Improved Wang-Landau sampling through the use of smoothed potential-energy surfaces

Phuong H. Nguyen[a)]
Institute of Physical and Theoretical Chemistry, J. W. Goethe University, Marie-Curie-Strasse 11, D-60439 Frankfurt, Germany

Emil Mittag[b)] and Andrew E. Torda
Center for Bioinformatics, University of Hamburg, Bundesstrasse 43, D-20146 Hamburg, Germany

Gerhard Stock
Institute of Physical and Theoretical Chemistry, J. W. Goethe University, Marie-Curie-Strasse 11, D-60439 Frankfurt, Germany

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A method is presented to improve the speed of convergence of Wang-Landau simulations as used to calculate the density of states of continuous systems. The density of states is first crudely estimated with calculations employing a smoothed potential-energy surface. This estimate is then used as a seed for subsequent Wang-Landau simulations using the original potential. The performance of the method is demonstrated by employing several simple models, including an analytically solvable harmonic system as well as a Gō model of a protein. For all systems considered, the seeded simulations were found to converge significantly faster and with higher accuracy than the standard Wang-Landau simulations. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2191060]

I. INTRODUCTION

The density of states is a sought-after quantity, especially in complex systems. If it is known, it can be used to calculate thermodynamic properties of a system via standard relationships.[1] In other contexts, it may serve to provide sampling weights for efficient Monte Carlo schemes.[2,3] Here, we propose a method purely aimed at speeding up the estimation of the density of states and apply it in the framework of the recursion introduced by Wang and Landau.[4,5]

Obviously, the density of states is intimately tied to the statistical description of a system. Given some temperature T, it is the key to the canonical probability distribution

$$P(E) = g(E)\exp(-\beta E), \tag{1}$$

where $\beta = 1/kT$, k is Boltzmann's constant, and $g(E)$ is the density of states as a function of energy. Rather than treating the density of states as an end in itself, it may have another important role. Estimation of the density of states is the first step in the Wang-Landau procedure,[4,5] which is an example of a broader class of two stage simulation methods.

In conventional Metropolis Monte Carlo,[6] one uses the Boltzmann weight of an energy ($e^{-\beta E}$) as the basis of the acceptance criterion, but this will not always be the best choice. Given the exponential term, there may only be a minute probability of visiting high energy states and escaping from local energetic minima. With the introduction of umbrella sampling came the idea that perhaps one should aim for uniform sampling of some other property.[7] This idea is probably best known from the multicanonical approach.[8,9] Here, one uses an initial simulation to obtain weights which can then be used for a single long simulation afterwards. The ideas have been generalized and applied in many ensembles[10–14] and to ever larger systems.[15–18]

In a related method, the so-called Wang-Landau scheme, a different recursion was proposed for obtaining the density of states of a system and perhaps using this as the basis for the weighting in a single longer simulation.[4,5] The idea is that the density of states can be estimated quite directly from a simulation where the acceptance criterion is explicitly based on the history of visiting different energy levels. This method has been applied in various contexts, ranging from random spin models[19] and quantum systems[20] to simple models for proteins.[21] Despite the advantages, it can still be difficult to obtain a reliable density of states with the Wang-Landau method. For example, it has not yet been proven if the Wang-Landau recursion necessarily leads to the true density of states or if a subsequent conventional Monte Carlo simulation is necessary for consistency reasons.[3] Furthermore, the general problem may remain that it is difficult to obtain sufficient sampling of a system's low energy configurations.[22,23] These configurations are both relevant to real world temperatures and essential for convergence of the approach. This is the issue with which this work is concerned. Can one simply speed up the convergence for the initial step of a Wang-Landau calculation?

The approach used here is to apply a general and controllable method for smoothing a potential-energy surface. On this surface, one begins a standard Wang-Landau calculation to provide a crude, but well-sampled initial estimate of the density of states. In a second step, one restores the true

[a)]Corresponding author. Electronic mail: phuong@theochem.uni-frankfurt.de
[b)]Corresponding author. Present address: Research School of Chemistry, Australian National University, Canberra ACT 0200, Australia. Electronic mail: mittag@rsc.anu.edu.au

surface and continues to improve the estimation of the density of states. Effectively, the initial simulation serves a seeding for a conventional calculation. Intuitively, it is hard to imagine the method producing worse results than a conventional Wang-Landau simulation since even a crude estimate of the density of states should be better than the normal initialization, $g(E)=1$.

