A new approach to Monte Carlo simulations in statistical physics: Wang-Landau sampling

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We describe a Monte Carlo algorithm for doing simulations in classical statistical physics in a different way. Instead of sampling the probability distribution at a fixed temperature, a random walk is performed in energy space to extract an estimate for the density of states. The probability can be computed at any temperature by weighting the density of states by the appropriate Boltzmann factor. Thermodynamic properties can be determined from suitable derivatives of the partition function and, unlike ''standard'' methods, the free energy and entropy can also be computed directly. To demonstrate the simplicity and power of the algorithm, we apply it to models exhibiting first-order or second-order phase transitions. © *2004 American Association of Physics Teachers.*
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I. INTRODUCTION

Computer simulation now plays a major role in statistical physics,[1] particularly for the study of phase transitions and critical phenomena, and is an important tool for teaching and understanding thermodynamics and statistical mechanics.[2] The reason for its importance is that all but the simplest models are theoretically intractable, and only approximate methods can generally be used. In particular, stochastic techniques known as Monte Carlo (MC) simulations have proven to be very powerful. The standard MC method, developed a half-century ago, is the Metropolis importance sampling algorithm,[3] but more recently new, more efficient algorithms have begun to play a role in allowing simulations to achieve the resolution that is needed to accurately locate and characterize phase transitions.[1]

The motivation for these new developments is that traditional methods exhibit long time scales, thus requiring long simulations. At first-order phase transitions metastable states appear, and critical slowing down becomes a problem at continuous transitions. For spin systems, beginning with the seminal work of Swendsen and Wang,[4] and extended by Wolff,[5] cluster algorithms have been used to reduce critical slowing down. The multicanonical ensemble method[6–10] was introduced to overcome the tunneling barrier between coexisting phases at first-order transitions, and has general utility for systems with a rough energy landscape.[7,11,12] In both situations, histogram reweighting techniques[13] can be applied in the analysis to increase the amount of information that can be gleaned from simulational data, but the applicability of reweighting is severely limited in large systems by the statistical quality of the wings of the histogram. This limitation is important in systems with competing interactions for which short-range order effects might occur over very broad temperature ranges or give rise to frustration and produce a very complicated energy landscape, thus reducing the efficiency of standard methods.

The partition function can be expressed in terms of a density of states $g(E)$, the number of all possible states (or configurations) for an energy level E of the system, but direct estimation of $g(E)$ has not usually been the goal of simulations. Instead, most conventional MC algorithms,[1] such as Metropolis importance sampling and Swendsen-Wang cluster flipping, generate an unnormalized canonical distribution

$$P(E,T)=g(E)e^{-E/k_BT} \tag{1}$$

at a given temperature T (k_B denotes the Boltzmann constant). Such distributions are so narrow that multiple runs are usually needed to describe thermodynamic quantities over a significant range of temperatures. Because $g(E)$ does not depend on the temperature, we can construct canonical distributions at any temperature if we can estimate $g(E)$ with high accuracy for all energies. Once $g(E)$ is known, we can calculate the partition function as

$$Z=\sum_{\{\text{configurations}\}} e^{-E/k_BT}=\sum_E g(E)e^{-E/k_BT}, \tag{2}$$

and the model is essentially ''solved,'' because most thermodynamic quantities can be calculated from Z.

Although MC methods are already very powerful,[1] there has been no efficient algorithm to calculate $g(E)$ very accurately for large systems. Even for exactly solvable models, such as the two-dimensional (2D) Ising model, $g(E)$ cannot be calculated exactly for large systems.[14] All methods based on accumulation of histogram entries[13,15–18] have the problem of scalability for large systems.

In this paper we describe a new, general, and efficient MC algorithm (generally known as the ''Wang-Landau algorithm'') that offers substantial advantages over existing approaches.[19] We will explain the algorithm in detail and describe its application for first- and second-order phase transitions. Unlike conventional MC methods that directly generate a canonical distribution $g(E)e^{-E/k_BT}$ at a given temperature T, this approach estimates $g(E)$ directly and accurately via a random walk that produces a flat histogram in energy space. The estimate for $g(E)$ is improved at each step of the random walk, using a carefully controlled modification factor, to produce a result that converges to the real value quickly.

Wang-Landau sampling[19] has proven to be very useful and efficient in many different applications, including studies of complex systems with rough energy landscapes. For example, the method has been used in studies of a Potts antiferromagnet,[20] random spin systems,[21] quantum

systems,[22–24] fluids,[25,26] binary Lennard-Jones glass,[27] liquid crystals,[28] polymers,[25,29] proteins,[30,31] other molecular systems,[32,33] atomic clusters,[34] optimization problems,[35] and combinatorial number theory.[36] Generalizations and further studies of this sampling technique have been carried out by several authors.[37–41]

Although the Wang-Landau method can be applied to many different types of systems, we will describe it here only in the context of classical spin systems with discrete energy values. Therefore, when we refer to the density of states $g(E)$, we do not mean an actual *density*, but the number of states for a given energy E. The two simple models of interest are the Ising model,[42] which has a second-order phase transition, and the Q-state Potts model[43] with $Q=8$, which undergoes a first-order phase transition.

