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Monte Carlo computation of the free energy in quantum two-dimensional Heisenberg ferromagnets using the expanded-ensemble method

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Abstract. The expanded-ensemble method, initially formulated for classical systems, is combined with the Handscomb quantum Monte Carlo approach to develop a new method for the free-energy MC calculation of quantum systems. It was applied to a quantum 2D Heisenberg ferromagnetic system and proved effective. Comparisons were performed between the Takahashi spin-wave approximation for free energy and our numerical results.

1. Introduction

In our previous paper (Favorsky *et al* 1992) we reported Monte Carlo (MC) simulations of a quantum two-dimensional (2D) Heisenberg system with the ferromagnetic interaction of nearest neighbours with the Hamiltonian

$$\mathcal{H} = -2J \sum_{(ij)} \boldsymbol{S}_i \cdot \boldsymbol{S}_j . \tag{1}$$

Such thermodynamic characteristics of the canonical ensemble as the internal energy, the zero-field susceptibility and the correlation functions were calculated using the Handscomb (1962, 1964) quantum MC (HQMC) method. The latter proved to be an effective tool for MC simulations of quantum systems. Still, both the Handscomb method in its conventional form and the other available quantum MC method (Suzuki 1976) fail to deal with the simulation of such an important characteristic as the canonical ensemble free energy.

We demonstrate in this paper the way that we have combined the HQMC method with an expanded-ensemble approach (Lubartsev *et al* 1992) to produce a method for the numerical calculation of the free energy for a class of quantum systems previously open only to analytical approximations (Takahashi 1986) or finite-system Hamiltonian diagonalizations.

1.1. Existing approaches to free-energy numerical simulation

Over the last 20 years a number of methods have been developed aimed at MC calculation of the free energy for classical systems. The problem of such an estimation lies in the fact that the Gibbs factors (statistical weights) $\exp[-\beta\mathcal{H}(q)]$ are normalized by the partition function Z itself. As a result, it is impossible to calculate Z within the

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framework of any single canonical ensemble; there is no corresponding 'microscopic' MC variable to average.

Some of the approaches taken to overcome this difficulty, such as the particle insertion method (Widom 1963), are unsuitable for a lattice system, classical or quantum. Some of the others, although formulated for classical systems, can be quite readily applied to Handscomb-inspired MC treatment of quantum Heisenberg systems.

The multistage sampling method proposed by Valleau and Card (1972) calculates the free-energy differences (FEDs) between two canonical ensembles, provided that we know their energy distributions. In fact, we can obtain such distributions naturally in the course of the Handscomb quantum MC simulation of energy and susceptibility. Still, the weakest point of the approach remains the same as with the classical case, i.e. the distributions in question must overlap at least partially.

The approach closest to that used in our simulations is the acceptance ratio method of Bennett (1976). However, it involves, for a pair of ensembles, optimization over two parameters, while the expanded-ensemble method has only one parameter to optimize.

In the expanded-ensemble concept proposed by Lubartsev *et al* (1992) a set of inverse temperatures $\beta_0, \beta_1, \ldots, \beta_M$ ($\beta = J/kT$) is introduced. β_0 is zero, and the high-temperature limit is the usual reference point for the free-energy calculation. Then an expanded ensemble is composed of individual canonical ensembles at the inverse temperatures β_i. The partition function of the composite ensemble is a sum over all the subensembles:

$$Z = \sum_{i=0}^{M} Z(\beta_i). \tag{2}$$

The MC procedure is then organized with two kinds of step involved: conventional steps over configurational space of a canonical ensemble at the inverse temperature β_i ($0 < i < M$) and transitions between temperature points.

Theoretically, this approach should have provided us with an opportunity to estimate ensemble weight ratios p_i/p_k by means of calculating the probability for a subensemble to occur during an MC run:

$$p_i/p_k = N_i/N_k \tag{3}$$

where N_i and N_k are the numbers of occurrences for the ith and kth subensembles, respectively.

On the other hand,

$$p_i/p_k = [Z(\beta_i)/Z]/[Z(\beta_k)/Z]. \tag{4}$$

The partition function of the whole ensemble cancels out.

