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Monte Carlo solution of antiferromagnetic quantum Heisenberg spin systems

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A Monte Carlo method is introduced that overcomes the problem of alternating signs in Handscomb's method of simulating antiferromagnetic quantum Heisenberg systems. The scheme is applied to both bipartite and frustrated lattices. Results of internal energy, specific heat, and uniform and staggered susceptibilities are presented suggesting that quantum antiferromagnets may now be studied as extensively as classical spin systems using conventional Monte Carlo techniques.

The Monte Carlo technique has been proven to be effective in obtaining nonperturbative solutions to many-body problems. For example, the application of Monte Carlo to classical spin systems is well established.[1] This is not true, however, for quantum spin problems. Recent work in this area can be classified into two categories. In the first category, one maps a d-dimensional quantum problem onto a $(d+1)$-dimensional classical problem using Trotter's formula.[2] A slight disadvantage in this approach is that one must extrapolate to the limit of an infinite number of lattice sites in the time coordinate as well as in the physical spatial coordinates. In the second category[3,4] one uses Handscomb's scheme[5] to transform the original quantum problem into a classical averaging problem *exactly*. A difficulty arises with both methods, however, when one attempts to study *antiferromagnetic* Heisenberg models. In particular, with Handscomb's scheme, one is immediately faced with a severe obstacle: the problem of alternating signs.

In this paper we introduce a technique which overcomes the problems of alternating signs and generalizes Handscomb's approach to include antiferromagnets. We are thus left with a Monte Carlo method for calculating thermodynamic properties of quantum spin systems which is tractable and convenient for both ferromagnets and antiferromagnets.

For simplicity, let us concentrate on the *isotropic* Heisenberg model $H=2J\sum_{\langle ij\rangle}\vec{S}_i\cdot\vec{S}_j$. In terms of the spin permutation operators

$$H=J\sum_{\langle ij\rangle}P_{ij}-\frac{N_b}{2}J \quad . \tag{1}$$

Where $J>0$, N_b is the total number of bonds and P_{ij} permutes the spin states at sites i and j. To calculate the partition function, one expands $\exp(-\beta H)$ as a power series in (βH). The partition function is given by

$$Z=\sum_{n=0}^{\infty}(-1)^n\frac{(\beta J)^n}{n!}\sum_{C_n}\mathrm{Tr}(P_{m_1},\ldots,P_{m_n}) \quad , \tag{2}$$

where $m_j=1,\ldots,N_b$ and $C_n\equiv(P_m,\ldots,P_{m_n})$ is a particular string of n permutation operators. One can transform (2) into a Monte Carlo problem by first defining an abstract sample space $S=\{C_n;n=1,\ldots,\infty\}$. Equation (2) is then viewed as an ensemble average of $(-1)^n$ with the statistical weight of C_n given by

$$\pi(C_n)\equiv\frac{(\beta J)^n}{n!}\mathrm{Tr}(P_{m_1},\ldots,P_{m_n}) \quad ,$$

so that

$$Z=\sum_n\sum_{C_n}(-1)^n\prod(C_n) \quad . \tag{3}$$

The problem of alternating signs is evident in Eq. (3). Whereas the partition function is manifestly positive, it is expressed in terms of the difference of quantities which increase in magnitude as the temperature decreases and must, therefore, cancel to an increasing extent. Although *in principle* one could get the right answer from (3) if one would perform an arbitrarily large number of Monte Carlo steps, in practice stochastic sampling errors yield unacceptably large variances for any practical number of Monte Carlo steps. This has been pointed out by Lyklema[3] who had difficulty in getting good statistics at low temperatures even for a *ten* spin system. The aim of our work is to reexpress (2) such that all, or at least dominantly many, terms in the partition-function sum are positive.