Within the scope of this paper, we are solely concerned with the estimation of the density of states and not their application afterwards. This means that we first consider a simple system where one can compare with analytical results before moving to a very idealized model of a protein. It also means that we judge the results in terms of the conventional Wang-Landau convergence criteria and not by a simulation of the system using the implied weights.

II. METHODOLOGY

A. Wang-Landau method

In the Wang-Landau method,[4,5] the probability P_{acc} of accepting a move from an energy level E_1 to a new trial level E_2 is given by

$$P_{\mathrm{acc}}(E_1 \to E_2) = \min\left(1, \frac{g(E_1)}{g(E_2)}\right), \tag{2}$$

where $g(E)$ denotes the current estimate of the density of states for the system. As a consequence of the criterion, a "flat" histogram of energies, $H(E)$, is generated. Before running the simulation, an energy range is specified and discretized into bins, and the initial density of states of the system is set to a uniform arbitrary value, e.g., $g(E)=1$. As the simulation progresses, the density of states is updated according to $g(E) \to f g(E)$, where f is a convergence factor and $g(E)$ is the density of states of the current or trial state, depending on if a trial configuration is rejected or accepted. Every time $g(E)$ is modified, the histogram of energies $H(E)$ is also updated. If all energy levels are about equally well sampled, the histogram becomes flat, defined as $H(E)/\langle H(E)\rangle > s$ for all values of E and where $s = 0.8\langle H(E)\rangle$ is a typical value. Once this flatness criterion for the histogram is satisfied, the value of the convergence factor f is modified such that it is monotonically decreasing, e.g., $f_{\mathrm{new}} = \sqrt{f_{\mathrm{old}}}$. The histogram of energies is then set to zero, and the procedure is repeated with the updated convergence factor f used to modify $g(E)$. The process is repeated until the value of f reaches a predefined value f_{final}. In this work, we used initial values of $f=\exp(10^0)$ and $f_{\mathrm{final}}=\exp(10^{-8})$ for the convergence criterion.

B. Generalized effective potential

To define a smoothed energy landscape, several approaches have been suggested.[16] Recently, a method based on the Tsallis statistic[24] was proposed by Andricioaei and Straub[25,26] and has been shown to be quite efficient in biomolecular simulations.[27–29] In this approach, the potential energy surface (PES) can be made arbitrarily flat by defining a generalized effective potential $V_q(x)$, which is obtained by applying a simple analytical transformation to the original potential-energy function $V(x)$ through

$$V_q(x) = \frac{q}{\beta(q-1)} \ln(1 + \beta(q-1)[V(x) + \epsilon]). \tag{3}$$

Here, ϵ is a constant that shifts the base line of the deformed energy surface and the smoothness of the PES is controlled by the parameter $q>1$. That is, the potential becomes smoother as the value of q increases and for $q \to 1$, $V_q(x) = V_q(x) + \epsilon$. In this work, we set $\epsilon=0$ and measure the energy in units of kT, i.e., $E=\beta V$. This yields

$$E_q(x) = \frac{q}{(q-1)} \ln(1 + (q-1)E(x)), \tag{4}$$

showing that the smoothness of the PES is controlled by the single parameter $1-q$, which in this work ranges between 10^{-3} and 10^{-2}.

As an illustrative example, Fig. 1 shows the double-well potential defined below in Eq. (7) and its smoothed versions for $q=1.0025$ and 1.05. It is seen that transformation (4) essentially reduces the barriers of the PES, while the positions of local minima and maxima remain unchanged. As a consequence, it may be expected that the density of states of the smoothed system is qualitatively similar to its real counterpart. By making the potential flatter, however, the position range, required to cover a given energy interval, increases. For example, to cover energies up to 100, the original $(q=1)$ potential-energy curve requires a range of $-3 \lesssim x \lesssim 4$, while for $q=1.0025$ this range increases to $-9 \lesssim x \lesssim 9$ and much larger for $q=1.05$. As a Monte Carlo sampling obviously becomes more time consuming with increasing x range