II. THE WANG-LANDAU ALGORITHM

If we perform an unbiased random walk in energy space by changing the states of the spins at random and accepting all energy values thus obtained, the histogram of the energy distribution should converge to the density of states $g(E)$ in the limit of a very long random walk that visits all possible spin configurations of the system. In practice it is forbiddingly difficult to realize such a long random walk with our current computer resources, given the extremely large number of spin configurations. For example, the Ising model on a 10×10 square lattice already has $2^{100}\approx 1.3\times 10^{30}$ spin configurations!

The Wang-Landau sampling method performs random walks in energy space by changing the states of spins randomly, but the energy E associated with each spin configuration is only accepted with a probability that is proportional to the reciprocal of the density of states. During the random walk, we also accumulate the histogram $H(E)$ in energy space, a quantity that keeps track of the number of visits at each energy level E [each time an energy E is visited, the corresponding entry in $H(E)$ is incremented by 1]. The algorithm modifies the estimate of the density of states by a multiplicative factor f, and uses the updated density of states to perform a further random walk in energy space. With this choice of acceptance probability, each random walk generates a flat histogram for the energy distribution. The modification factor f is carefully controlled, and at the end of the simulation, it should be very close to 1, which is the ideal case of the random walk with the true density of states.

At the beginning of the simulation, $g(E)$ is unknown, and we make an initial guess for it. The simplest approach is to set $g(E)=1$ for all possible energies E. The initial spin configuration for the entire lattice can be chosen arbitrarily. Then, a random walk in energy space is begun by forming trial states, each of which is produced by randomly picking a spin and randomly changing its state. In general, if E_1 and E_2 are energies before and after a spin value is changed, the transition probability from energy E_1 to E_2 is

$$p(E_1\rightarrow E_2)=\min\left(\frac{g(E_1)}{g(E_2)},1\right). \tag{3}$$

Equation (3) implies that if $g(E_2)\leqslant g(E_1)$, the state with energy E_2 is accepted; otherwise it is accepted with a probability $g(E_1)/g(E_2)$ [that is, the state with energy E_2 is accepted if a random number picked uniformly between 0 and 1 is smaller than or equal to the ratio $g(E_1)/g(E_2)$]. If the trial state with energy E_2 is accepted, we multiply the existing value of $g(E_2)$ by a modification factor $f>1$, that is, $g(E_2)\rightarrow f\times g(E_2)$, and we update the existing entry for $H(E_2)$ in the energy histogram, that is, $H(E_2)\rightarrow H(E_2)+1$. If the random walk rejects the trial move and remains at the same energy level E_1, we modify the existing density of states $g(E_1)$ by the same modification factor; that is, $g(E_1)\rightarrow f\times g(E_1)$, and we update the existing entry for $H(E_1)$; that is, $H(E_1)\rightarrow H(E_1)+1$. Because $g(E)$ becomes very large, in practice it is preferable to work with the logarithm of the density of states, so that all possible $\ln[g(E)]$ will fit into double precision numbers. Therefore, each update of the density of states is implemented as $\ln[g(E)]\rightarrow\ln[g(E)]+\ln(f)$, and the ratio of density of states in Eq. (3) is computed as $\exp\{\ln[g(E_1)]-\ln[g(E_2)]\}$.

A reasonable, although not necessarily optimal choice of the initial modification factor is $f=f_0=e^1\simeq 2.71828$, which allows us to reach all possible energy levels quickly even for a large system. If f_0 is too small, the random walk will spend a very long time to reach all possible energies; however, too large a choice of f_0 will lead to large statistical errors. We proceed with the random walk in energy space until we obtain a ''flat'' histogram $H(E)$. We typically check whether the histogram is flat after every 10 000 MC sweeps, where one MC sweep corresponds to randomly picking N spins and thus generating N trial states (N denotes the total number of spins on the lattice). When the histogram is flat, all the possible energies have been roughly visited an equal number of times, and the density of states converges to the true value with an accuracy proportional to the modification factor $\ln(f)$. We then reduce the modification factor by using a function such as $f_1=\sqrt{f_0}$, reset the histogram to $H(E)=0$ for all values of E, and begin the next level random walk during which we modify the density of states with the smaller modification factor f_1 for each step. Each level random walk is referred to as one iteration in the algorithm. Note that the spin configuration and the density of states are never reset during the simulation. We continue performing the random walk until the histogram $H(E)$ is flat again, and then we reduce the modification factor $f_{i+1}=\sqrt{f_i}$, reset the histogram to $H(E)=0$ for all values of E, and restart the random walk. We stop the simulation when the modification factor is smaller than a predefined value (such as $f_{\text{final}}=\exp(10^{-8})\simeq 1.000\,000\,01$). The modification factor acts as a control parameter for the accuracy of the density of states during the simulation and also determines how many MC sweeps are necessary for the whole simulation.

It is impossible to obtain a perfectly flat histogram and the phrase ''flat histogram'' in this paper means that the histogram $H(E)$ for all possible E is not less than x% of the average histogram $\langle H(E)\rangle$, where x% is chosen according to the size and complexity of the system and the desired accuracy of the density of states. For the 2D Ising model with only nearest-neighbor couplings on small lattices, this percentage can be chosen as high as 95%, but for large systems the criterion for ''flatness'' may never be satisfied if we choose too high a percentage, and the program might run forever.

