave function the lower line. Even though the simple wave function (1) and the improved wave function give the same energy within error bars, the improved is superior because it is consistent with the sum rules. (b) The staggered magnetization vs L^{-1}.

determined in a way analogous to that used in the case of quantum liquids to calculate the sound velocity. This technique is known to be accurate for numerical studies because it is not too sensitive to finite-size effects.

In the variational calculation we used the form (1a), including in the sum (1c) only states with zero magnetization, and took $u(1)$ and $u(\sqrt{2})$ as variational parameters and

$$u(\mathbf{r}) = a u_{\mathrm{LR}}(\mathbf{r}), \text{ for } (x^2+y^2)^{1/2} \geq 2, \tag{12}$$

where $u_{\mathrm{LR}}(\mathbf{r})$ is that given in Eq. (1b) and a is a parameter of order 1. We did not treat a as a variational parameter because the ground-state energy is not too sensitive to its precise value. Instead it is determined self-consistently by satisfying the third sum rule (11). Therefore, given a value of a we perform the variational calculation and determine $\varepsilon(M)$ and the spin-wave velocity c from the slope of $\varepsilon(M)$ via (11). Using the Chester and Reatto[16] relation (3) we obtain a new value of a from the relation $a = c/\sqrt{2}$. This can be iterated until the input and the output value of a are the same. This procedure converges very quickly since, as mentioned, the energy $\varepsilon(M)$ is not sensitive to the a. Starting from $a = 1$ the output value of a obtained from $\varepsilon(M)$ is ~ 1.2. In the next step using the new value of a as input we obtain practically the same output value from $\varepsilon(M)$.

The calculation is performed on lattices of several sizes up to 20^2. In Fig. 1(a) we present the ground-state energy per site as function of L^{-3} (Ref. 18) for lattices of size $N = L^2$. The energy obtained with the wave function (1) (upper line) and with the improved wave function (lower line) are the same within error bars. The optimal values of the variational parameters are $u(1) = 0.61$ and $u(\sqrt{2}) = 0.34$. The advantage of the improved wave func-

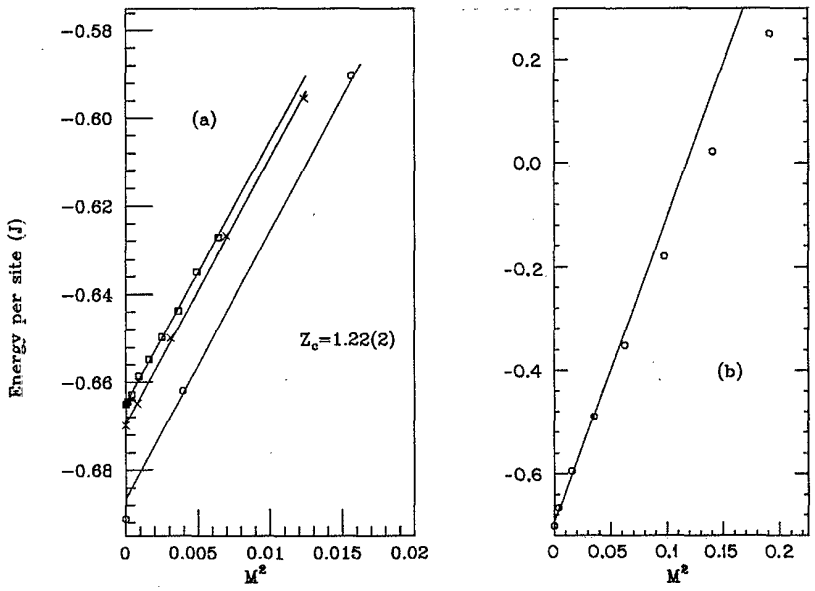

FIG. 2. (a) The energy per site $\varepsilon(M)$ as a function of the square of the total magnetization M for the spin-$\frac{1}{2}$ AF Heisenberg model for 4×4, 6×6, and 10×10 lattices (the results for the 20×20 size lattice are very close to those for the 10×10 on this scale). The slope is related to the spin-wave velocity via Eq. (11). (b) Exact diagonalization results for ε vs M^2 obtained on a 4×4 lattice. The solid line has the same slope as the curves in (a).