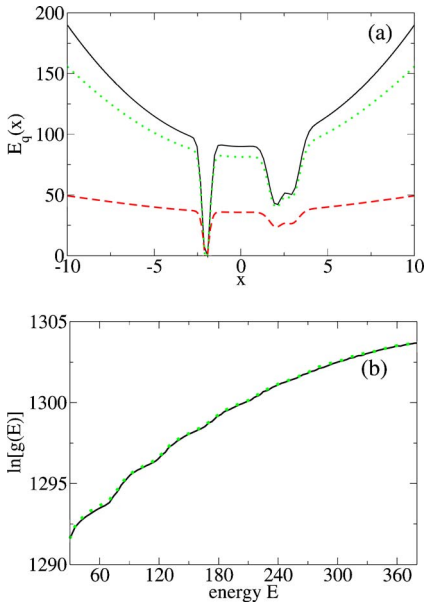

FIG. 1. (Color online) (a) Potential-energy curve of the double-well model [Eq. (7)]. Compared are the original potential ($q=1$, black solid line) and smoothed potentials obtained for the smoothness parameters $q=1.0025$ (green dashed line) and $q=1.05$ (red dotted line). (b) Density of states for a system of four particles, each modeled by the double-well potential. Compared are standard (black solid line) and seeded (green dashed line) Wang-Landau simulations.

to cover, Fig. 1 also serves to indicate a practical consideration in choosing the smoothness parameter q. That is, q should be large enough to allow for an efficient sampling of the overall PES and, at the same time, as small as possible to keep the x range small. From this explanation, it is clear that the optimal choice of the smoothness parameter will depend on the system under consideration. For the considered double-well potential, the choice of $q=1.0025$ appears to be a reasonable compromise that was chosen in the calculations of the density of states described below.

C. Seeding of Wang-Landau simulations

As explained in the Introduction, we wish to combine the PES smoothing and the Wang-Landau method in the following way: (i) First, Wang-Landau simulations on a smoothed PES are performed which provide an overall yet rough estimate for the density of states. (ii) Employing the original potential, this estimate is used as a seed for subsequent Wang-Landau simulations which provide accurate results for the density of states.

It is important to note that the simulations on the smoothed PES need not run so long that the density of states converges. Since the seeding simulations are supposed to yield a well-sampled but rough estimate for the density of states, the simulations on the smoothed PES can be stopped once the density of states exhibits the typical overall increase with energy. For the systems under consideration, the seeding simulations were usually run until the convergence factor f was rescaled about ten times. Extending the seeding simulations for longer times hardly improves the convergence of the subsequent Wang-Landau simulations. On the other hand, stopping the seeding simulation after only a few rescalings of f, the subsequent Wang-Landau simulations took almost as long to converge as for standard unseeded calculations. Apart from the smoothness variable q, the proposed procedure therefore introduces a second system-dependent parameter, which needs to be determined. However, since in practice the effort of the seeding calculations is negligible compared with the second part of the procedure, the number of rescalings in the seeding calculations is not critical.

To give a better idea of typical performance of the proposed method, we conducted 100 independent simulations with different random number seeds. In the discussion below we are mainly concerned with the average of the logarithm of the density of states, which is given by

$$\langle \ln[g(E)] \rangle = \frac{1}{n} \sum_{i=1}^{n} (\ln[g_i(E)] + C_i). \tag{5}$$

Because the Wang-Landau method determines the density of states only up to a multiplicative constant, the $g_i(E)$ of each individual run needs to be shifted by a constant C_i. To determine these constants, we define the corresponding statistical error of $\ln g$ as[22,23]

$$\Delta = \sum_{l} \sum_{i=1}^{n} \left(\ln[g_i(E_l)] + C_i - \frac{1}{n} \sum_{j=1}^{n} [\ln[g_j(E_l)] + C_j] \right)^2, \tag{6}$$

where the index l labels energy states E_l. Setting $C_1=0$ and minimizing the total variance Δ with respect to the C_i, the constants are readily obtained.

III. RESULTS

A. Noninteracting particles model

As a first simple model system, we consider a model of four noninteracting particles, each modeled by a one-dimensional double-well potential of the form

$$E(x) = x^2 - 100e^{-10(x+2)^2} - 50e^{-3(x-2)^2} - 45e^{-3(x-3)^2} + 90. \tag{7}$$

As shown in Fig. 1(a), the parameters have been chosen such that the potential comprises a narrow deep minimum as well as a shallow higher-lying minimum. As a consequence of the shape of the potential energy, the density of states of the system is expected to rise in a nonuniform way. Also shown is the generalized effective potential for the smoothness parameter $q=1.0025$, for which the seeding calculations have been performed. All simulations used 150 histogram bins of equal width and a histogram flatness criterion of 80%.