Clearly, one essential constraint is that $g(E)$ should converge to the true value. The accuracy of the estimate for

$g(E)$ is proportional to $\ln(f)$ at that iteration. However, $\ln(f_{\text{final}})$ cannot be chosen arbitrarily small or the modified $\ln[g(E)]$ will not differ from the unmodified one to within the number of digits in the double precision numbers used in the simulation. If this happens, the algorithm no longer converges to the true value, and the program may run forever. If f_{final} is within the double precision range but is too small, the calculation might take excessively long to finish.

A simple recipe for reducing the modification factor is to take a square-root function, and f approaches 1 as the number of iterations approaches infinity. (There is no reason why any function cannot be used as long as it decreases f monotonically to 1. A simple and efficient formula is $f_{i+1}=f_i^{1/n}$, where $n>1$. The value of n can be chosen according to the available CPU time and the expected accuracy of the simulation. For the systems that have been studied, the choice of $n=2$ yields good accuracy in a relatively short time, even for large systems.)

For the initial modification factor $\ln(f_0)=1$ and the final factor $\ln(f_{\text{final}})=10^{-8}$, the total number of iterations is 27. We do not set a predetermined number of MC sweeps for each iteration, but rather let the program check periodically whether the established criterion for a flat histogram is satisfied. Generally, the number of MC sweeps needed to satisfy the criterion increases as we reduce the modification factor, but we cannot predict the exact number of MC sweeps needed for each iteration before the simulation. It is preferable to allow the program to decide how much simulational effort is needed for a given modification factor f_i. Nonetheless, we need to perform some test runs to make sure that the program will finish within a given time.

The simulation method can be further enhanced by performing multiple random walks, each for a different range of energy, either serially or in parallel. We can restrict the random walk to remain in the range by rejecting any move out of that range.[19,41] The resultant parts of the density of states can then be joined together.

During the random walk (especially in the early iterations), the algorithm does not satisfy the detailed balance condition exactly, because $g(E)$ is modified constantly during the random walk. After many iterations, however, $g(E)$ converges to the true value as the modification factor approaches 1. If $p(E_1\rightarrow E_2)$ is the transition probability from energy E_1 to energy E_2, the ratio of the transition probabilities from E_1 to E_2 and from E_2 to E_1 can be calculated very easily as

$$\frac{p(E_1\rightarrow E_2)}{p(E_2\rightarrow E_1)}=\frac{g(E_1)}{g(E_2)}, \tag{4}$$

where we have used Eq. (3). In other words, the random walk algorithm satisfies the detailed balance:

$$\frac{1}{g(E_1)}p(E_1\rightarrow E_2)=\frac{1}{g(E_2)}p(E_2\rightarrow E_1), \tag{5}$$

where $1/g(E_1)$ is the probability at the energy E_1 and $p(E_1\rightarrow E_2)$ is the transition probability from E_1 to E_2. We conclude that the detailed balance condition is satisfied with accuracy proportional to the modification factor $\ln(f)$.

Almost all recursive methods update the density of states by using the histogram data directly, and only after enough histogram entries are accumulated.[6,9,11,44–51] Because of the exponential growth of the density of states in energy space, this process is inefficient because the histogram is accumulated linearly. Instead, in Wang-Landau sampling we modify $g(E)$ at each step of the random walk, and this modification allows us to approach the true distribution much faster than conventional methods, especially for large systems. (We also accumulate histogram entries during the random walk, but we only use them to check whether the histogram is flat enough to go to the next level random walk.)

Although the total number of configurations increases exponentially with the size of the system, the total number of possible energies increases linearly with the size of system, so it is easy to calculate $g(E)$ with a random walk in energy space for a large system. Consider, for example, a Q-state Potts model on a $L\times L$ lattice with nearest-neighbor interactions.[43] For $Q\geqslant 3$, the number of possible energies is about $2N$, where $N=L^2$ is the total number of the lattice sites. However, the average number of possible states for each energy level is as large as $Q^N/2N$, where Q^N is the total number of possible configurations of the system. This large number is why we cannot simply use a computer to realize all possible states and why efficient and fast algorithms are required.

At the end of the simulation, the Wang-Landau algorithm provides only a relative density of states for different energies. To extract the correct density of states $g_n(E)$ for the Q-state Potts model, we can either use the fact that the total number of possible states is $\Sigma_E g_n(E)=Q^N$, or that the number of ground states (where $E=-2N$) is Q. By using the former rescaling condition, the correct normalized density of states $g_n(E)$ can be obtained from the simulation data $g(E)$, by the relation $\ln[g_n(E)]=\ln[g(E)]-\ln[\Sigma_E g(E)]+N\ln(Q)$, whereas the latter condition leads us to use $\ln[g_n(E)]=\ln[g(E)]-\ln[g(E=-2N)]+\ln(Q)$. For simplicity, we will denote the normalized density of states simply as $g(E)$ in the following. The latter normalization guarantees the accuracy of the density of states at low energy levels, which is important in the calculation of thermodynamic quantities at low temperature. With this normalization, when $T=0$, we can obtain exact solutions for the internal energy, entropy, and free energy when we calculate these quantities from the density of states. If we apply the normalization that the total number of states is Q^N, we cannot guarantee the accuracy of $g(E)$ for energies at or near the ground state, because the rescaling factor is dominated by the maximum density of states. We can use one of these two normalizations to obtain the absolute density of states, and use the other normalization to check the accuracy of the result.

