Quantum Monte Carlo and transfer-matrix calculations for one-dimensional easy-plane ferromagnets

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We have applied previously used quantum Monte Carlo (QMC) techniques to obtain numerically the thermodynamics of two well-studied quasi-one-dimensional (1D) easy-plane ferromagnetic models, in the presence of an applied magnetic field in the easy plane. The checkerboard decomposition form of the Trotter approximation to the partition function has been used. Internal energy, specific heat, magnetization, and susceptibility have been obtained for model Hamiltonians believed appropriate for spin $S=\frac{1}{2}$ [$(C_6H_{11}NH_3)CuBr_3$ (CHAB)] and $S=1$ ($CsNiF_3$), in temperature and field ranges where classical theories have predicted solitonlike kink excitations. The $S=\frac{1}{2}$ QMC calculations are verified and superseded by a numerically exact quantum transfer-matrix (QTM) technique. Results for the temperature dependence of the peak in the specific heat versus field are compared with available experimental results. For the model applicable to CHAB, it is found that there is no value of the easy-plane anisotropy parameter from 4% to 10% for which the QTM calculation can adequately reproduce the experimentally obtained peak height and position. On the other hand, the QMC results for the model assumed for $CsNiF_3$ do roughly reproduce the temperature dependence of the experimental peak positions, but not the peak heights. However, statistical errors present in our QMC data are large, and a better method is still needed for computing the quantum statistical mechanics of $S=1$ systems.

I. INTRODUCTION

Obtaining a correct theoretical description of the low-temperature thermodynamics of the easy-plane spin-1 ferromagnet $CsNiF_3$ has been the subject of a number of investigations in recent years, both theoretical[1–6] and experimental.[7–10] More recently, a similar spin-$\frac{1}{2}$ compound, $(C_6H_{11}NH_3)CuBr_3$, or CHAB, has inspired even further interest in easy-plane ferromagnets (EPF's), especially because of the possible existence of (or effective consequences of) classical solitary wave excitations in such a low-spin quantum chain.[11] A typical model Hamiltonian for a spin-1 EPF is[1]

$$\hat{H}_{S=1}=\sum_{n=1}^{N}\left[-J\mathbf{S}_n\cdot\mathbf{S}_{n+1}+A(\hat{S}^y_n)^2-g\mu_B\mathbf{B}\cdot\mathbf{S}_n\right], \quad (1)$$

where $J=23.6$ K, $A=9.0$ K, and $g=2.4$ for $CsNiF_3$.[8] N is the number of spins and μ_B is the Bohr magneton. For a spin-$\frac{1}{2}$ EPF, the easy-plane anisotropy must be in the exchange, then the Hamiltonian is usually taken to be

$$\hat{H}_{S=1/2}=\sum_{n=1}^{N}\left[-(J_x\hat{S}^x_n\hat{S}^x_{n+1}+J_y\hat{S}^y_n\hat{S}^y_{n+1}+J_z\hat{S}^z_n\hat{S}^z_{n+1})-g\mu_B\mathbf{B}\cdot\mathbf{S}_n\right]. \quad (2)$$

For CHAB, we take[12] $J_x=J_z=110$ K, $J_y=104.5$ K, and $g=2.0$. In both Hamiltonians, we will find it convenient to choose the xz plane as the easy plane, and to apply the field in the z direction.

Because of the low spin number, quantum effects might be expected *a priori* to be strong in both these materials. However, it has been customary to apply classical mechanics as a first approximation. With a field in the easy plane which is small in comparison to the anisotropy ($g\mu_BB\ll 2AS$), and at low temperatures ($T\lesssim J$), the continuum limit classical dynamics is approximately described by the sine-Gordon (SG) equation.[1] Then the possible excitations include small-amplitude spin waves, solitons, and breathers. Early neutron-scattering experiments on $CsNiF_3$ (Refs. 7 and 8) and tetramethyl ammonium manganese trichloride (TMMC, a similar antiferromagnet with $S=\frac{5}{2}$; Ref. 13) were interpreted in terms of a gas of weakly interacting solitons and spin waves. More recently this interpretation has been challenged and the need to include alternative mechanisms[4–6] has been discussed—for example, including higher-order spin-wave processes. Also, the soliton-gas model[14] predicts a peak in the soliton specific heat versus field at fixed temperature, whose position and height are proportional to T^2 and T, respectively. The T^2 dependence of the peak position is approximately observed in experiments on CHAB and $CsNiF_3$, but the constants of proportionality are only correct if one assumes an *ad hoc* renormalization of the soliton rest mass. There is no consistent fit for the peak heights in either material.

Furthermore, linear stability analyses[15,16] and numerical simulations of the full classical dynamics[17] (without approximating the equations of motion by the SG equation) have shown that there is an intrinsic instability for the solitons, henceforth referred to as kinks, to deviate strongly from SG-like behavior for applied fields greater than a critical field given by $g\mu_BB_c=2AS/3$. As the field is increased toward the critical field, the spins show an increasing tendency to tilt out of the easy plane, a

motion which is assumed small in the SG approximation. The critical fields are around 18 kG for $CsNiF_3$ and 13 kG for CHAB, somewhat above the ranges where most experiments have been done. This spin tilting increases continuously as the field approaches the critical field, and therefore its effects can generally play a role in the dynamics. For fields greater than the critical field, the kinks move in a direction opposite to that expected of SG solitons; these have been referred to as "backwards" negative effective mass kinks.[17] It is only in the limit of small field and zero velocity that the out-of-plane tilting vanishes, and then the SG approximation becomes exact. Otherwise, the complete *classical* dynamics is rather poorly described by the SG equation. This behavior of the kinks also has been obtained by a variational ansatz calculation.[18]

The classical EPF Hamiltonian has been further shown to be inadequate for explaining experimental data through classical transfer-matrix (TM) calculations. In particular, TM calculations[6] of specific heat using the full discrete EPF Hamiltonian give results much higher than experiment[10] for $CsNiF_3$, whereas SG theory (without a renormalized rest mass) fits much closer to experiment. Similar results have been found for easy-plane antiferromagnets[19] comparing experiment with classical Monte Carlo results. It has been suggested that quantum mechanics could effectively restrict spins to the easy plane (tilted by zero-point fluctuations), thereby making SG theory more appropriate than the full classical Hamiltonian, even for $S=\frac{1}{2}$ CHAB.[20]

There could be several corrections necessary to the classical model, including effects of next-nearest-neighbor interactions, impurities, or discreteness. Perhaps even a quite different Hamiltonian is necessary, especially since parameters are usually determined from fits to *linear* properties, whereas soliton-bearing systems exhibit intrinsically *nonlinear* phenomena. Certainly the most obvious question to consider, however, is how to include quantum mechanics in the model. One approach has been to reinsert the quantum mechanics by simply replacing the classical SG equation with its quantized version, then the leading correction to the classical theory is a reduction of the SG soliton rest mass.[21] More recently, Johnson and Wright[22] reported on the Bethe ansatz method[23] applied to solving the quantized SG equation relevant to easy-plane ferro- and antiferromagnets—a similar rest-mass reduction is found, but still theory and experiment for CHAB, $CsNiF_3$, and TMMC disagree (for specific heat, and therefore probably for other thermodynamic properties). These authors point out, in particular, that the corrected classical SG theory, including kink-kink interactions,[24] would require a rest mass *increase* to bring the calculated specific heat into agreement with experiment for CHAB. This approach of quantizing a particular limit of the full classical Hamiltonian (the SG limit) seems questionable. By so doing, the out-of-plane degree of freedom is not treated properly; it is essentially transformed to a linear degree of freedom. In view of continuing controversies over the importance of out-of-plane classical motions[25] versus the quantization of the SG model, it seems necessary to include *both* out-of-plane and quantum aspects simultaneously.