For this system, Fig. 1(b) shows a comparison of the density of states obtained from a standard and a seeded Wang-Landau simulation. While both calculations essentially agree with each other, the seeded calculations were faster by a factor of about 2. That is, the unseeded simulations required on average $(5.9\pm1)\times10^7$ trial moves to converge, while the average number of trial moves for the simulations on the smoothed PES was $(2.2\pm0.2)\times10^6$ and the subsequent seeded simulations required $(2.9\pm0.9)\times10^7$ trial moves.

As a supposedly even simpler model, we next consider a four-particle model using noninteracting harmonic oscillators with potential energy $E(x)=\frac{1}{2}x^2$. Since a harmonic potential only exhibits a single minimum, it represents a somewhat pathological case for a method designed for rugged PES. This is reflected in the fact that only a very small smoothness parameter $q=1.0005$ proved advantageous for the harmonic model. Using 50 histogram bins of equal width and a histogram flatness criterion of 80%, Fig. 2(a) shows a comparison of the density of states obtained from a standard and a seeded Wang-Landau simulation. Again, both calculations essentially agree with each other. Moreover, the seeded calculations were still faster by a factor of about 2. Remarkably, the proposed computational approach is still advantageous, although an obviously unfavorable problem was chosen.

Since the density of states of an N-dimensional harmonic system is available in analytical form $[g(E) \propto E^{N-1}]$, the harmonic model may readily be employed to study the effect of the histogram flatness criterion in Wang-Landau simulations. To this end, we performed two additional simulations using the same conditions, but with the histogram flatness criterion set to 60% or 95% for both the seeding and subsequent Wang-Landau simulations. Figure 2(b) compares the thus

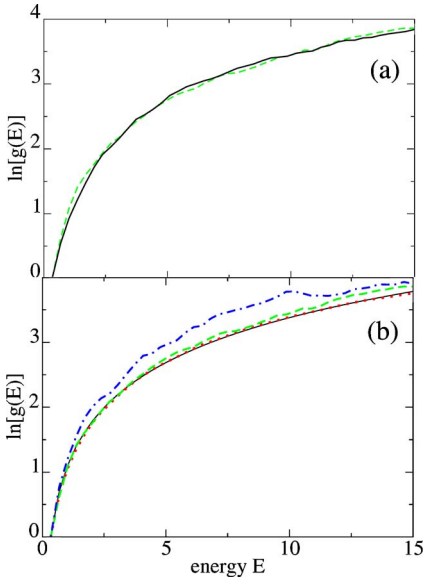

FIG. 2. (Color online) The density of states of a harmonic four-particle model. (a) Comparison of standard (black solid line) and seeded (green dashed line) Wang-Landau simulations. (b) Seeded simulations using flatness criteria of 60% (blue dashed dotted line), 80% (green dashed line), and 95% (red dotted line) are compared with exact analytical results (black solid line).

obtained density of states with the histogram flatness criteria of 60%, 80%, and 95% with the exact analytical result. Generally speaking, by requiring a flatter histogram one expects a higher accuracy of the calculation, albeit at the cost of a longer computation time. As might be expected, the agreement using a 95% flatness criterion is excellent. While at 60% flatness the deviation from the analytical result is significant; the results are quite adequate at 80% flatness, which is the value also suggested by Wang and Landau.[4]

B. Protein model

Finally, we consider a Gō-type model[30] of a 20-amino-acid protein, whose structure was taken from the protein data bank[31] [pdb code 1APM (Ref. 32)]. The main properties of this very simple model are that (i) the protein is a chain of particles, each centered at the C^{α} position and representing a whole residue and (ii) inter-residue interactions are described by harmonic terms centered at the native configuration. The potential energy of the model can be written as

$$E(d) = \sum_{i=1}^{N-1} \sum_{j>i}^{N} (E_{ij}^{I}(d) + E_{ij}^{\mathrm{NI}}(d)), \tag{8}$$

where $E_{ij}^{I}(d)$ denotes the potential felt by two residues that are regarded as interacting in the native state. It is given by

$$E_{ij}^{I}(d) = k_I (d_{ij} - d_{ij}^{(0)})^2, \tag{9}$$

where k_I represents the force constant of the interaction, $d = \{d_{ij}\}$ represents the distances between residues i and j at a given step of the simulation, and $d_{ij}^{(0)}$ is the native-state distance between those two residues. In order to prevent natively noninteracting residues from coming too close to each other during the simulation, the second term