One of the advantages of the Wang-Landau method is that the density of states does not depend on the temperature. For example, the internal energy $U(T)$ can be calculated by

$$U(T)=\frac{\Sigma_E E g(E) e^{-E/k_BT}}{\Sigma_E g(E) e^{-E/k_BT}}\equiv\langle E\rangle, \tag{6}$$

and the specific heat $C(T)$ can be determined from the fluctuations in the internal energy

$$C(T)=\frac{\partial U(T)}{\partial T}=\frac{\langle E^2\rangle-\langle E\rangle^2}{k_BT^2}. \tag{7}$$

We can also access some quantities, such as the Helmholtz free energy and entropy, that are not directly available from

conventional MC simulations. For example, by using conventional MC methods the entropy can be estimated by integrating over other thermodynamic quantities, such as the specific heat, but the result is not always reliable because the specific heat itself is not easy to accurately determine, particularly considering its divergence at a phase transition. However, the free energy $F(T)$ can be calculated directly from the partition function Z using

$$F(T) = -k_B T \ln(Z) = -k_B T \ln\left(\sum_E g(E) e^{-E/k_B T} \right), \quad (8)$$

and the entropy can then be easily computed by

$$S(T) = \frac{U(T) - F(T)}{T}. \quad (9)$$

We point out that even for relatively small lattices, the partition function may be too large to fit into a double precision number, in which case it cannot be easily computed in practice. Nevertheless, the thermodynamic quantities can still be readily computed if we note that

$$\sum_E X(E) g(E) e^{-E/k_B T} = e^{\lambda} \sum_E X(E) e^{\ln[g(E)] - E/k_B T - \lambda}, \quad (10)$$

where $X(E)$ is a general function of E and λ is the largest exponent, $\ln[g(E)] - E/k_B T$. The summation on the right-hand side of Eq. (10) can be computed and the factor e^{λ} does not have to be evaluated. Because this factor appears in the numerator and the denominator of Eq. (6), it cancels. This cancellation also occurs in the evaluation of the specific heat. The free energy, which is proportional to the *logarithm* of the partition function, can also be computed without evaluating e^{λ} explicitly.

Statistical errors in the thermodynamic quantities can be estimated by repeating the simulation several times using different random number sequences, and then computing the averages and fluctuations in these quantities.

With the histogram reweighting method,[13] it is possible to use simulational data at specific temperatures to obtain complete thermodynamic information near, or between, those temperatures. Unfortunately, it is usually quite difficult to obtain accurate information in the region far away from the simulated temperature due to difficulties in obtaining good statistics, especially for large systems where the canonical distributions are very narrow. With Wang-Landau sampling, the histogram is "flat," and we have essentially the same statistics for all energy levels. Because the output of the simulation is the density of states, which does not depend on the temperature, we can then calculate most thermodynamic quantities at any temperature without repeating the simulation. The algorithm is especially useful for obtaining thermodynamic information at low temperatures, or at the transition temperature where the conventional MC algorithm is not so efficient.

III. APPLICATION TO A SECOND-ORDER PHASE TRANSITION

Wang-Landau sampling is very efficient for the study of second-order phase transitions, because it sidesteps critical slowing down at the critical temperature T_c and the slow dynamics at low temperature. To check the accuracy and convergence of the method, we apply it to the 2D ferromagnetic Ising model[42] with nearest-neighbor interactions on a $L \times L$ square lattice with periodic boundary conditions. Each of the $N = L^2$ lattice sites i has a spin denoted as σ_i, which can assume the values $\sigma_i = +1$ for spin up and $\sigma_i = -1$ for spin down. The interaction Hamiltonian is given by

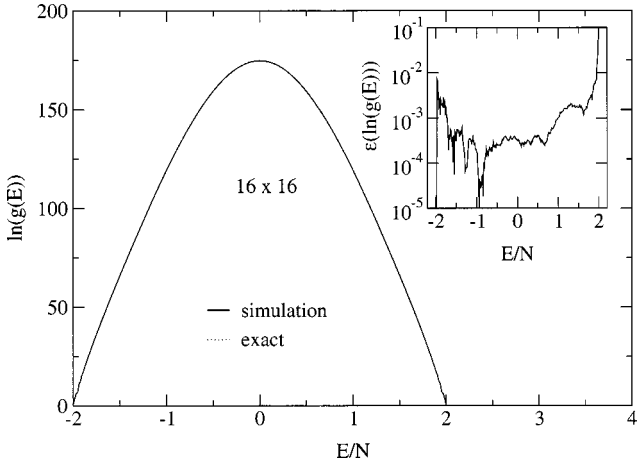

Fig. 1. Logarithm of the density of states, $\ln[g(E)]$, of the 2D Ising model for $L = 16$. The relative errors of the simulational densities of states are shown in the inset.

$$\mathcal{H} = -\sum_{\langle i,j \rangle} \sigma_i \sigma_j, \quad (11)$$

where $\langle i,j \rangle$ denotes distinct pairs of nearest-neighbor sites; the number of energies for this system is $N-1$ for even L. This model provides an ideal benchmark for new algorithms,[13,52] and is also an ideal laboratory for testing theory,[14,53] because this model can be solved exactly.