Since the negative contributions in (3) come from those terms with an odd number of operators, the problem of alternating signs can be completely solved if we can reexpress $\vec{S}_i\cdot\vec{S}_j$ in terms of new operators such that only terms with even number of operators give nonzero trace. This can be achieved by first shifting the zero of the energy by $(N_b/2)J$. The new Hamiltonian is $H'=J\sum_{\langle ij\rangle}(P_{ij}-1)$. Now if we define $h_{ij}\equiv S_i^+S_j^-+S_i^-S_j^+$ it is possible to show $(P_{ij}-1)=h_{ij}-h_{ij}^2$, so that

$$-\beta H'=\beta J\sum_{\langle ij\rangle}(h_{ij}^2-h_{ij}) \quad . \tag{4}$$

The partition function is now given by

$$Z'=\sum_{n=0}\frac{(\beta J)^n}{n!}\sum_{C_n}(-1)^{n'}\mathrm{Tr}(O_{m_1},\ldots,O_{m_n}) \quad , \tag{5}$$

where $O_m=h_{ij}$ or h_{ij}^2, and n' is the number of unpaired h's in a particular string C_n. We note that when h_{ij} acts on sites i and j which are in opposite spin states, it interchanges their spins, and yields zero if the spins on site i and j are the same. Thus, the only nonzero terms in (5) are those for which the unpaired h's form closed loops. For any lattice in which nonintersecting closed loops have an even number of bonds, such as a bipartite lattice, all terms in (5) are positive. In other lattices, such as a triangular lattice in which odd loops arise, there are negative terms, but, in each order of (J/kT) the dominant contribution comes from those positive terms in which the h's are paired.

The trace of a particular string of h's is either zero or 2^{n_c}

where n_c is the number of clusters formed by connected lattice sites.[6] The trace is nonzero if and only if among all possible terms generated by multiplying out the h's there exists a term such that the following is satisfied. For each lattice site the number of S^+ and the number of S^- appearing in the string are equal and for each lattice site, S^+ and S^- appear alternately. In the presence of an external magnetic field H, the same quantity is replaced by

$$\mathrm{Tr}(O_{m_1}, \ldots, O_{m_n}) = \prod_{\alpha=1}^{n_c} 2\cosh\left(\frac{q\mu_B}{2} a_\alpha H\right) . \tag{6}$$

where $\frac{1}{2}q\mu_B$ is the magnetic moment of each spin and a_α is the "effective moment"[7] of the αth cluster. Equation (6) can be interpreted as the partition function of n_c independent renormalized spins of magnetic moment $(q\mu_B/2)a_\alpha$. The Monte Carlo calculation sums over all nonzero traces of the strings formed by h's which correspond to different ways of grouping the "bare spins." The average energy, magnetization, zero field susceptibility, and specific heat are given by

$$\begin{aligned} E(H) &= -HM(H) - \frac{1}{\beta}\langle n\rangle + \frac{N_b}{2}J , \\ M(H) &= \left\langle \sum_\alpha \frac{q\mu_B}{2} a_\alpha \tanh\left(\frac{q\mu_B}{2} a_\alpha H\right)\right\rangle , \\ \chi_0 &= \beta\left\langle \sum_\alpha \left(\frac{q\mu_B}{2} a_\alpha\right)^2\right\rangle , \\ C_H &= k_B\left(\langle n^2\rangle - \langle n\rangle^2 - \langle n\rangle\right) , \end{aligned} \tag{7}$$

where $\langle\ \rangle$ represents the Monte Carlo average. In carrying out the random walk, we allow the addition or removal of N_b operators at each Monte Carlo step in order to guarantee that the random walk path covers the entire sample space. This generalization is essential when we do calculations on a triangular lattice. Each Monte Carlo step in our calculation, therefore, consists of $N \leq N_b$ consecutive "nearest-neighbor hops." (In each nearest-neighbor hop only one operator is added or removed) and the transition probability is given by

$$\Gamma(i, i_1, \ldots, i_{n-1}, j) = \frac{1}{N_b} P_{ii_1}, \ldots, P_{i_{n-1}j} ,$$

where $P_{i_m i_n}$ is the nearest-neighbor transition probability. For convenience $P_{i_m i_n}$ is constructed so that the detailed balance principle is satisfied for each nearest-neighbor hop and the expression can be found in Refs. 3 and 4.