One way of achieving this is to use the recently developed Trotter-Suzuki transformation, whereby the thermodynamics of the original one-dimensional (1D) *quantum* system is mapped onto the thermodynamics of a 2D *classical* system.[26] Numerical evaluation of the internal energy, specific heat, etc., is carried out by using either Monte Carlo or transfer matrix methods.[27] Although this will give no direct information about the excitations (e.g., the question of existence of solitons) it can nevertheless give crucial indications of the importance of quantum effects and the validity of the assumed Hamiltonians.

In this paper we begin with a brief review of the Trotter-Suzuki formalism, which converts the trace operation in the partition function definition into a discrete path integral, thereby adding one dimension. New Monte Carlo spin-1 updating algorithms will be given,[28] these being somewhat different from the previous spin-$\frac{1}{2}$ schemes.[27] Results relevant to the $CsNiF_3$ model will be presented, and compared with experiment and SG theory. Unfortunately, these $S=1$ data have rather large statistical errors making quantitative comparisons of limited value. Methods other than the present quantum Monte Carlo (QMC) technique may prove to be more precise. One possibility is the "numerically exact" quantum transfer-matrix (QTM) method as studied by Betsuyaku,[29] which will be applied here to the spin-$\frac{1}{2}$ CHAB thermodynamics. We use a technique to extrapolate from the finite-size lattice to the infinite limit in both directions on the 2D lattice, thereby making this preferred over the previous $S=\frac{1}{2}$ QMC method.[30] We find that there is *no* value of exchange anisotropy from 4% to 10% for which the QTM results for specific-heat peaks will agree with experiment. We have, however, tested that the QTM calculation gives results consistent with the QMC calculation. Finally, a method for calculating the $S=1$ fundamental matrix elements is sketched in the Appendix;[28] these matrix elements determine the effective energy of the 2D lattice, and therefore control the Monte Carlo updating.

II. TROTTER-SUZUKI FORMALISM: CHECKERBOARD DECOMPOSITION

The original 1D quantum thermodynamics spin problem is mapped onto an approximately equivalent 2D classical thermodynamics problem via an application of a generalized Trotter formula, as suggested by Suzuki.[26] First, the partition function Z is defined in terms of a trace

$$Z=\mathrm{tr}(e^{-\beta\hat{H}})=\sum_{\sigma}\langle\sigma\,|\,e^{-\beta\hat{H}}\,|\,\sigma\rangle\ , \tag{3}$$

where β is the reciprocal of the temperature T (we use Boltzmann's constant $k_B=1$) and $\sigma=\{S_n,\ n=1,2,3,\ldots,N\}$, where the S_n are eigenvalues of some appropriately chosen operators, usually $\hat{S}^z_n$. It is generally not known how to compute the required matrix elements in Eq. (3), so the Trotter formula[31] is used to approximate the operator by one for which the matrix elements are easier. The generalized Trotter formula for the

exponential of a sum of k operators is

$$\exp\left[\sum_{i=1}^{k}\hat{\theta}_i\right]=\lim_{m\to\infty}\left[\prod_{i=1}^{k}\exp(\hat{\theta}_i/m)\right]^m . \tag{4}$$

Typically the integer k is N for what has been called[32] the "real space decomposition," and k can be 2 or 4 or even 6 for the "checkerboard decomposition." The integer m is referred to as the "Trotter index," and represents the number of discrete path integral steps.

The Hamiltonian is written as a sum of two-body operators $H_{n,n+1}$, and it is convenient to assume that each of these two-body operators can be written as a sum of two parts

$$\hat{H}=\sum_{n=1}^{N}\hat{H}_{n,n+1} , \tag{5a}$$

$$\hat{H}_{n,n+1}=\hat{H}^{0}_{n,n+1}+\hat{V}_{n,n+1} . \tag{5b}$$

Furthermore, the odd and even n two-body operators are summed separately to define four parts of $\hat{H}$ (i.e., $k=4$);

$$\hat{H}^{0}_{\substack{\text{odd}\\\text{even}}}=\sum_{\substack{n=\text{odd}\\\text{even}}}\hat{H}^{0}_{n,n+1},\quad \hat{V}_{\substack{\text{odd}\\\text{even}}}=\sum_{\substack{n=\text{odd}\\\text{even}}}\hat{V}_{n,n+1} , \tag{6a}$$

$$\hat{H}=\hat{H}^{0}_{\text{odd}}+\hat{V}_{\text{odd}}+\hat{H}^{0}_{\text{even}}+\hat{V}_{\text{even}} . \tag{6b}$$

The mth-order Trotter approximation to the partition function, $Z^{(m)}$, is then defined by

$$Z^{(m)}=\mathrm{tr}\{[(e^{-\beta\hat{H}^{0}_{\text{odd}}/m}e^{-\beta\hat{V}_{\text{odd}}/m})\times(e^{-\beta\hat{H}^{0}_{\text{even}}/m}e^{-\beta\hat{V}_{\text{even}}/m})]^m\} . \tag{7}$$

All four operators $\hat{H}^{0}_{\text{odd}}$, etc., are sums of commuting operators, and so this is rewritten as

$$Z^{(m)}=\mathrm{tr}[(\hat{L}_{\text{odd}}\hat{L}_{\text{even}})^m] , \tag{8a}$$

where

$$\hat{L}_{\substack{\text{odd}\\\text{even}}}=\prod_{\substack{n=\text{odd}\\\text{even}}}(e^{-\beta\hat{H}^{0}_{n,n+1}/m}e^{-\beta\hat{V}_{n,n+1}/m}) . \tag{8b}$$

Now, $(2m-1)$ complete sets of states are introduced (in addition to that of the trace itself) labeled by $\sigma_r=\{S_{n,r}, n=1,2,3,\ldots,N\}$, $r=1,2,3,\ldots,2m$. The eigenvalues now have both a position index n and a state index r. Then

$$Z^{(m)}=\sum_{\sigma_1,\sigma_2,\ldots,\sigma_{2m}}\langle\sigma_1|\hat{L}_{\text{odd}}|\sigma_2\rangle\langle\sigma_2|\hat{L}_{\text{even}}|\sigma_3\rangle\cdots\times\langle\sigma_{2m}|\hat{L}_{\text{even}}|\sigma_1\rangle . \tag{9}$$

From (9) it is evident that we have an expression for a classical partition function on a 2D lattice of size $N\times 2m$:

$$Z^{(m)}=\sum_{\sigma_1,\sigma_2,\ldots,\sigma_{2m}}\prod_{\langle n,r\rangle}e^{-\beta E(n,r)} . \tag{10}$$

Here brackets $\langle n,r\rangle$ indicate a product restricted to terms in which n and r are both odd or both even, and the 2D energy function $E(n,r)$ for a block of four spins on the lattice is given by the fundamental matrix element

$$e^{-\beta E(n,r)}=\langle S_{n,r}S_{n+1,r}|e^{-\beta\hat{H}^{0}_{n,n+1}/m}e^{-\beta\hat{V}_{n,n+1}/m}\times|S_{n,r+1}S_{n+1,r+1}\rangle . \tag{11}$$

The r variable is the new added dimension. Since the only terms which contribute to the energy of the 2D lattice are restricted to n and r both odd or both even, this has been called a checkerboard decomposition. In what follows the $S_{n,r}$ variables will be eigenvalues of $\hat{S}^{z}_{n}$ operators, for the rth set of states. In the spin-$\frac{1}{2}$ problem these can be $\pm\frac{1}{2}$, while for spin-1 the possibilities are ±1 and 0.