With the exact solution for the partition function on finite-size systems,[54] and the expansion of the expression by Mathematica, the density of states for the Ising model on a square lattice can be obtained exactly.[14] Beale[14] obtained the exact density of states up to $L = 32$, and using Beale's program, Wang and Landau[19] were able to compute $g(E)$ for $L = 50$. In this paper we show results for $L = 16$, but Wang-Landau sampling has been used to determine $g(E)$ for lattices up to $L = 256$ for which there is currently no exact solution.[19]

The estimate of the density of states for $L = 16$ using Wang-Landau sampling is shown in Fig. 1, along with the exact results by Beale.[14] The initial and final modification factors for the random walks were $\ln(f_0) = 1$ and $\ln(f_{\text{final}}) = 10^{-8}$. The histogram $H(E)$ was considered flat when all entries were not less than 80% of the average $\langle H(E) \rangle$. The absolute density of states in Fig. 1 is obtained by the condition that the number of ground states is 2 for the 2D Ising model. With the logarithmic scale used in Fig. 1, the simulational data and exact solution overlap perfectly with each other. In the inset of Fig. 1, we show the relative error ε, which is defined by the ratio between the error of the simulational data and exact values for any quantity X as

$$\varepsilon(X) \equiv \frac{|X_{\text{sim}} - X_{\text{exact}}|}{X_{\text{exact}}}. \quad (12)$$

We see that $\varepsilon[\ln(g)]$ is smaller than 0.2% for most of the region.

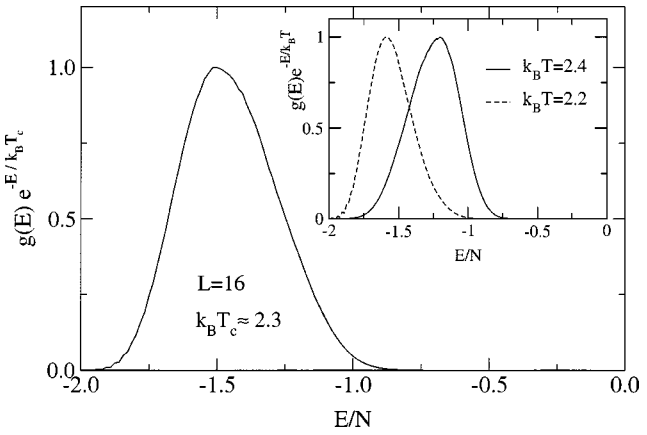

Fig. 2. The canonical distribution at the transition temperature $P(E,T_c)=g(E)e^{-E/k_BT_c}$ for the $L=16$ Ising model. The inset shows the canonical distribution at a temperature slightly above and below T_c for the same system.

We can calculate the canonical distribution using Eq. (1) at essentially any temperature without performing multiple simulations. In Fig. 2 we show the resultant canonical distribution at the critical temperature T_c, which exhibits a single peak. The distributions at temperatures above and below T_c are also single peaked, as illustrated in the inset of Fig. 2.

It is also important to study the influence of the errors in the density of states on the calculated thermodynamic quantities. In Fig. 3 we show the internal energy, the specific heat, the Helmholtz free energy, and the entropy as a function of temperature for $L=16$. Both the simulational results computed with Eqs. (6)–(9), and the exact solutions are plotted and overlap almost perfectly over a wide temperature region from $k_BT=0-8$. Because no difference is visible in these figures, more stringent tests of the accuracy are provided by the insets, which show the relative errors for the respective thermodynamic quantities. The relative errors are quite small for the entire temperature region from $k_BT=0-8$.

Note that because the system has a second-order phase transition, the first derivative of the free energy is a continuous function of temperature. There are no jumps in either the internal energy or the entropy even in the limit as the system size goes to infinity.

The random number generator used in our simulation was a shift-register algorithm denoted as R1279.[1] The average number of visits to each energy for the entire duration of the simulation (adding the average number of visits for all iterations) was roughly 10^6. The CPU time of the simulation to obtain the density of states shown in Fig. 1 was less than 3 min using a GNU compiler on a Pentium 4 (1.3 GHz) processor.

IV. APPLICATION TO A FIRST-ORDER PHASE TRANSITION

In this section, we apply the algorithm to a model with a first-order phase transition.[55,56] In such cases, the internal energy and the entropy have discontinuities at the transition, at which both ordered and disordered states coexist. We consider the 2D $Q=8$ Potts model[43] on $L\times L$ square lattices with nearest-neighbor interactions and periodic boundary conditions. The total number of spins is $N=L^2$, and the Hamiltonian can be written as

$$\mathcal{H}=-\sum_{\langle i,j\rangle}\delta(q_i,q_j), \tag{13}$$

where $q_i=1,2,\ldots Q$ denotes the Potts spin at site i and $\delta(q_i,q_j)$ is a Kronecker delta. During the simulation, we select lattice sites randomly and choose integers between $[1,Q]$ randomly for new Potts spin values. The modification