We have tested our method by comparison to exact calculations on linear spin chains. The results obtained by running maximally 10^5 Monte Carlo steps[8] are listed in Table I. The numbers in parentheses are obtained by direct diagonalizations. The statistical error in our calculations is estimated by dividing the entire run into $\simeq 20$ bins and calculating the standard deviation of the bin averages with respect to the total average. It is significant that good statistics are obtained for all observables including even the specific heat at very low temperatures with modest Monte Carlo runs.

Having this positive confirmation of the method we now apply it to study the square lattice. We calculate the evarage energy, specific heat, and the staggered susceptibility for 4×4, 8×8, 16×16, and 32×32 lattices under periodic boundary conditions. The results are obtained by averaging over maximally 6×10^6 Monte Carlo steps and are shown in Fig. 1. To make the figures legible we have only included the largest lattice results available at each temperature. The solid curves represent the results of high-temperature series (HTMP) expansion and the dashed curves are drawn merely to guide the eye. In Fig. 1(a) we show the average energy and the inverse staggered susceptibility for temperature from $kT/J = 0.5$ to $kT/J = 5.0$. In the same units the extrapolated ground-state energy obtained by Oimaa and Betts[9] is 1.31 ± 0.01. The staggered susceptibility diverges as the temperature is lowered. From the present calculation we cannot tell whether a nonzero temperature transition exists, but from the size dependence studied so far this seems un-

TABLE I. Comparison of stochastic and direct diagonalization results (enclosed in parenthesis) for internal energy, specific heat, and magnetic suspectibilities for $N = 10$ and $N = 32$ spin linear chains.

K_BT/J	$-U/NJ$	C_H/NK	$4JX/g\mu_B$
$N=10$			
5.0	0.159 ± 0.008 (0.162)	0.034 ± 0.004 (0.034)	0.161 ± 0.001 (0.161)
4.0	0.193 ± 0.009)0.204)	0.052 ± 0.007 (0.053)	0.190 ± 0.001 (0.189)
3.0	0.272 ± 0.010 (0.275)	0.093 ± 0.008 (0.094)	0.228 ± 0.001 (0.227)
2.0	0.412 ± 0.009 (0.409)	0.188 ± 0.014 (0.189)	0.273 ± 0.001 (0.273)
1.0	0.688 ± 0.008 (0.683)	0.341 ± 0.028 (0.353)	0.288 ± 0.002 (0.288)
0.8	0.751 ± 0.088 (0.754)	0.337 ± 0.058 (0.346)	0.269 ± 0.005 (0.274)
0.6	0.826 ± 0.008 (0.819)	0.317 ± 0.060 (0.296)	0.248 ± 0.005 (0.250)
0.4	0.864 ± 0.008 (0.870)	0.239 ± 0.060 (0.206)	0.199 ± 0.013 (0.198)
$N=32$			
0.4	−0.845 ± 0.020 (−0.855)	0.195 ± 0.040 (0.178)	0.246 ± 0.005 (0.240)
0.6	−0.825 ± 0.020 (−0.810)	0.250 ± 0.040 (0.275)	0.260 ± 0.002 (0.260)
0.8	−0.745 ± 0.015 (−0.755)	0.325 ± 0.030 (0.345)	0.279 ± 0.002 (0.277)
1.0	−0.675 ± 0.015 (−0.690)	0.340 ± 0.030 (0.350)	0.292 ± 0.002 (0.290)
2.0	−0.405 ± 0.010 (−0.410)	0.215 ± 0.020 (0.193)	0.270 ± 0.001 (0.272)