tion, however, is that it is consistent with the sum rules and gives an accurate excitation spectrum. The extrapolated value of the energy upper bound to the infinite-size lattice is $-0.6637J \pm 0.0002$, while the best estimate for the exact value obtained by Green's-Function Monte Carlo (GFMC) calculations[19,20] is $-0.6692J \pm 0.0001$. Figure 1(b) shows the square root of the expectation value of the square of the staggered magnetization obtained with the improved wave function. We obtain $m^{\dagger} = 0.349 \pm 0.002$ for the infinite lattice. This is close to the values of 0.34 ± 0.01 and 0.31 ± 0.01 reported in Refs. 19 and 20, respectively, while their guiding trial functions give a somewhat higher value of $m^{\dagger}$ than ours. In Fig. 2(a), we give $\varepsilon(M)$ for several lattice sizes. Notice that ε'' is independent of the lattice size within error bars and we obtain $\alpha = 1.22 \pm 0.02$. The value of this parameter (commonly called Z_c) is in good agreement with the value 1.158 obtained by spin-wave theory[3] and that obtained by GFMC (Refs. 19 and 20) calculations and series expansions.[21] The same value of ε'' is required to fit at small M the exact diagonalization results for a 4×4 size lattice [Fig. 2(b)] which have been obtained with methods explained in Ref. 22. Hence, we expect the error in the calculation of c due to the approximate nature of our wave function to be small. In Fig. 3, we give the calculated $S(\mathbf{q})$ using $\alpha = 1.22$ in the wave function. The straight line has a slope $s_1 = f/c$. The inset is the $\omega(\mathbf{q})$ calculated with the use of Eq. (9). The straight line in the inset has a slope $c = 1.22\sqrt{2}$. Hence, the spin-wave spectrum in the long-wavelength limit, although noisy as expected, is consistent with this sum rule within error bars also. In our units $Z_\chi = 8\chi J = 8J/\varepsilon'' = 0.667 \pm 0.004$ which is higher than the spin-wave value of 0.449 and close to that reported in Ref. 23.

Using our relation $c = 1.22\sqrt{2}Ja$, $a = 3.79$ Å and the value of c inferred from the most recent high-energy neutron scattering experiments[5] done on La_2CuO_4, we find $J = 0.13$ eV. In Ref. 2, in order to fit the correlation length as a function of temperature calculated from the same model to that measured by the thermal neutron scattering experiments,[4] we used a somewhat smaller value of $J \sim 0.11$ eV. Inversely, using the value of J found in Ref. 2, we find $c = 0.72$ eV Å which is between the experimental values reported in Refs. 4 and 5. This means that either the experimental value of c has been overestimated or the Heisenberg model may be too simple to accurately describe the physics behind the undoped La_2CuO_4 and another more realistic Hamiltonian may be necessary to account for the difference. It is interesting, however, that we can achieve this agreement using such a basic model.

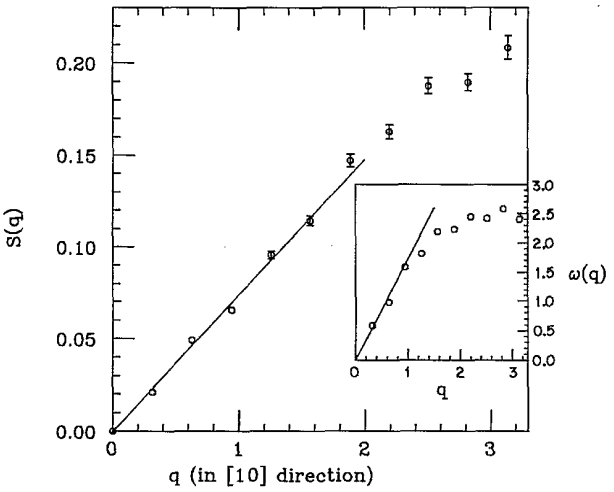

FIG. 3. The $S(\mathbf{q})$ calculated with the wave function (12) with $\alpha = 1.22$. The straight line has a slope $s_1 = f/c$. Therefore it is consistent with Eqs. (10) and (11). The inset is the $\omega(\mathbf{q})$ calculated with the use of Eq. (9). The straight line in the inset has a slope equal to $c = 1.22\sqrt{2}$ as obtained from (11).

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