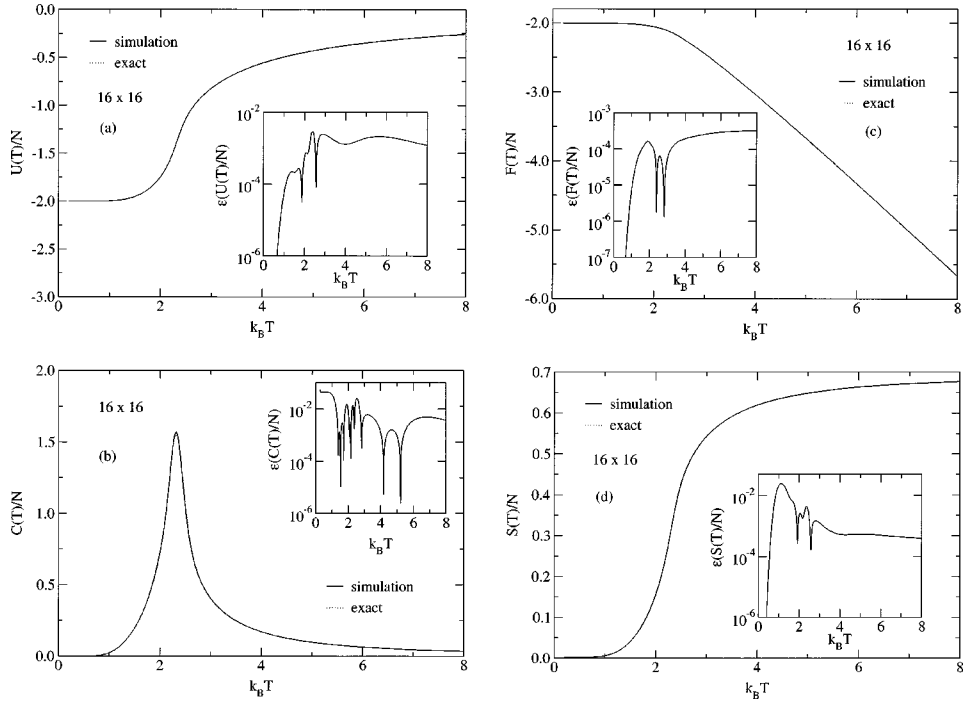

Fig. 3. Thermodynamic quantities for the $L=16$ 2D Ising model calculated from the density of states. The relative errors with respect to the exact solutions by Ferdinand and Fisher[54] are shown in the insets: (a) internal energy, (b) specific heat, (c) Helmholtz free energy, and (d) entropy.

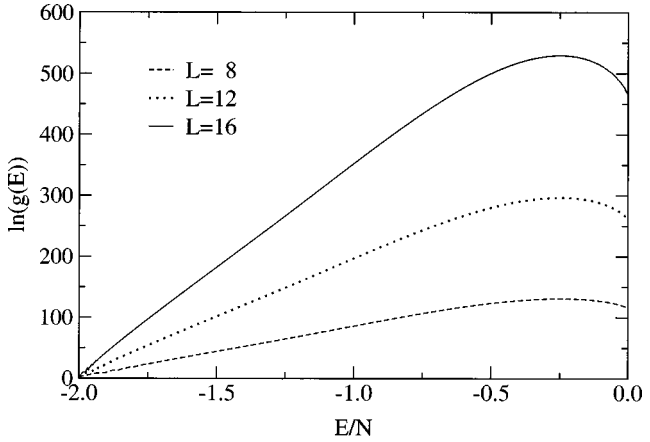

Fig. 4. Logarithm of the density of states $g(E)$ for the 2D $Q=8$ Potts model as a function of energy per lattice site, E/N, for $L=8$, 12, and 16. With the scale in the figure, the errors of the simulational data are within the width of the lines.

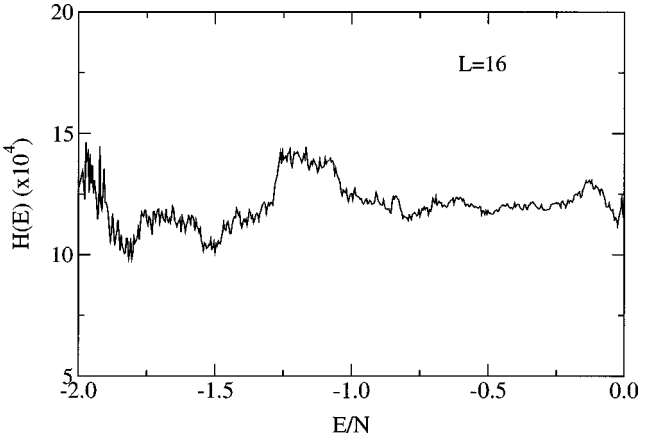

Fig. 6. Final histogram of energy for the last iteration of random walks to estimate the density of states of a $Q=8$ Potts model for $L=16$.

factor $\ln(f_i)$ changes from $\ln(f_0)=1$ at the beginning, to $\ln(f_{\text{final}})=10^{-8}$ by the end of the random walks. The histogram of energy $H(E)$ is considered flat when all entries are not less than 80% of the average $\langle H(E)\rangle$. To guarantee the accuracy of thermodynamic quantities at low temperatures, we use the condition that the number of the ground states is $Q=8$ to normalize the density of states. The densities of states for $L=8$, 12, and 16 lattices are shown in Fig. 4. We see that the maximum density of states from our data for $L=16$ is very close to e^{530}, which is about 1.5×10^{230}.

In Fig. 5 we show the double-peaked canonical probability distribution[56] at the transition temperature T_c for the first-order transition, computed from the simulational data using Eq. (1). The "transition temperature" $k_BT_c(L)$ is approximately 0.7519 for $L=16$ and is the temperature where the double peaks are of the same height. The transition temperature for the infinite lattice is known exactly to be $k_BT_c=1/\ln(1+\sqrt{Q})\approx0.7449$.[43] The valley between the two peaks is approximately 0.37 for $L=16$, and becomes deeper as L increases. The latent heat for this temperature-driven first-order phase transition can be estimated from the energy difference between the double peaks. When T is slightly away from T_c, one of the double peaks increases dramatically in magnitude and the other decreases as shown in the inset of Fig. 5.