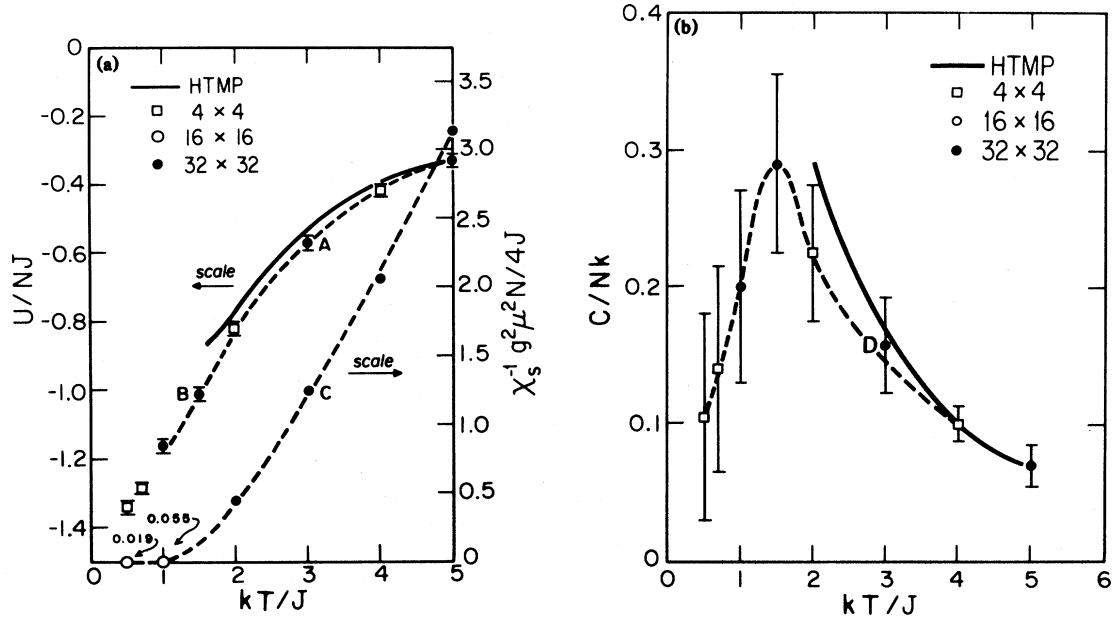

FIG. 1. Thermodynamic properties on a square lattice. The points represent different size lattices, the dashed lines are meant to guide the eye, and the solid lines represent high-temperature series expansion results. (a) Average energy and the staggered susceptibility as a function of temperature; (b) specific heat as a function of temperature.

likely. In Fig. 1(b) we show the specific heat. Clearly, fluctuations are much more severe and longer Monte Carlo runs are required to improve the statistics. The specific heat peaks around $kT/J = 1.5$ and the peak value saturates at about $0.3k_B$ per spin. In Fig. 2 we show the size dependence of the various quantities indicated as *A*, *B*, *C*, and *D* in Figs. 1(a) and 1(b).

Analogous calculations on the triangular lattice produced the interesting results shown in Fig. 3. Here, we plotted the inverse uniform susceptibility as a function of temperature

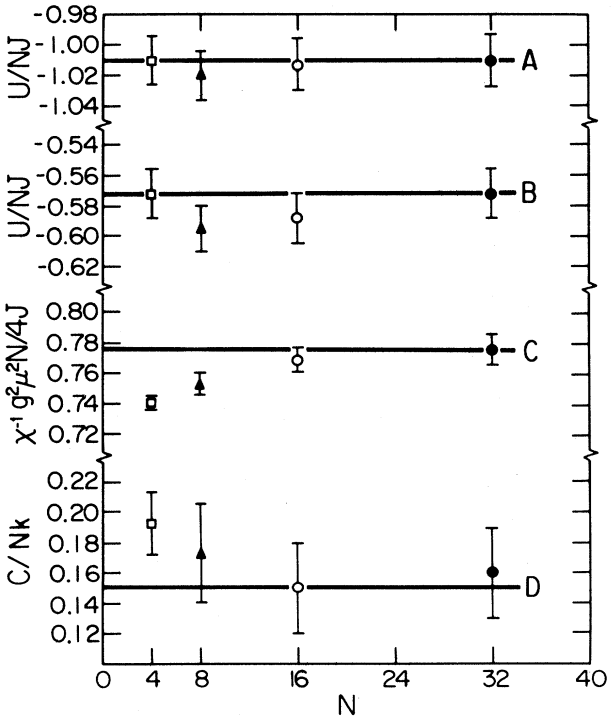

FIG. 2. Size dependence of various quantities for the square lattice. The labels *A*, *B*, *C*, and *D* correspond to the points marked the same way in Figs. 1(a) and 1(b). The solid lines indicate the values represented by the dashed curves in 1(a) and 1(b).