The 2D lattice consists of $\frac{1}{2}Nm$ blocks of four spins, or "vertices." In the limit $m\to\infty$, $Z^{(m)}$ approaches the exact partition function of the original 1D quantum system. The 2D lattice has periodic boundary conditions in the r (or Trotter) direction, as a result of the trace operation. For the Monte Carlo calculations, periodic boundary conditions will also be imposed in the spatial direction. For the transfer matrix calculations, however, it is very advantageous to use free end boundary conditions in the spatial direction.

Expression (11) needs to be modified slightly, since the resulting matrix elements will not in general be symmetric with respect to the interchange of r and $r+1$ (i.e., the matrix is non-Hermitian). We redefine $E(n,r)$ and restore this symmetry:

$$e^{-\beta H(n,r)}=\langle S_{n,r}S_{n+1,r+1}|e^{-\beta\hat{V}_{n,n+1}/2m}e^{-\beta\hat{H}^{0}_{n,n+1}/m}e^{-\beta\hat{V}_{n,n+1}/2m}|S_{n,r+1}S_{n+1,r+1}\rangle . \tag{12}$$

This is equivalent to using $k=6$ in the Trotter formula. These matrix elements (or vertex weights) for the spin-1 model are calculated in the appendix. Spin-$\frac{1}{2}$ matrix elements have already been given elsewhere.[26-30]

The properties of the spin-$\frac{1}{2}$ matrix elements have also been discussed.[26-30] It is important to note that zero matrix elements correspond to infinite energies and therefore prohibited states of the four-spin blocks or vertices. For the spin-1 problem, out of the possible $(2S+1)^4=81$ matrix elements, only 19 are nonzero for the isotropic case in the absence of a field ($A=B_z=0$). If a field is added parallel to the quantization axis, there will still be only 19 allowed vertices. However, if a field is added perpendicular to the quantization axis, this produces an 81 vertex model. By choosing the quantization axis in the easy plane (when $A\neq0$), parallel to the field, one obtains a 41-vertex model, independent of the size of the field. Since we are interested in studying the field dependence of the thermodynamics, it is most convenient to choose this last case, so that the model under consideration always has 41 allowed vertices. In this way, updating acceptance rates will depend only weakly on the field strength for any

chosen spin updating algorithm. For this 41-vertex model, the allowed vertices are the ones which contain an even number of zeros (or ± 1's), and this influences the choice of an appropriate Monte Carlo updating algorithm.

III. MONTE CARLO SPIN UPDATING: $S=1$

At each of the $N \times 2m$ sites of the 2D lattice, there is a classical spin variable whose value can be -1, 0, or $+1$. Starting from any given allowed state of the system, an algorithm is needed which has equal *a priori* probability of transition to any other allowed state. Then the new state will be accepted or rejected according to the usual Metropolis *et al.* scheme:[33] If ΔE is the energy change of the 2D lattice, the acceptance probability is 1 if $\Delta E \leq 0$ and $e^{-\beta \Delta E}$ if $\Delta E > 0$. One can follow the example of spin-$\frac{1}{2}$ QMC and generate trial columns, rows, or squares (in the "holes" of the checkerboard) along which spins are altered.[27] One simply flips $+\frac{1}{2}$ to $-\frac{1}{2}$, or vice versa, at each site along the trial path; there is only one possible output state (plus the identity). For this spin-1 problem, however, the situation is more complicated—there are many choices for possible output states of the selected path, and they all need to be weighted equally, as follows.[28]

A move is considered which will alter p spins along some closed path in the lattice. That the path must be closed is necessitated by the choice of a 41-vertex model. One has $p=N$ for a row, $p=2m$ for a column, and $p=4$ for a square. Any nearest-neighbor pair of spins in the path belongs to one vertex (block of four spins) in the lattice. If the number of 0's in the pair is odd (even) before the attempted move, then within the 41-vertex model it must remain odd (even) after the move, since the only allowed vertices have an even number of 0's. This restriction limits the moves to two types:

(i) "unflipped;" all 0's in the path remain 0; all ± 1's in the path go to $+1$ or -1 with equal probabilities.

(ii) "flipped;" all 0's in the path go to $+1$ or -1 with equal probabilities; all ± 1's in the path go to 0.

The decision whether to attempt the flipped or unflipped move is based on the number of zeros, N_0, along the path before the move. For a given initial path, there are 2^{N_0} flipped moves possible, and 2^{p-N_0} unflipped ones, for a total of $2^{N_0}+2^{p-N_0}$ possible output states. Therefore the flipped moves should be attempted with probability $2^{N_0}/(2^{N_0}+2^{p-N_0})$, and the unflipped moves should be attempted with probability $2^{p-N_0}/(2^{N_0}+2^{p-N_0})$. In the absence of any interactions (all vertex weights equal), we have tested that this algorithm generates equal numbers of -1's, 0's, and $+1$'s in the lattice, using an equal number of row, column, and square moves at randomly chosen locations.

IV. MONTE CARLO DETAILS

We used an approximately constant value of $mT \approx 60$ K, in order to make the errors due to the Trotter approximation reasonably independent of temperature. This necessitates a larger lattice in the Trotter direction at lower T. For the $CsNiF_3$ parameters, in the temperature range 5 K $< T <$ 15 K and field range $0 \leq B_z \leq 10$ kG (in the easy plane), acceptance rates for row and column moves are $\ll 1\%$, while square moves have larger acceptance rates $\approx 10\%$. Because of the inefficiency of this method compared to spin-$\frac{1}{2}$, we present data here for only 16 spins, using only square moves.

The initial configuration was taken to be the state with all $S_{n,r}=0$. Vertex weights were found for a temperature $2T$, and then the Monte Carlo algorithm was applied for 3000 to 5000 "steps," where a step involved attempting Nm square moves chosen at random positions; this is two sweeps through the lattice of $Nm/2$ vertices. The temperature was then lowered to T, new vertex weights were calculated, and the Monte Carlo algorithm was begun using the final configuration of the stirring or heating interval as the initial configuration. The first 64 000 steps were discarded for equilibration, and then data for expectation values were saved for 192 000 steps. Expectation values of the internal energy, specific heat, in-plane magnetization, and susceptibility were computed in a manner identical to that for the spin-$\frac{1}{2}$ problem, as expectation values of appropriate derivatives of the vertex weights.[27] Six bins of 32 000 steps were used for estimation of errors. Finally, data from five such calculations were averaged to obtain the results presented here.

At zero applied field, it was found to be difficult to obtain zero average magnetization, due to the strong effective ferromagnetic exchange, especially at low T. This strong exchange, which is also seen in spin-$\frac{1}{2}$ QMC, causes an effective freezing of the QMC algorithm. This was somewhat alleviated by adding a global spin move (where all spins of the lattice are reversed in sign) attempted once every step. At zero field this move is always accepted and the average magnetization must come out as zero. At nonzero fields the move is accepted with probability 1 if $\Delta E \leq 0$ and probability $e^{-\beta \Delta E}$ if $\Delta E > 0$, as for any other elementary move.