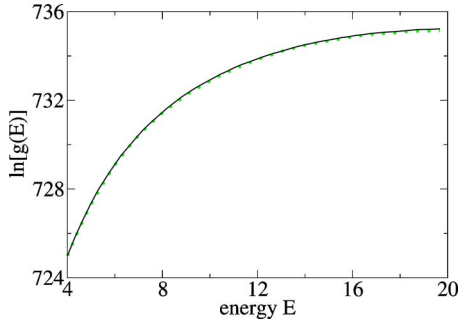

FIG. 3. (Color online) The density of states for a Gō model of a 20 residue protein. Compared are standard (black solid line) and seeded (green dashed line) Wang-Landau simulations.

$$E_{ij}^{\mathrm{NI}}(d) = \begin{cases} k_{\mathrm{NI}}(d_{ij} - d_{ij}^{(0)})^2, & d_{ij} \leqslant d_C \\ 0, & d_{ij} \geqslant d_C \end{cases} \tag{10}$$

introduces a repulsion between the two residues whenever their distance becomes smaller than a cutoff radius d_C. The parameters of the present model are $d_C = 6.4$ Å, $k_I = 1.0$ Å^{-2}, and $k_{\mathrm{NI}} = 0.2$ Å^{-2}. Despite its simplicity, the Gō model gives rise to a complicated energy landscape with numerous local minima.[30]

To generate the generalized effective potential for the Gō model, a smoothness parameter $q = 1.01$ was employed. The energy range was discretized into 50 energy bins of equal width and the criterion for histogram flatness was set to 80%. Comparing again standard and seeded Wang-Landau simulations, Fig. 3 shows the results of the density of states for the protein model. Being in perfect agreement, the seeded and unseeded simulations required on average $(5.9 \pm 4) \times 10^6$ and $(16.6 \pm 2) \times 10^6$ trial moves, respectively, thus resulting in roughly a threefold speedup.

To study the performance of the proposed method in more detail, it is instructive to consider the convergence be-

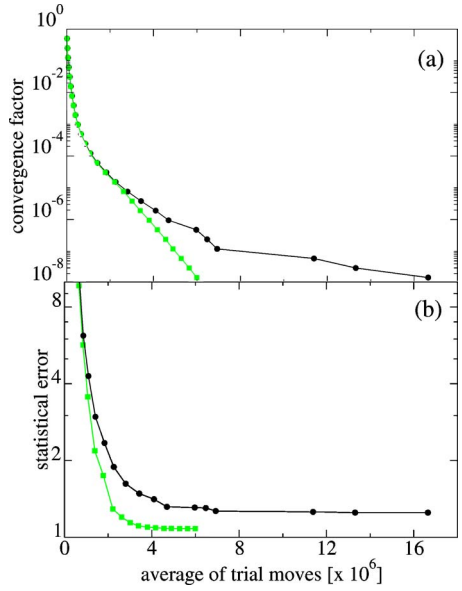

FIG. 4. (Color online) Convergence behavior of the standard (black solid line) and seeded (green dashed line) Wang-Landau simulations as obtained for the protein model. Shown are (a) the convergence factor $\ln f$ and (b) the statistical error Δ of $\ln g(E)$ as defined in Eq. (6).

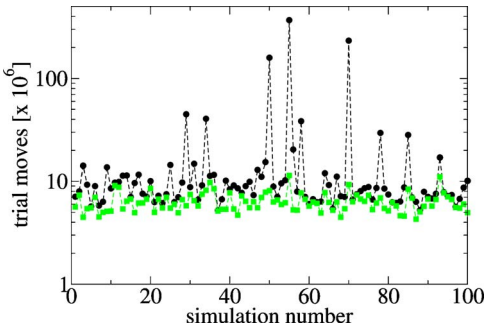

FIG. 5. (Color online) Number of trial moves required for 100 statistically independent Wang-Landau simulations, comparing again standard (black dots) and seeded (green squares) calculations for the protein model. The lines only serve as a guide for the eyes.