Since

$$Z(\beta_i)/Z(\beta_k) = \exp[\beta_k F(\beta_k) - \beta_i F(\beta_i)] \tag{5}$$

knowledge of the occurrence ratios yields the FEDs between the subensembles at β_i and β_k.

Note that from now on we shall call the free energy divided by the temperature, βF, and the energy divided by the temperature, βE, the free energy and the energy, respectively. Unless it is specified otherwise, all values describe the whole system.

However attractive, a straightforward application of the outlined scheme is unfeasible; the system will invariably rapidly collapse into the lowest-temperature (highest-weight) subensemble and will leave it extremely rarely.

In order to equalize the weights of the subensembles, the balancing factors $\exp(\eta_i)$ were introduced by Lubartsev *et al* (1992). The partition function Z of the thus modified expanded ensemble takes the form

$$Z = \sum_{i=0}^{M} Z(\beta_i)\exp(\eta_i) \tag{6}$$

and the FEDs can be easily calculated:

$$\beta_k F_k - \beta_i F_i = \eta_k - \eta_i + \log(N_i/N_k). \tag{7}$$

2. Expanded-ensemble method applied to quantum Heisenberg ferromagnets: Handscomb quantum Monte Carlo approach

The Handscomb quantum MC method developed by Handscomb (1962, 1964), Lyklema (1982) and Favorsky *et al* (1992) made MC simulations of various quantum spin-$\frac{1}{2}$ Heisenberg and Ising systems possible. According to the Handscomb approach, sampling is performed in the space of permutation operator sequences C_r, $r = 0, 1, 2, \ldots$:

$$C_r = P_{i_1} \ldots P_{i_k} \ldots P_{i_r} \tag{8}$$

where a permutation operator P_{i_k} switches the spins occupying currently the sites connected by the i_k bond.

The expanded-ensemble approach combined with the Handscomb approach can be described best by writing a sum for the partition function of the canonical ensemble using MC variables:

$$Z(\beta) = \sum_{r=0}^{\infty}\sum_{C_r} p(C_r, \beta) \tag{9}$$

where $p(C_r, \beta)$ is the non-normalized weight of the C_r sequence:

$$p(C_r, \beta) = (\beta^r/r!)\,\mathrm{Tr}(C_r) = (\beta^r/r!)2^{n(cl)}. \tag{10}$$

The permutation induced by the C_r-sequence can be reduced to cycles independent of each other (Lyklema 1982); $n(cl)$ is the current number of those cycles. The balanced weight p_{bal} of the C_r-sequence will depend on which subensemble it belongs to:

$$p_{\mathrm{bal}} = p(C_r, \beta_i)\exp(\eta_i). \tag{11}$$

2.1. MC procedure realization

Mostly because of the computational resources available we have been using four-point sets $\{\beta_i\}$. They overlapped and covered the whole of our temperature range (from 40.0 to 0.5), although it is possible to treat all temperatures at once. Steps over 'configurational' space (r-steps) were identical with those in the original Handscomb approach and their acceptance probability can be found elsewhere (Handscomb 1962, 1964, Lyklema 1982, Favorsky *et al* 1992).

Except for the finite-size effect study, we have used in the course of the simulations 24×24 systems with periodic boundary conditions. The MC chain lengths ranged from 3000 to 10 000 MC steps per spin per temperature point. Error margins were estimated

in a conventional way: by dividing the whole run into ten or so bins and monitoring the difference between the overall and the bin averages. Actual error bars for the free energy and related variables will be presented in section 3, where the free-energy results are presented and discussed.

To achieve a sufficient amount of fluctuations in the r-space between the attempted β-steps, the probability of initiating an r-step was chosen to be nine times that of the β-step. For the β-step acceptance probability $T(\beta_1 \rightarrow \beta_2)$, using the Handscomb expansion for the partition function in equation (9) and detailed balance principle, one can write

$$T(\beta_1 \rightarrow \beta_2) = \min[1, p(\beta_2 \rightarrow \beta_1)p_{\mathrm{bal}}(C_r, \beta_2)/p(\beta_1 \rightarrow \beta_2)p_{\mathrm{bal}}(C_r, \beta_1)] \tag{12}$$

where $p(\beta_i \rightarrow \beta_k)$ is the probability, when at the β_i point, of initiating the $\beta_i \rightarrow \beta_k$ step.