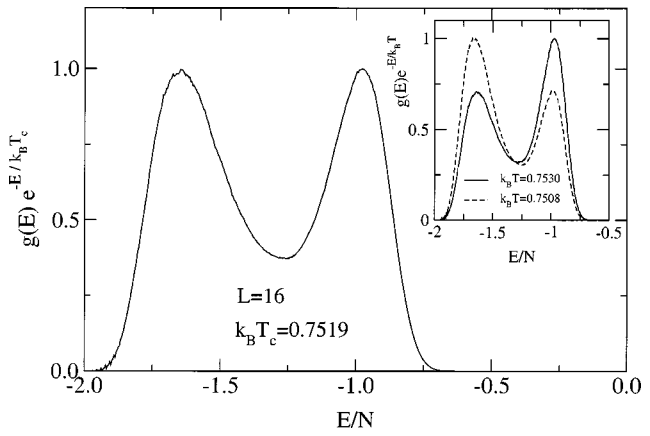

Fig. 5. The canonical distribution at the transition temperature $P(E,T_c)=g(E)e^{-E/k_BT_c}$ for the $Q=8$ Potts model for $L=16$. The inset shows the canonical distribution at a temperature slightly above and below T_c for the same system.

Because of the double-peaked structure at a first-order phase transition, conventional MC simulations are not efficient because an extremely long time is required for the system to travel from one peak to the other in energy space. With the Wang-Landau algorithm, all possible energy levels are visited with equal probability, so it overcomes the barrier between the coexisting phases in the conventional MC simulations. The final flat histogram for $L=16$ is shown in Fig. 6, and it describes the total number of visits to each energy level for the random walk of the last iteration.

Figure 7 illustrates some thermodynamic quantities calculated from the density of states using Eqs. (6)–(9). Near the transition temperature T_c, the internal energy, shown in Fig. 7(a), has a steplike change that becomes sharper as the lattice size increases and transforms into a discontinuous jump when the system size goes to infinity. The magnitude of this jump [shown in Fig. 7(a) for an infinite lattice] equals the latent heat for the phase transition.

The specific heat, shown in Fig. 7(b), has a peak in the vicinity of T_c, and both the maximum value and the position of the peak depend on the finite size of the lattice. As L increases, the peak in the specific heat becomes narrower and goes to a delta function in the thermodynamic limit.

Our results for the Helmholtz free energy per lattice site are shown in Fig. 7(c) as a function of temperature. Because the transition is of first-order, the first derivative of the free energy has a discontinuity at T_c. (The location of this discontinuity can be used as an estimate of T_c.)

Like the internal energy, the entropy shown in Fig. 7(d) has a steplike change near T_c. This change becomes sharper as L increases and becomes a discontinuous jump when $L\to\infty$. Because the jump in the internal energy equals the latent heat of the phase transition, and the free energy is continuous at T_c, the magnitude of the jump in the entropy equals the latent heat divided by T_c [see Eq. (9)].

V. RANDOM WALK IN ENERGY AND ORDER PARAMETER SPACE

To study the effect of an applied magnetic field on the Ising and Potts models, we have to perform a random walk

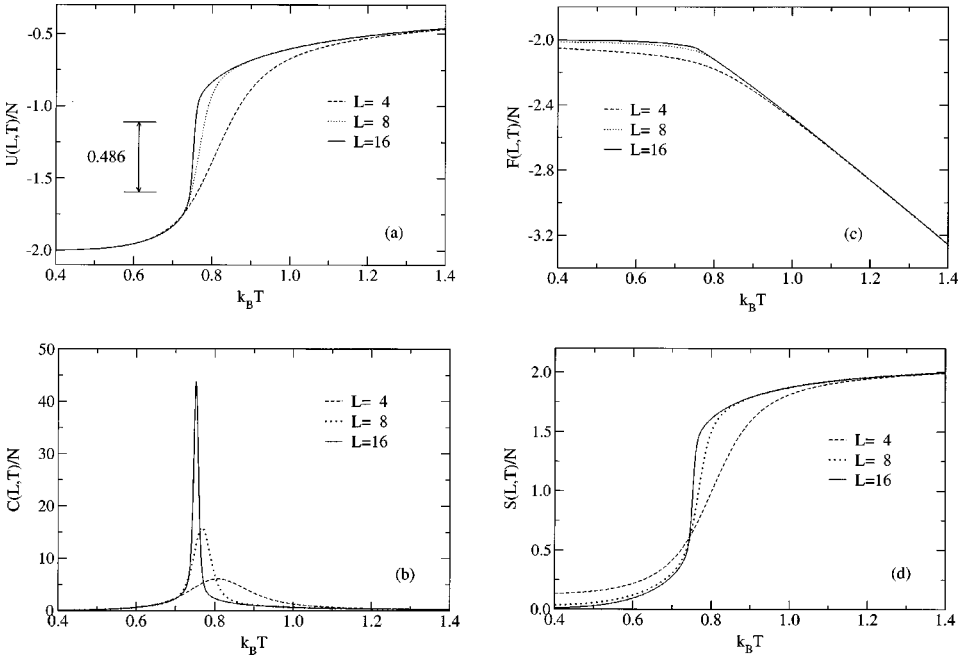

Fig. 7. Thermodynamic quantities calculated from the density of states for the $Q=8$ Potts model for $L=4$, 8, and 16: (a) internal energy, (b) specific heat, (c) Helmholtz free energy, and (d) entropy.

in both the energy and order parameter space. 2D random walks may also be required to study systems with more complex orders, such as a three-dimensional spin glass model, even in the absence of external fields.

