LA-UR -86-843

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36.

TITLE: QUANTUM MONTE CARLO AND TRANSFER MATRIX CALCULATIONS FOR 1-D EASY-PLANE FERROMAGNETS

AUTHOR(S): G. M. Wysin
A. R. Bishop

SUBMITTED TO: Physical Review B

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

FORM NO 836 R4
ST NO 2629 5/81

Phys. Rev. B 34, 3377 (1986).

Quantum Monte Carlo and Transfer Matrix Calculations for 1-D Easy-Plane Ferromagnets

G. M. Wysin* and A. R. Bishop
Theoretical Division
Los Alamos National Laboratory
Los Alamos, NM 87545

Abstract

We have applied previously used quantum Monte Carlo (QMC) techniques to obtain numerically the thermodynamics of two well-studied quasi-one-dimensional (1-D) 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 soliton-like kink excitations. The $S = \frac{1}{2}$ QMC calculations are verified and superceded by a numerically exact quantum transfer matrix (QTM) technique.

Results for the temperature dependence of the peak in the specific heat verses 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.

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, $(C_6,H_{11}NH_3)CuBr_3$, or "CHAB", has inspired even further interest in easy-plane ferromagnets (EPFs), 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]

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

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

$$\hat{H}_{S=1/2} = \sum_{n=1}^{N} [(-J_x\hat{S}_n^x\hat{S}_{n+1}^x + J_y\hat{S}_n^y\hat{S}_{n+1}^y + J_z\hat{S}_n^z\hat{S}_{n+1}^z) - g\mu_B\vec{B}\cdot\vec{S}_n] \quad . \tag{2}$$

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 ($g\mu_B B \ll 2AS$), 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 $CsNiF_3$ (Ref. 7,8) and TMMC (a similar antiferromagnet with S = 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 verses 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 $g\mu_B B_c = 2AS/3$. 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 (with 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 = 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 re-insert 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 antiferro-magnets -- 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] verses 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 1-D quantum system is mapped onto the thermodynamics of a 2-D 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 QMC may prove to be more precise. One possibility is the "numerically exact" 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 2-D 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 2-D lattice, and therefore control the Monte Carlo updating.

II. Trotter-Suzuki Formalism: Checkerboard Decomposition

The original 1-D quantum thermodyanmics spin problem is mapped onto an approximately equivalent 2-D 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

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

where β is the reciprocal of the temperature T (we use Boltzmann's constant $k_B = 1$) and $\sigma = \{S_n, n = 1,2,3...N\}$, 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

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

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

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

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

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

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

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

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

$$Z^{(m)} = \mathrm{tr}\{[(e^{-\frac{\beta\hat{H}^{o}_{odd}}{m}} e^{-\frac{\beta\hat{V}_{odd}}{m}})(e^{-\frac{\beta\hat{H}^{o}_{even}}{m}} e^{-\frac{\beta\hat{V}_{even}}{m}})]^{m}\} \quad . \tag{7}$$

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

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

where

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

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

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

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

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

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

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

The r variable is the new added dimension. Since the only terms which contribute to the energy of the 2-D 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 2-D lattice consists of $\frac{1}{2}Nm$ blocks of 4 spins, or "vertices". In the limit $m \to \infty$, $Z^{(m)}$ approaches the exact partition function of the original 1-D quantum system. The 2-D 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:

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

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 4-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, 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 zeroes (or ±1's), and this influences the choice of an appropriate Monte Carlo updating algorithm.

The decision whether to attempt the flipped or unflipped move is based on the number of zeroes, N_o, along the path before the move. For a given initial path, there are 2^{N_o} flipped moves possible, and 2^{P-N_o} unflipped ones, for a total of $2^{N_o} + 2^{P-N_o}$ possible output states. Therefore the flipped moves should be attempted with probability $2^{N_o}/(2^{N_o} + 2^{P-N_o})$, and the unflipped moves should be attempted with probability $2^{P-N_o}/(2^{N_o} + 2^{P-N_o})$. 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 $mT \simeq 60$ 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 « 1%, while square moves have larger acceptance rates $\simeq$ 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 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 alieviated 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 $5K \leq T \leq 15$ K for fields up to 10 kG. The internal energy, specific heat and in-plane magnetization and susceptibility verses T are shown in Fig. 1, for fields 0, 5 kG 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 verses field B is shown, at temperatures 5, 6, 7, 8 and 9K. Classical sine-Gordon soliton theory predicts a peak in ΔC verses B, whose position (at $B = B_{peak}$) and height (ΔC_{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} verses T^2 in Figure 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

$$\begin{aligned} B_{peak} &\simeq 0.14\ T^2 - 1.4 \quad , \quad \text{QMC data,} \\ B_{peak} &\simeq 0.131\ T^2 \quad , \quad \text{Ramirez and Wolf experiment,} \\ B_{peak} &= 0.114\ T^2 \quad , \quad \text{sG theory,}^{24} \text{ no mass renormalization.} \end{aligned} \tag{13}$$

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 verses 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