Since in our scheme all probabilities $p(\beta_i \rightarrow \beta_k)$ are equal, the acceptance ratio took the form of the weights ratio

$$p_{\mathrm{bal}}(C_r, \beta_2)/p_{\mathrm{bal}}(C_r, \beta_1) = (\beta_2/\beta_1)^r \exp(\eta_2 - \eta_1) = (T_2/T_1)^{-r} \exp(\eta_2 - \eta_1). \tag{13}$$

It is instructive to note here that $-r$ in equation (13) is the 'microscopic' equivalent of energy in our Handscomb process:

$$\beta\langle E\rangle = -\langle r\rangle + \beta N. \tag{14}$$

We could have incorporated the βN-term into the acceptance ratio equation (13) and it would have led to changes in η_i-values for the balanced ensemble but we chose to keep the formulae in the program simple and to 'rescale' the free energy to its conventional values later (see section 3.1).

2.2. *Balancing the system*

We started with a more or less arbitrary set of $\{\eta_i\}$ for a given set of $\{\beta_i\}$. After a few trial runs we were usually able to adjust η_i sufficiently to allow the system to visit all the temperature points repeatedly. Although at this stage the frequencies of occurrence varied greatly (something like $N_i/N_k = 0.01$ for some subensembles), for the purposes of statistically representative free-energy estimations we fine-tuned the system to achieve at least $N_i/N_k > 0.1$ for all subensembles.

2.3. *Absolute free-energy estimation*

It is clear that, with a method capable of calculating only FEDs, it is necessary to have a reference point that can be reached in the course of simulations.

In the case of spin-$\frac{1}{2}$ systems we know the free-energy high-temperature limit:

$$\beta F(0) = -S(0) = -N \log 2 \tag{15}$$

where N is the number of spins in the system ($N = 576$). However, the power rather than the classical exponential dependence of the acceptance ratio (13) rules out the possibility of including the $\beta = 0$ point into our chain of overlapping $\{\beta_i\}$ sets.

It was our belief that the way to overcome the difficulty lay in extending the MC simulation into the high-temperature region and estimating the first few terms of the Taylor series for the entropy near the $\beta = 0$ point. We used the finite-element method to do this.

The 'microscopic' equivalent of entropy differences can be easily found from the corresponding equations for the FEDs and the energy:

$$S(\beta_2) - S(\beta_1) = \langle r_2 \rangle - \langle r_2 \rangle + \eta_1 - \eta_2 + \ln(N_2/N_1). \tag{16}$$

Thus the entropy differences were estimated in our MC scheme together with the FEDs and the energy.

Using $T/J = 40$, 35 and 25 points for estimation we found that the difference between the entropy $S(1/40)$ and its high-temperature limit $S(0)$ appeared not to exceed 0.2%. So, after performing first-order corrections for the entropy, we went on to calculate the absolute free energy with a fair degree of assurance.

Table 1. Absolute free-energy simulation results.

T/J	Absolute free energy $-\beta F$, MC simulations	T/J	Absolute free energy $-\beta F$, MC simulations
40.0	413	1.20	1090
35.0	415	1.15	1120
30.0	418	1.10	1160
25.0	422	1.05	1210
22.0	425	1.00	1260
19.0	430	0.95	1310
16.0	436	0.93	1330
13.0	445	0.915	1350
10.0	460	0.9	1370
7.0	489	0.866	1420
6.0	505	0.833	1470
5.0	530	0.8	1520
4.0	570	0.766	1580
3.5	600	0.733	1640
3.0	630	0.7	1710
2.5	690	0.68	1760
2.3	720	0.665	1790
2.15	740	0.65	1830
2.0	770	0.63	1890
1.9	800	0.615	1930
1.8	820	0.6	1970
1.7	850	0.58	2040
1.6	890	0.565	2090
1.4	970	0.55	2140
1.35	1000	0.53	2220
1.30	1030	0.515	2280
1.25	1060	0.5	2350