To illustrate a simple case where a 2D random walk is required, we consider the 2D Ising model in the presence of an external magnetic field h. The Hamiltonian is given by

$$\mathcal{H}=-\sum_{\langle i,j\rangle}\sigma_i\sigma_j-h\sum_{i=1}^{N}\sigma_i. \tag{14}$$

The order parameter is the magnetization, defined as $M'=\Sigma_{i=1}^{N}\sigma_i$, and we denote the exchange energy as $E'=-\Sigma_{\langle i,j\rangle}\sigma_i\sigma_j$. The algorithm works as before, except that the random walk is now performed in both the energy E' and the order parameter M', and a 2D histogram $H(E',M')$ is accumulated.

With the estimate of the density of states $g(E',M')$, the partition function can be computed as

$$Z(T,h)=\sum_{E',M'}g(E',M')e^{-(E'-hM')/k_BT}. \tag{15}$$

From the partition function we can obtain thermodynamic quantities for all values of the temperature and magnetic field. For example, the mean magnetization of the system can be computed as

$$M(T,h)=\frac{\Sigma_{E',M'}M'g(E',M')e^{-(E'-hM')/k_BT}}{\Sigma_{E',M'}g(E',M')e^{-(E'-hM')/k_BT}}. \tag{16}$$

$M(T,h)$ is shown in Fig. 8 as a function of the external magnetic field h, for different values of the temperature T. Note that for fixed $T<T_c$ the Ising model has a first-order phase transition at $h=0$. Standard MC methods generate hysteresis in the magnetization curve at low T (shown as the solid line in Fig. 8), because of metastable states that appear near first-order phase transitions. This metastability hinders studies of first-order phase transitions using standard MC methods.

VI. DISCUSSION AND CONCLUSION

We have described an efficient algorithm to calculate the density of states directly for large systems. By modifying the estimate at each step of the random walk in energy space and carefully controlling the modification factor, we can determine the density of states very accurately. Using the density of states, we can then calculate thermodynamic quantities at essentially any temperature. An important advantage of this approach is that we can also calculate the Helmholtz free energy and entropy, quantities that are not directly available

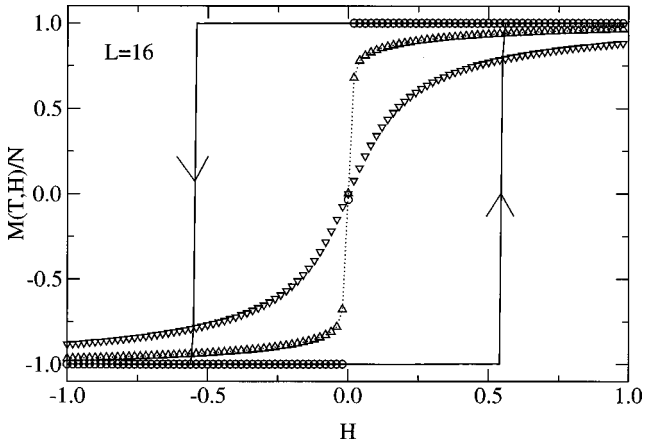

Fig. 8. Magnetization per site for the $L=16$ Ising model as a function of applied field for $T=1.0/k_B<T_c$ (circle), $T=2.3/k_B\approx T_c$ (upright triangle), $T=3.0/k_B>T_c$ (inverted triangle) obtained with Wang-Landau sampling, and the hysteresis curve obtained with the Metropolis algorithm at $T=1.0/k_B$ (solid line). The dotted line is a guide to the eye.

from conventional MC simulations. The method is applicable to a wide range of systems and is easy to implement. Although we have described its implementation in terms of single spin-flip sampling, it is straightforward to use other types of sampling for cases in which it will further accelerate the simulation.

Applications to the 2D $Q=8$ Potts model and to the 2D Ising model show that the method is effective for systems that exhibit first-order or second-order phase transitions. Our presentation concentrated on the random walk in energy space (and order-parameter space); however, the idea is very general and can be applied to any parameters. The energy levels of the models treated here are discrete, and the total number of possible energies is known before the simulation, but in general such information is not available. For models where all the possible energy levels cannot be fitted in the computer memory or the energy is continuous, for example, the Heisenberg model, we must bin the energy. Statistical and systematic errors in the density of states, and thus of the thermodynamic quantities, are controlled by the flatness of the histogram at the end of each iteration and the final modification factor f_{final}. These errors can be decreased by requiring a more strict condition for a flat histogram and by using a $\ln(f_{\text{final}})$ that is closer to zero.

In this paper, we only applied the Wang-Landau algorithm to simple models on small lattices, but the method is also efficient for large systems and has proven to be useful in the studies of general, complex systems with rough landscapes (see references given in Sec. I). However, more investigation is needed to better determine under which circumstances the method offers substantial advantages over other approaches.[57]

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