havior of the seeded and unseeded simulations. To this end, we consider the convergence factor f of the Wang-Landau simulation as well as the statistical error of the density of states, defined in Eq. (6). Using the protein model as a representative example, Fig. 4(a) shows the logarithm of f versus the average number of trial moves. Each marked point shows the time (in terms of the number of trial moves) when the convergence factor was rescaled. While initially seeded and unseeded simulations rescaled f quite similarly, at later times the seeded simulations become more efficient and converge clearly faster than their unseeded counterpart. The difference is even more significant for the mean statistical error [see Fig. 4(b)], which is seen to decrease rapidly as the number of trial moves increases. That is, the seeded simulations clearly converge faster and with higher accuracy. We note in passing that according to Fig. 4(b) the last 50% of the calculation did not help to improve the accuracy, despite additional rescaling of the convergence factor f. This well-known shortcoming of (both seeded and unseeded) Wang-Landau simulations has given rise to the development of improved variants of the Wang-Landau algorithm.[23]

In practical applications, of course, only a single calculation of the density of states rather than an average over several simulations is performed. Apart from the average convergence behavior discussed above, it is therefore of interest to consider the convergence of the individual simulations. To this end, Fig. 5 displays the total number of trial moves of all simulations. While the seeded simulations converge always faster than the unseeded ones, it interesting that the unseeded calculations show a much larger variance of the number of trial moves. It seems that without the seeding, the system spends much time stuck in energy bins which are difficult to emerge from. As a consequence, in numerous cases, the difference in efficiency of the two methods is about an order of magnitude.

IV. CONCLUSIONS

In order to calculate the density of states for a continuous system, we have proposed the use of a smoothed PES to obtain a seeded estimate for subsequent Wang-Landau simulations. For all the systems considered, a suitably seeded simulation converged significantly faster and with higher accuracy than the standard procedure. Because of the simple generalized effective potential for the seeding calculations, the method is general as well as very easy to implement.

Since the performance of Monte Carlo calculations may be sensitive to the details of the simulation, we have discussed the parameter dependence of the proposed approach. First, just as in the standard Wang-Landau method, the range and binning of the energy levels need to be fixed. This choice certainly depends on the system under consideration. On the other hand, the choice of the histogram flatness criterion seems to be fairly generic. Although it was only plotted for the harmonic model, we found for all three systems that a flatness criterion higher than 80% hardly leads to a significant improvement of the accuracy. For the seeding calculations two more parameters need to be chosen. The number of required rescalings of the convergence factor f (typically 10) assures that the seeding calculations provide an overall yet rough estimate of the density of states. Because of the negligible overall cost of the seeding calculations, the exact choice of this number is not critical. Finally, the value of the smoothness parameter q needs to be fixed such that q is large enough to allow for an efficient sampling of the overall PES and, at the same time, as small as possible to keep the x range small in the Monte Carlo calculations.

There are also possible improvements to the approach. The smoothing procedure chosen in this work is quite crude, in that we move in a single step from a deformed ($q>1$) to the original ($q=1$) PES. Alternatively, one could employ an annealing procedure which gradually decreases q. This would make the initial guess of the smoothness parameter q less critical. In this work, we chose the Tsallis statistic approach to smooth the potential-energy surface, because of its generality and the fact that it preserves the location of energetic minima. One could, of course, smooth an energy surface using ideas such as the diffusion equation method,[33,34] possibly augmented with soft-core terms.[35]

Once one is content with the performance of the method, there are several applications and extensions. Firstly, one should note that if the density of states estimate is only used to obtain a weighting in a subsequent simulation, then it need not be perfect.[36] One could use the quick, imperfect estimate of the density of states from a smoothed PES to generate better weights for a longer simulation. Unfortunately, for large or complex systems, it is difficult to know in advance just how crude the crude estimate of the density of states is. Continuing in this vein, one may note that there are several ways to obtain the weights for a multicanonical ensemble or one of the related methods. These techniques also have rather poor initialization conditions, so they should, in principle, also benefit from a seeding procedure. Without implementing all the possibilities, one should not speculate too much about the success. Some reports suggest that different generalized ensemble approaches have similar performance,[37] but that the convergence properties may be very different.[38,39]

As described, the methodology appears useful, but it will benefit from experience with more realistic systems. Ultimately, one would like recipes for methods which are fast, generally applicable, do not sacrifice accuracy, and are easy to use.

ACKNOWLEDGMENTS

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