3. Free-energy simulation results and discussion

Absolute free-energy simulation results are presented in table 1. The error margin is reflected in the number of significant digits left. In order to compare the results with the analytical theory low-temperature predictions for the free energy made by Takahashi (1986), we must 'rescale' them to conform with Takahashi's choice of energy ground states value, the Hamiltonian (Takahashi 1986) being

$$\mathcal{H} = -J \sum_{\{ij\}} (\boldsymbol{S}_i \cdot \boldsymbol{S}_j - \tfrac{1}{4}). \tag{17}$$

In figure 1 we have plotted the simulated free energy βf per spin together with the theoretical curve

$$\beta f = -(T/4\pi J)[\zeta(2)/6 + T\zeta(3)/8]. \tag{18}$$

Both were calculated for Takahashi's choice of energy ground-state value and our coupling constant.

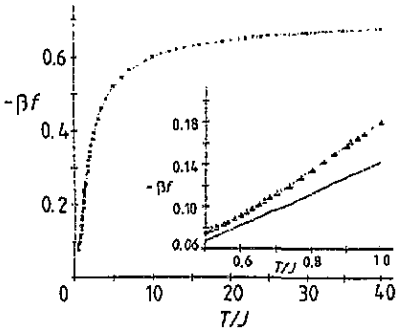

Figure 1. Absolute free energy per spin rescaled to the Takahashi choice of ground-state energy: ■, MC results. The inset shows a comparison of the MC results (Δ) with the TLTA (——) for the free energy.

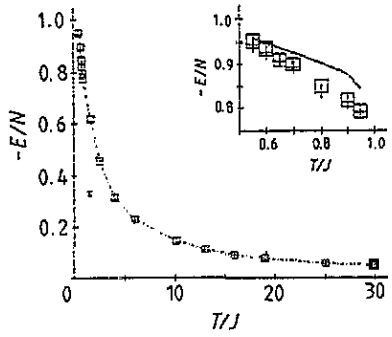

Figure 2. The internal energy of the system per spin: □, directly simulated in the course of an MC run; +, calculated from the FED using the thermodynamic identity. The inset shows a comparison of the MC results (□, +) with the TLTA (——) for the energy. Note that the size of the experimental points reflects the error bars only in the inset.

The discrepancies decrease from 20% for $T/J = 0.9$ to 13.5% for $T/J = 0.7$ and 9% for $T/J = 0.55$.

We had several possible ways to account for these large disagreements.

(1) There are possible faults and inaccuracies in our approach, such as size effects or internal discrepancies within our results.

(2) It is possible that the Takahashi low-temperature approximation (TLTA) was still invalid in the range of temperatures studied. This supposition is partly supported by earlier simulations of correlation lengths (Manousakis and Salvador 1989) and susceptibility (Favorsky *et al* 1992). The studies showed that, although the correlation lengths and susceptibility did exhibit for a certain range of temperatures the exponential dependence predicted by the TLTA ($\exp(b'/T)$ and $\exp(b/T)$, respectively), the b-coefficients differ considerably from the values obtained by Takahashi.

To study the possible size–temperature dependence of free energy, we performed simulations with lattices 40×40 and 50×50 for the lower temperatures where finite-size effects should have been at their most dramatic. The resulting FEDs are presented in table 2. The error margin of this particular MC run was about 1.5%; so the numbers are reliable only up to the third digit. The fourth digit was provided to show that

Table 2. Finite-size effects study. Note that in the last column we have listed the FEDs between $T/J = 0.63$ and $T/J = 0.6$. In the case of the 50×50 system we had to insert two additional temperature points between 0.63 and 0.6 to help with balancing.

	FED				
	$T/J = 0.665$	$T/J = 0.650$	$T/J = 0.630$	$T/J = 0.600$	Between $T/J = 0.63$ and $T/J = 0.6$
24×24 (βF)	0	38.2	92	180	
24×24 (βf)	0	0.006 62	0.1598	0.3126	0.1523
40×40 (βF)	0	106	255	500	
40×40 (βf)	0	0.006 61	0.1595	0.3123	0.1527
50×50 (βF)	—	—	0	380	
50×50 (βf)	—	—	0		0.1521

the $\Delta(\beta f)$ results agree exceptionally well and do not display any size dependence at all.