V. QMC RESULTS FOR $CsNiF_3$

Calculations using $CsNiF_3$ parameters were made in the temperature range 5 K $\leq T \leq$ 15 K for fields up to 10 kG. The internal energy, specific heat and in-plane magnetization and susceptibility versus T are shown in Fig. 1, for fields 0, 5, and 10 kG. In Fig. 2 we present the *changes* in energy and specific heat, $\Delta U = U(B) - U(0)$ and $\Delta C = C(B) - C(0)$, to isolate contributions present only under application of the field (including, but not exclusively, "soliton" contributions). These data have vague peaks and the scatter is considerable. In Fig. 3 the specific heat ΔC versus field B is shown, at temperatures 5, 6, 7, 8, and 9 K. Classical sine-Gordon soliton theory predicts a peak in ΔC versus B, whose position (at $B = B_{\text{peak}}$) and height ($\Delta C_{\max}$) are proportional to T^2 and T, respectively. For these results, it is estimated, very roughly, that the peaks are at fields 2, 4, 5, 7, and 10 kG. We plot B_{peak} versus T^2 in Fig. 4, and compare with the SG theory prediction[24] and with the linear fit to the Ramirez and Wolf experimental data.[10] Linear fits to the three are

$$B_{\text{peak}} \approx 0.14T^2 - 1.4, \quad \text{QMC data},$$
$$B_{\text{peak}} \approx 0.131T^2, \quad \text{Ramirez and Wolf experiment}, \quad (13)$$
$$B_{\text{peak}} = 0.114T^2,$$

SG theory (Ref.24), no mass renormalization .

Generally the lack of agreement between experiment and SG theory has been attributed to a quantum reduction of the rest mass of the solitons. One can also compare the results of a Bethe ansatz solution of the quantized SG equation in Ref. 22. Surprisingly the slopes of the QMC and experimental data are in good agreement, although this may be coincidental, especially considering the quality of the QMC data. Also, there is considerable scatter in the QMC data for peak heights versus T; we can draw no firm conclusion from those data.

There are several deficiencies of this QMC calculation, the largest of which is its inefficiency. A better algorithm is needed with higher acceptance rates so that the statistical errors are reduced. There is also a strong tendency for the system to "freeze" at low T; possible solutions to this

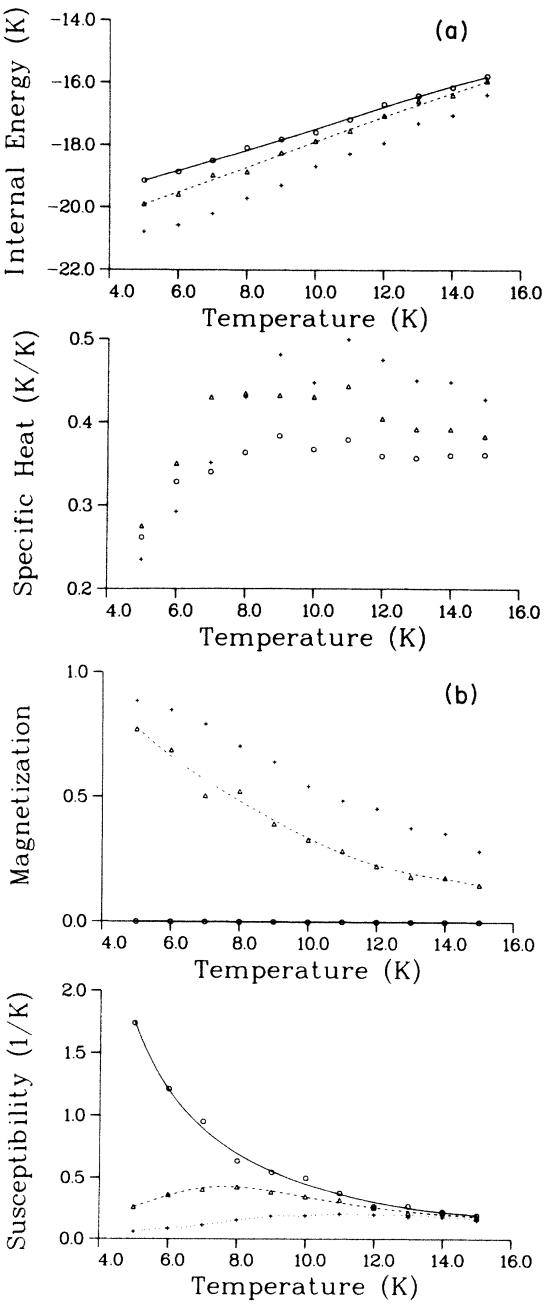

FIG. 1. Representative spin-1 QMC data for $CsNiF_3$ parameters, using 16 spins and $m \approx 60K/T$. The drawn curves are least-square polynomial fits to the QMC averages, for in-plane fields 0 kG (○, solid), 5 kG (Δ, dashed), and 10 kG (+ , dotted). All quantities are per spin.

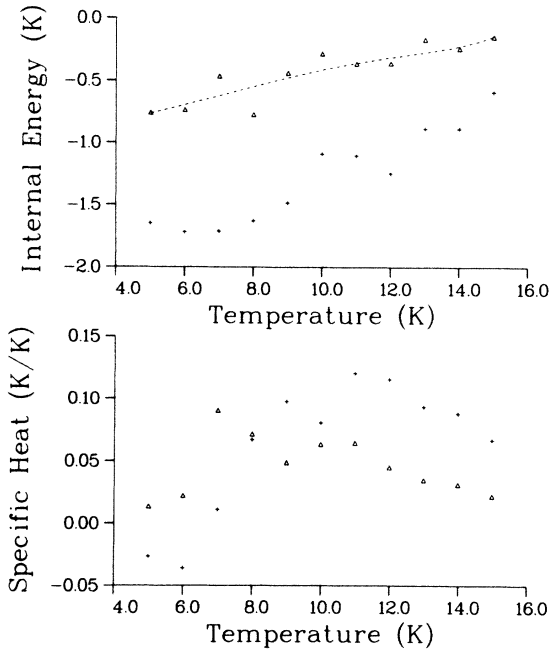

FIG. 2. The differences $\Delta U = U(B) - U(0)$ and $\Delta C = C(B) - C(0)$ versus temperature, derived from Figs. 1(a) and 1(b), for fields 5 kG (Δ, dashed) and 10 kG (+ , dotted). The drawn curves are the differences of the least-square polynomial fits in Fig. 1, while the data points are obtained by direct subtraction of the Fig. 1 data points.

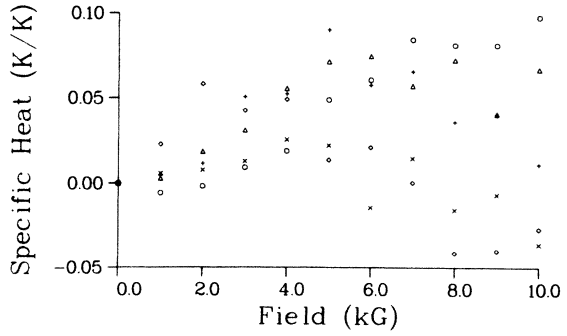

FIG. 3. The differences $\Delta C = C(B) - C(0)$ versus B, as obtained from the spin-1 QMC calculation for $CsNiF_3$ parameters (see text). The data points correspond to temperatures 5 K (◇), 6 K (×), 7 K (+), 8 K (Δ), and 9 K (○). There are only vague peaks in each set of data at fixed temperature, although these represent averages over $5 \times 192\,000$ states.