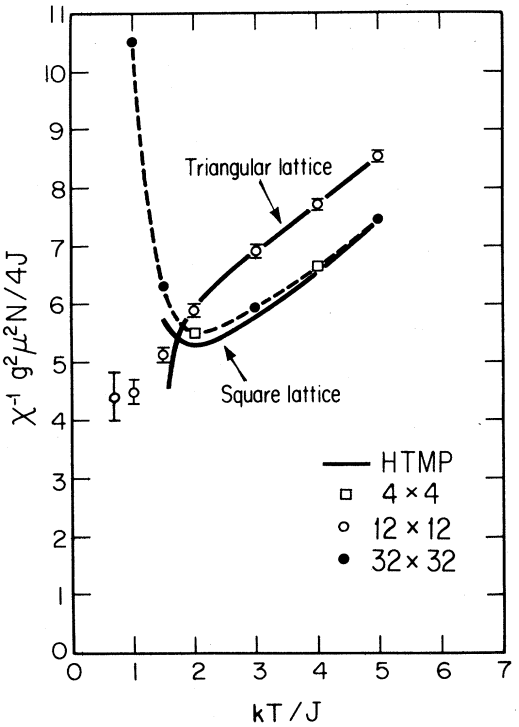

FIG. 3. In this figure we show the inverse uniform susceptibility for square and triangular lattices. The solid curves represent series-expansion results and the dashed curve is drawn to guide the eye.

for both square (nonfrustrated) and triangular (frustrated) lattices showing that the low-T behavior of these two models differs qualitatively. For the square lattice, the susceptibility decreases rapidly as kT/J is lowered below 2.0 indicating an "activated behavior." However, the gap is found to scale as $1/\sqrt{N}$ where N is the total number of spins. For the triangular lattice the uniform susceptibility is less sensitive to temperature at low T and the crossover to "activated behavior" is not seen down to $kT/J=0.75$. This could mean that there is a much smaller gap or much larger entropy (greater degeneracy) associated with the excited states. Although the explanation of this unusual behavior is not exactly clear to us, we speculate that it may be a consequence of the large entropy associated with the existence of additional elementary excitations in frustrated lattices.[10] Much further work is needed before this can be understood completely.

In conclusion, we have developed a Monte Carlo scheme which now makes the study of both quantum antiferromagnets and ferromagnets as tractable as classical spin systems using conventional Monte Carlo methods. Although we have concentrated on the isotropic antiferromagnet for simplicity, the generalization to the anisotropic case is straightforward. For example, given

$$H=2\sum_{\langle ij\rangle}(J_x S_i^x S_j^x+J_y S_i^y S_j^y+J_z S_i^z S_j^z)\ , \tag{8}$$

with $J_x\neq J_y\neq J_z>0$ the zero of energy can be chosen to be $(J_z/2)N_b$ and the new Hamiltonian becomes

$$H'=\sum_{\langle ij\rangle}\left[-J_z h_{ij}^2+\frac{(J_x+J_y)}{2}h_{ij}\right]\ . \tag{9}$$

Thus, the algorithm for evaluating the partition function

$$Z'=\sum_{n=0}^{\infty}\frac{1}{n!}\sum_{C_n}(-1)^{n'}(\beta J_z)^{n-n'}\times\left[\beta\frac{(J_x+J_y)}{2}\right]^{n'}\mathrm{Tr}(O_{m_1},\ldots,O_{m_n})\ . \tag{10}$$

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[2]See, e.g., J. J. Cullen and D. P. Landau, Phys. Rev. B **27**, 297 (1983); M. Suzuki, Prog. Theor. Phys. **56**, 1454 (1976).

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[4]S. Chakravarty and D. B. Stein, Phys. Rev. Lett. **49**, 582 (1982).

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[6]Two lattice sites i,j are connected if h_{ij} appears in the particular string. A single isolated lattice site is considered to be a cluster by itself.

[7]The "effective moment" a_α of the αth linked part is given by $a_\alpha\equiv|n_+^\alpha-n_-^\alpha|$, where n_+^α (n_-^α) is the number of lattice sites in that linked part for which the first operator appearing in the string is S^+ (S^-).

[8]One Monte Carlo step in our method should be compared with the flipping of one spin in classical Monte Carlo calculations.

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