To prove the validity of our approach we have tested our results for self-consistency. Since the temperature points were situated close enough for us to calculate the inverse temperature derivatives $\mathrm{d}(\beta f)/\mathrm{d}\beta$, we used the well known thermodynamical equation

$$\mathrm{d}(\beta f)/\mathrm{d}\beta = E/N \tag{19}$$

to find the internal energy of the ensemble for the entire temperature range.

Although the free energy of an ensemble is obviously intrinsically related to the 'microscopic' distribution of the ensemble's energy (Valleau and Card 1972), the internal energy and FEDs have been estimated in the course of our MC sampling independently of each other. The results of the application of equation (19) are presented in figure 2. They agree with the directly simulated energy within the error bars. In the temperature range below $T/J = 5$ the error bars amount to 3% for both the calculated and the simulated energies. The increase in the relative error, which can be detected in the variance data in table 3, is explained by the fact that $\langle r \rangle / N\beta$ and $\mathrm{d}(\beta f)/\mathrm{d}\beta$ give us the energy $\langle E \rangle / N$ plus a constant (see equation (14) and the last paragraph of section 2.1). The errors in both $\langle r \rangle / \beta$ and βf decrease from 1.5% at $T/J = 0.5$ to 0.4% at $T/J = 40.0$, thus also reducing the absolute error in the energy (estimated before the constant is subtracted) but evidently not enough in our case to prevent the relative error in the energy from increasing to 5% for $T/J = 30.0$.

The Takahashi spin-wave low-temperature approximation for quantum 2D Heisenberg ferromagnets was one of the analytical theories that we compared our results with in our previous paper. Unfortunately, the low-temperature energy formula that we used for comparison there was based on that in the work of Okabe and Kikuchi (1988). It was only after the original Takahashi paper became available to us that we discovered an error which was obviously typographical in the Okabe–Kikuchi formula. The internal energy E/N per spin calculated from βf in equation (18) and presented in the inset of figure 2 shows good agreement with the simulated energy only for lower temperatures ($T < 0.7$). This agrees with the Okabe–Kikuchi MC simulation results for the same system: their energy results began to agree with those of Takahashi only from $T/J \simeq 0.5$.

After considering the observations mentioned above, together with the fact that discrepancies between our numerical simulations and the TLTA appear to decrease with increasing temperature, we came to the conclusion that the approximation, when

Table 3. Self-consistency testing: comparison with the TLTA.

	Energy calculated			
T/J	Using PED $E/N = \partial(\beta f)/\partial\beta$	Direct HQMC $\langle E\rangle/N$	TLTA	Statistical variance
30.0	0.052	0.049		0.003
25.0	0.060	0.060		0.003
19.0	0.080	0.078		0.003
16.0	0.094	0.093		0.004
13.0	0.115	0.113		0.005
10.0	0.149	0.143		0.006
6.0	0.231	0.228		0.007
4.0	0.317	0.319		0.009
2.5	0.456	0.458		0.005
1.7	0.62	0.61		0.01
1.0	0.77	0.77		0.02
0.95	0.79	0.79	0.8452	0.02
0.9	0.82	0.81	0.8765	0.02
0.8	0.85	0.84	0.9040	0.03
0.7	0.90	0.89	0.9276	0.03
0.65	0.91	0.91	0.9381	0.03
0.6	0.93	0.93	0.9477	0.03
0.55	0.95	0.94	0.9564	0.03

applied to Heisenberg 2D quantum ferromagnets, describes the internal energy of the system better than the free energy; while the energy approximation is valid up to the temperature $T/J = 0.7$, the accuracy of the free-energy approximation is still far from satisfactory, even for $T/J = 0.55$.

4. Conclusion

The results of numerical simulations presented in this paper prove the effectiveness of combining the β expanded-ensemble method with the Handscomb quantum MC procedure for the simulation of the free energy of a 2D quantum Heisenberg ferromagnet. Following the principles proposed in our scheme it should be easy to use the method outlined for other quantum Heisenberg-like systems.

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