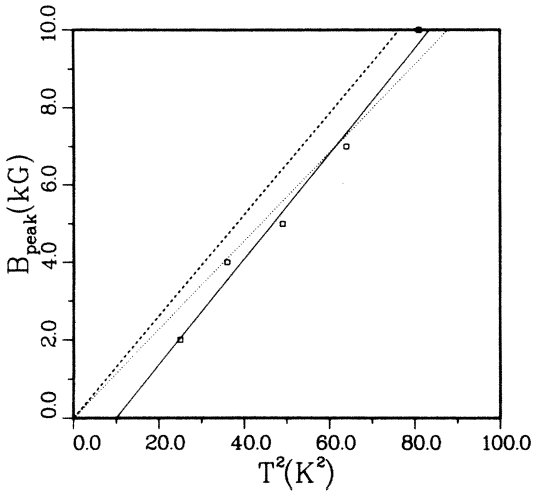

FIG. 4. Field B_{peaks}, at which ΔC vs B is maximum, versus T^2, from experimental and model calculations. The dashed line is a linear fit to the Ramirez and Wolf (Ref. 10) experimental data for $CsNiF_3$. The data points are rough estimates from the QMC calculation, and the solid line is a linear fit to those data. The dotted line is the prediction of classical SG theory of Sasaki and Tsuzuki (Ref. 24), which includes effects of soliton-soliton interactions, as well as multiple spin wave and spin wave-soliton processes.

problem might include some annealing procedure, or perhaps a different decomposition of $\hat{H}_{n,n+1}$ into $\hat{H}^0_{n,n+1}$ and $\hat{V}_{n,n+1}$. The other major problem is the finite lattice size, in *both* the spatial and Trotter directions. This latter problem is difficult to correct without first tending to the inefficiency problem.

Because of these difficulties, we cannot make any strong conclusions, especially concerning the question of a "quantum soliton" interpretation to the experimental data. Furthermore, it is not possible to verify the applicability and parameters of the presently accepted Hamiltonian for $CsNiF_3$ without a more accurate calculation. It is likely that quantum effects *are* important here, and further study, possibly by other methods, is needed. As a step in this direction, we present next a quantum transfer matrix calculation for the spin-$\frac{1}{2}$ model of CHAB, where the accuracy is much better and does show that there are some difficulties to be cleared up in the understanding of the Hamiltonian currently considered to be appropriate.

VI. QUANTUM TRANSFER MATRIX CALCULATION FOR $S=\frac{1}{2}$

The Hamiltonian in Eq. (2) is used here. The Trotter formula applies just as for $S=1$, but now the matrix elements are the spin-$\frac{1}{2}$ functions given previously.[27–30] The coordinate system is oriented so that it is an eight-vertex model, although this simplification is in no way necessary.

The computing method used was given by Betsuyaku,[29] who adapted that of Morgenstern and Binder[34] as originally applied to spin-glass models, by allowing for the four-spin interactions. It is necessary to choose free boundary conditions in the spatial direction, while periodic boundary conditions are imposed in the Trotter direction as a result of the trace.

Initially the 2^{2m} partial partition functions of the first column $(S_{1,r}, r=1,2,3,\ldots,2m)$ are stored in memory. Now consider adding the spins $S_{2,1}$ and $S_{2,2}$ to the lattice, in each of four possible states. This adds one four-spin block to the lattice. Since all the interactions involving $S_{1,1}$ and $S_{2,1}$ in the first column have been taken into account, we can perform the trace over them, and save in memory 2^{2m} partial partition functions labeled by the states of $S_{2,1},S_{2,2},S_{1,r}$, $r\geq 3$. Next, new spins are added at sites $S_{2,3},S_{2,4}$, and thus the trace over $S_{1,3}$ and $S_{1,4}$ can be performed, and the states are now labeled by $S_{2,1},S_{2,2},S_{2,3},S_{2,4},S_{1,r}$, $r\geq 5$. This procedure is continued until the whole second column of spins has been added to the lattice, at which point the partial partition functions saved will be labeled by states of the second column, $S_{2,r}$, $r=1,\ldots,2m$. This procedure is repeated, adding the spins of the third column by pairs, and tracing over the spins of the second column pair after pair. Columns are added iteratively, thereby transferring from the n-particle system to the $(n+1)$-particle system. The procedure is stopped after adding the Nth column and taking the trace over it, obtaining the total partition function $Z^{(m)}$ for any specified temperature. Internal energy, specific heat, magnetization, and susceptibility are then obtained by taking appropriate derivatives numerically.

The method requires storing the 2^{2m} Boltzmann factors—for this calculation we have used $1\leq m\leq 9$. Computing time rises exponentially with m and linearly with N. Presently the practical limit is $m=9$ for storage as well as CPU time using a CRAY-1 800K word machine, while $N\gtrsim 100$ is no practical problem.

For 1D magnets, it has been noted that for large N the total internal energy U scales like $U(N)/N=U_\infty+a/N$ (Ref. 35) where U_∞ and a are constants. Similarly, it has been shown that the leading errors in the Trotter approximation should be proportional to $1/m^2$ (Ref. 36), a result which has been demonstrated in the numerical calculations of Betsuyaku. We have made use of these two facts to extrapolate to the infinite N, infinite m limit. First, for a given value of m, data from $N=8$, 14, 20, 26, and 32 was extrapolated to obtain the $N\rightarrow\infty$, finite m limit. A weighted least-squares linear fit was used, where the weights were proportional to N. See Fig. 5 for an example. Then these data, for a series of m values satisfying $JS^2/mT<1$, were used to extrapolate to the $N\rightarrow\infty$, $m\rightarrow\infty$ limit (also in Fig. 5). Here a weighted least-squares linear fit was also made, where the weights were $1,2.5,2.5^2,\ldots,$ as m increases. The restriction $JS^2/mT<1$ is necessary since the Trotter errors actually scale with the square of this parameter. Curiously this parameter must be much smaller to obtain good results in an XY model than for a Heisenberg model.[27]

VII. QTM RESULTS FOR CHAB

First the method was tested for $m=8$, $N=32$, at 5% anisotropy $(J_y/J_x=0.95)$ to compare with previous spin-$\frac{1}{2}$ QMC data.[28,30] Results for internal energy, specific

heat, magnetization, and susceptibility all agreed to within about 5% over the temperature range 4 to 20 K. Then we applied this method to model (2) with anisotropy ranging from 4 to 10%, in order to compare with the experimental specific heat data of Kopinga *et al.*[11] As for $CsNiF_3$, ΔC is plotted versus field for a series of temperatures, and then the peak position and height are determined and plotted versus T^2 and T, respectively. Some representative ΔC versus B curves are shown in Fig. 6, for the case of 5% anisotropy. The data lie on smooth curves, making the determination of peak positions and heights possible. Interpolation, using a parabolic fit to the peaks, provided

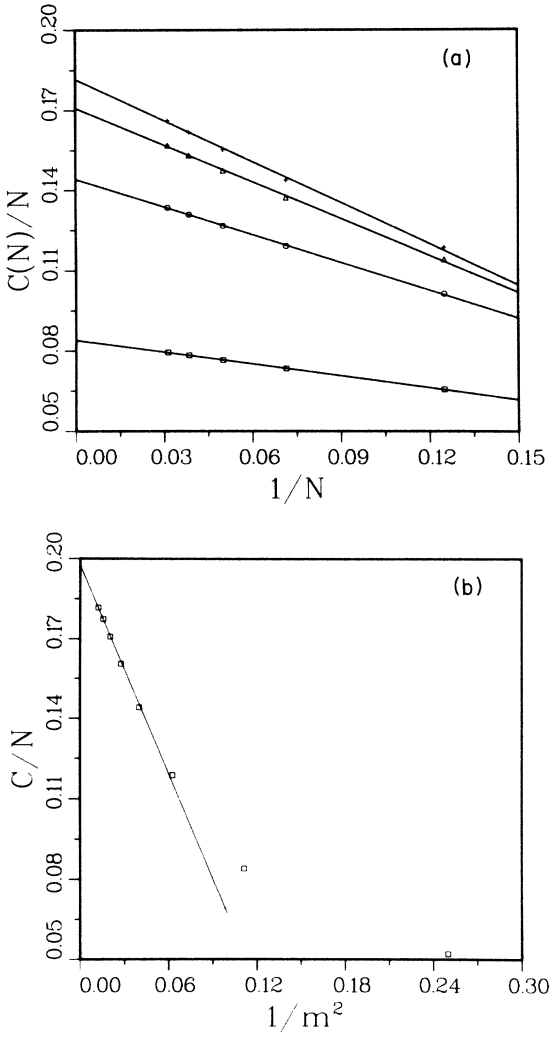

FIG. 5. An example of the extrapolation to $N\to\infty$, $m\to\infty$, for the specific heat per particle C/N, as used in the spin-$\frac{1}{2}$ QTM calculation. (Similar curves can be obtained for the internal energy, and in-plane magnetization and susceptibility.) CHAB parameters are used here; $J_x=J_z=110$ K, $J_y=104.5$ K. The in-plane field is $B_z=5.5$ kG, and the temperature is $T=7.20$ K. (a) Extrapolations to $N\to\infty$ at fixed values of $m=3$ (□), $m=5$ (○), $m=7$ (Δ), and $m=9$ (+). The straight lines are weighted least-square fits. (b) Extrapolation to $m\to\infty$, using $3\le m\le 9$. The data points are the values already extrapolated to $N\to\infty$, as found from the intercepts of curves as in (a). The straight line is a weighted least-squares fit.

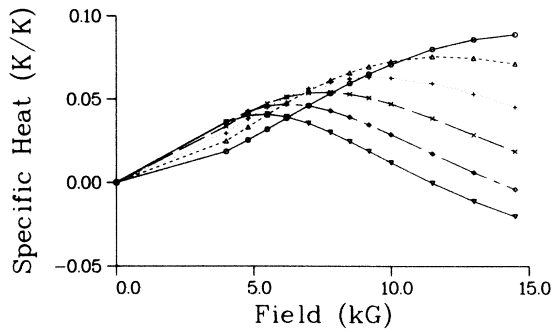

FIG. 6. Some typical results for ΔC vs B as obtained with the spin-$\frac{1}{2}$ QTM calculation using CHAB parameters (5% anisotropy). The data correspond to temperatures $T=4.0$ K (∇), 4.4 K ( ), 4.9 K (×), 5.5 K (+), 6.2 K (Δ), and 7.2 K (○), and have been extrapolated to $N\to\infty$, $m\to\infty$.

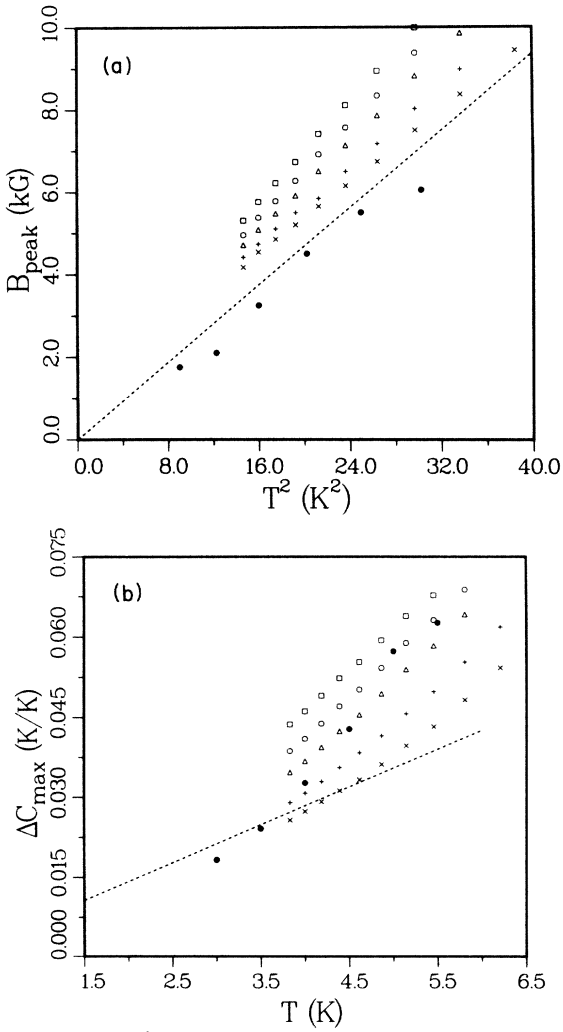

FIG. 7. Spin-$\frac{1}{2}$ QTM results for (a) B_{peak} and (b) ΔC_{max}, using model (2) with $J_x=J_z=110$ K, for a series of values of anisotropy $J_y/J_x=0.96$ (□), 0.95 (○), 0.94 (Δ), 0.92 (+), and 0.90 (×). These are all data from the extrapolation to $N\to\infty$, $m\to\infty$. The solid data points (●) are the experimental data on CHAB by Tinus *et al* (Ref. 20). The dashed lines are the classical SG theory of Sasaki and Tsuzuki (Ref. 24).

a simple accurate way to determine the heights and positions. In Fig. 7 the resulting $B_{\rm peak}$ and $\Delta C_{\rm max}$ are shown, for anisotropies 4, 5, 6, 8, and 10%, and compared with classical SG theory and experiment.

The drawn curves in Fig. 7 are classical SG theory results using a soliton rest mass $E_{SG}^0=8(JS^3g\mu_B B_z)^{1/2}$, that is, with no adjusted parameters, as in Ref. 24. The predictions of classical SG theory are independent of the anisotropy. Expressions given by Sasaki and Tsuzuki[24] include contributions from spin waves, solitons, and soliton-soliton interactions. Their calculations predict that the general result for a SG ferromagnet is

$$B_{\rm peak}=AT^2,\quad A=(64t_m^2 g\mu_B JS^3)^{-1}, \tag{14}$$

where $t_m=T/E_{\rm SG}^0=0.190$ has determined the peak position. The corresponding peak height is found to be given by

$$\Delta C_{\rm max}=A'T,\quad A'=0.196/JS^2 . \tag{15}$$

We see that agreement between this classical theory and experiment is fair for $B_{\rm peak}$ but not as good for $\Delta C_{\rm max}$. None of the chosen values of anisotropy for the QTM fit well to the experimental CHAB data over this temperature range. If the SG soliton rest mass is *ad hoc* renormalized such that the slopes of the SG theory $B_{\rm peak}$ curves agree with the experimental slope, then the implied changes in the SG theory $\Delta C_{\rm max}$ are not adequate to cause them to simultaneously fit the experimental data. It has not been apparent how to resolve this problem with classical SG theory. The QTM data presented here obviously should require no such quantum renormalization, but nevertheless systematically disagree with experiment.

VIII. DISCUSSION OF RESULTS

The lack of efficiency and resulting lack of accuracy of the $S=1$ QMC algorithm presented here led us to consider other methods of obtaining the low-temperature thermodynamics, the first of which is a quantum transfer matrix method. This transfer matrix method as introduced by Betsuyaku was applied here to the $S=\frac{1}{2}$ CHAB thermodynamics, thereby making earlier $S=\frac{1}{2}$ QMC data obsolete, due to the much greater precision of the newer method (i.e., no statistical errors). The results obtained for specific heat with an in-plane field are in disagreement with presently available experimental data. Other values of easy-plane anisotropy, from 4 to 10%, different from the accepted value of 5%, produced no good fit to the experimental data. It continues to be somewhat surprising how well the classical SG theory is capable of explaining the CHAB specific heat data. We have shown that the quantized version of the ferromagnet Hamiltonian gives approximately the same low-T thermodynamics as the classical SG Hamiltonian, for the case of $S=\frac{1}{2}$ CHAB. This can be compared with the classical transfer matrix calculations[20] for the ferromagnet Hamiltonians, which give *much* larger low-temperature specific-heat peaks. Apparently the quantum mechanics plays a strong role in restricting the spins to the easy plane (perhaps including a zero-point out-of-plane component) thereby making the classical theory more appropriate than might at first be expected.

Unfortunately, this QTM method cannot be applied in its present form to the $S=1$ $CsNiF_3$ problem, essentially because of computer memory limitations. Generally one expects that the errors due to a finite value of m in the Trotter approximation should scale with the parameter JS^2/mT. For the m-extrapolation method to work well, at a given temperature one needs data at several values of m satisfying $JS^2/mT\ll 1$. Coincidentally, $JS^2\approx 25$ K for $CsNiF_3$ as well as for CHAB, and since the interesting soliton regimes are both near $T=5$ K, one necessarily must use several points for which $m>5$. For $S=\frac{1}{2}$, the computer memory needed is of the order of 2×2^{2m} words, which is 520 000 words for $m=9$. This size was the practical limit (also in terms of CPU time) of available CRAY-1 machines (800K words) at Los Alamos. For $S=1$, the computer memory needed will be of the order of 2×3^{2m} words, which is about 1×10^6 words for $m=6$. This memory requirement is excessive and yet $m=6$ is too small for the extrapolation. The present QTM method cannot be used for this $S=1$ problem without taking into account the intrinsic symmetry of the Trotter subsystems of the 2D lattice (the columns, along the Trotter direction). (See Ref. 36 and 37.) Including these symmetries, possibly by using coherent spin states as in Ref. 37, will reduce the number of states being stored, thereby easing the computer memory limitation problem.

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APPENDIX I: CALCULATION OF SPIN-1 MATRIX ELEMENTS

The two-site Hamiltonian is broken up into $\hat{H}^0_{n,n+1}$ and $\hat{V}_{n,n+1}$ as

$$-\beta\hat{H}^0_{n,n+1}/m=K(x_nx_{n+1}+y_ny_{n+1}+z_nz_{n+1}) , \tag{A1}$$

$$\begin{aligned}-\beta\hat{V}_{n,n+1}/m&=-\beta\hat{V}_n/m-\beta\hat{V}_{n+1}/m\\&=-\alpha(y_n^2+y_{n+1}^2)+b(z_n+z_{n+1}) ,\end{aligned} \tag{A2}$$

where

$$K\equiv\beta J/m,\quad \alpha\equiv\beta A/2m,\quad b=\beta B/2m , \tag{A3}$$

and x_n, y_n, and z_n represent spin-1 operators $\hat{S}^x_n$, $\hat{S}^y_n$, and $\hat{S}^z_n$. The motivation here is that $\hat{V}_{n,n+1}$ is a moderate perturbation on $\hat{H}^0_{n,n+1}$ ($2A/J \approx 0.38$ for $CsNiF_3$), with very weak dependence on the field strength. Matrix elements of the two parts will be found separately; we sketch the method for $\hat{H}^0_{n,n+1}$; the same method can be applied even more easily to $\hat{V}_{n,n+1}$.

The exponential of (A1) is needed, i.e.,

$$\hat{\mathcal{M}} = \exp K\hat{M} = \sum_{p=0}^{\infty} \frac{1}{p!} K^p \hat{M}^p , \tag{A4}$$

where

$$\hat{M} = x_n x_{n+1} + y_n y_{n+1} + z_n z_{n+1} . \tag{A5}$$

A method is given here which produces $\hat{M}^{p+1}$ from $\hat{M}^p$, using the anticommutator for symmetrizing the algebra;

$$\hat{M}^{p+1} = \tfrac{1}{2}\{\hat{M}, \hat{M}^p\} . \tag{A6}$$

By iteratively taking higher powers of $\hat{M}$, it is found that $\hat{M}^p$ will generally involve eight different simple spin-1 operators, with coefficients of these operators depending on p, so we can write

$$\hat{M}^p = \sum_{k=1}^{8} C_k(p) \hat{O}_k . \tag{A7}$$

The sum is over the eight operators $\hat{O}_k$. One can use the fundamental commutator

$$z_n = i[x_n, y_n] \tag{A8}$$

to write all operators in terms of x and y operators. Then the symmetry of $\hat{M}$ with respect to interchange of x and y operators simplifies the calculation. The eight operators are

$$\begin{aligned}
\hat{O}_1 &= x_n x_{n+1} + \{x \leftrightarrow y\} , \\
\hat{O}_2 &= x_n^2 x_{n+1}^2 + \{x \leftrightarrow y\} , \\
\hat{O}_3 &= (xy)_n (xy)_{n+1} + \{x \leftrightarrow y\} , \\
\hat{O}_4 &= (xxy)_n (xxy)_{n+1} + \{x \leftrightarrow y\} , \\
\hat{O}_5 &= (xxyy)_n (xxyy)_{n+1} + \{x \leftrightarrow y\} , \\
\hat{O}_6 &= (xyyx)_n (xyyx)_{n+1} + \{x \leftrightarrow y\} , \\
\hat{O}_7 &= (xy)_n (yx)_{n+1} + \{x \leftrightarrow y\} , \\
\hat{O}_8 &= (xyyx)_n (yxxy)_{n+1} + \{x \leftrightarrow y\} ,
\end{aligned} \tag{A9}$$

where $\{x \leftrightarrow y\}$ indicates another term with the x and y operators interchanged. The anticommutators of these, divided by two as in (A6), are

$$\begin{aligned}
\tfrac{1}{2}\{\hat{O}_1, \hat{M}\} &= \hat{O}_2 + \hat{O}_3 - \tfrac{1}{2}\hat{O}_4 , \\
\tfrac{1}{2}\{\hat{O}_2, \hat{M}\} &= \hat{O}_1 - \hat{O}_3 + \tfrac{1}{2}\hat{O}_4 , \\
\tfrac{1}{2}\{\hat{O}_3, \hat{M}\} &= \tfrac{1}{2}\hat{O}_4 - \hat{O}_6 , \\
\tfrac{1}{2}\{\hat{O}_4, \hat{M}\} &= \hat{O}_3 - \tfrac{1}{2}\hat{O}_4 + \hat{O}_5 + \hat{O}_6 , \\
\tfrac{1}{2}\{\hat{O}_5, \hat{M}\} &= \hat{O}_4 , \\
\tfrac{1}{2}\{\hat{O}_6, \hat{M}\} &= -\hat{O}_3 + \tfrac{1}{2}\hat{O}_4 , \\
\tfrac{1}{2}\{\hat{O}_7, \hat{M}\} &= \hat{O}_8 , \\
\tfrac{1}{2}\{\hat{O}_8, \hat{M}\} &= \hat{O}_7 .
\end{aligned} \tag{A10}$$

Inserting (A7) into (A6), we see that the $C_k(p+1)$ coefficients are found from the $C_k(p)$ coefficients by two matrix relationships:

$$\begin{bmatrix} C_1 \\ C_2 \\ C_3 \\ C_4 \\ C_5 \\ C_6 \end{bmatrix}_{p+1} = \begin{bmatrix} 0 & 1 & 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 & 0 \\ 1 & -1 & 0 & 1 & 0 & -1 \\ -\frac{1}{2} & \frac{1}{2} & \frac{1}{2} & -\frac{1}{2} & 1 & \frac{1}{2} \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 1 & 0 & 0 \end{bmatrix} \begin{bmatrix} C_1 \\ C_2 \\ C_3 \\ C_4 \\ C_5 \\ C_6 \end{bmatrix}_p , \tag{A11}$$

$$\begin{bmatrix} C_7 \\ C_8 \end{bmatrix}_{p+1} = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \begin{bmatrix} C_7 \\ C_8 \end{bmatrix}_p . \tag{A12}$$

Now since

$$\hat{M} = \hat{O}_1 - \hat{O}_3 + \hat{O}_7 , \tag{A13}$$

the system is solved subject to the initial condition

$$\begin{aligned}
&C_1(1) = C_7(1) = 1 , \\
&C_3(1) = -1 , \\
&\text{all other } C_k(1) = 0 .
\end{aligned} \tag{A14}$$

The simplest way to solve the six-variable system is to expand the initial condition in terms of the eigenspectrum of the matrix transformation. One finds the eigenvalues $\lambda = -1$, $+1$, -2, and $-\frac{1}{2}$ with -1 triply degenerate. Then applying the transformation $(p-1)$ times to the initial condition easily gives the following solutions for the operator coefficients:

$$\begin{aligned}
C_1(p) &= C_7(p) = \tfrac{1}{2}[1 - (-1)^p] , \\
C_2(p) &= C_8(p) = \tfrac{1}{2}[1 + (-1)^p] , \\
C_3(p) &= \tfrac{1}{3}[(-2)^p - 1] , \\
C_4(p) &= -C_6(p) = -\tfrac{1}{6} + \tfrac{1}{2}(-1)^p - \tfrac{1}{3}(-2)^p , \\
C_5(p) &= -\tfrac{1}{6} - \tfrac{1}{2}(-1)^p + \tfrac{1}{6}(-2)^p .
\end{aligned} \tag{A15}$$

Multiplying these by $K^p/p!$ and summing on p, the following operator result is found for $\hat{\mathcal{M}} = \exp K\hat{M}$:

$$\hat{\mathcal{M}}=1+(\sinh K)(\hat{O}_1+\hat{O}_7)+(\cosh K-1)(\hat{O}_2+\hat{O}_8)-\tfrac{1}{3}(e^{K}-e^{-2K})\hat{O}_3+(\tfrac{1}{6}e^{K}-\tfrac{1}{2}e^{-K}+\tfrac{1}{3}e^{-2K})(\hat{O}_6-\hat{O}_4)$$
$$+(\tfrac{1}{2}-\tfrac{1}{6}e^{K}-\tfrac{1}{2}e^{-K}+\tfrac{1}{6}e^{-2K})\hat{O}_5 . \tag{A16}$$

A calculation similar to the above gives the following expression for the other part of the Hamiltonian:

$$\exp(-\beta\hat{V}_n/2m)=\exp(-\tfrac{1}{2}\alpha y_n^2+\tfrac{1}{2}bz_n)=1+(e^{-\alpha/2}-1)y_n^2+b\gamma^{-1}e^{-\alpha/4}(\sinh\gamma)z_n$$
$$+[e^{-\alpha/4}(\cosh\gamma+\tfrac{1}{4}\alpha\gamma^{-1}\sinh\gamma)-1](xyyx)_n$$
$$+[e^{-\alpha/4}(\cosh\gamma-\tfrac{1}{4}\alpha\gamma^{-1}\sinh\gamma)-e^{-\alpha/2}](yxxy)_n ,$$
$$\gamma\equiv\tfrac{1}{2}(b^2+\tfrac{1}{4}\alpha^2)^{1/2} . \tag{A17}$$

The nonzero matrix elements of $\hat{\mathcal{M}}$, the isotropic exchange operator, are given here, where the notation is

$$\langle S_n^z S_{n+1}^z\,|\,\hat{\mathcal{M}}\,|\,S_n^{z'}S_{n+1}^{z'}\rangle,\quad \langle 00\,|\,\mathcal{M}\,|\,00\rangle=\tfrac{1}{3}(2e^{K}+e^{-2K}),\quad \langle 11\,|\,\mathcal{M}\,|\,11\rangle=\langle -1-1\,|\,\mathcal{M}\,|\,-1-1\rangle=e^{K} ,$$
$$\langle 01\,|\,\mathcal{M}\,|\,01\rangle=\langle 10\,|\,\mathcal{M}\,|\,10\rangle=\langle 0-1\,|\,\mathcal{M}\,|\,0-1\rangle=\langle -10\,|\,\mathcal{M}\,|\,-10\rangle=\cosh K ,$$
$$\langle 01\,|\,\mathcal{M}\,|\,10\rangle=\langle 10\,|\,\mathcal{M}\,|\,01\rangle=\langle 0-1\,|\,\mathcal{M}\,|\,-10\rangle=\langle -10\,|\,\mathcal{M}\,|\,0-1\rangle=\sinh K ,$$
$$\langle 00\,|\,\mathcal{M}\,|\,1-1\rangle=\langle 1-1\,|\,\mathcal{M}\,|\,00\rangle=\langle 00\,|\,\mathcal{M}\,|\,-11\rangle=\langle -11\,|\,\mathcal{M}\,|\,00\rangle=\tfrac{1}{3}(e^{K}-e^{-2K}) ,$$
$$\langle 1-1\,|\,\mathcal{M}\,|\,1-1\rangle=\langle -11\,|\,\mathcal{M}\,|\,-11\rangle=\tfrac{1}{6}e^{K}+\tfrac{1}{2}e^{-K}+\tfrac{1}{3}e^{-2K} ,$$
$$\langle 1-1\,|\,\mathcal{M}\,|\,-11\rangle=\langle -11\,|\,\mathcal{M}\,|\,1-1\rangle=\tfrac{1}{6}e^{K}-\tfrac{1}{2}e^{-K}+\tfrac{1}{3}e^{-2K} . \tag{A18}$$

There are a total of 19 different nonzero vertex weights; all others are zero. When this interaction is combined with that due to the $\hat{V}_{n,n+1}$ term [symmetrized as in (12)], one finds that 41 of the possible 81 vertices are allowed. These allowed vertices are the only ones with an even number of 0's.

We also note that for arbitrary S, the isotropic exchange Hamiltonian will relate to a N_v-vertex model, where

$$N_v=2(1+2^2+3^2+\cdots+(2S)^2+(2S+1)^2 , \tag{A19}$$

obtained by counting those vertices which conserve total magnetization between states. For example, $S=\frac{3}{2}$ would be described by a 44-vertex model in the isotropic limit.

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