Efficiencies of Dynamic Monte Carlo Algorithms for Off-Lattice Particle Systems with a Single Impurity

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The efficiency of dynamic Monte Carlo algorithms for off lattice systems composed of particles is studied for the case of a single impurity particle. The theoretical efficiencies of the rejection-free method and of the Monte Carlo with Absorbing Markov Chains method are given. Simulation results are presented to confirm the theoretical efficiencies.

1. INTRODUCTION

One of the most difficult problems to solve in simulations of physical systems is the problem of disparate time scales. In materials, this problem is evident from looking at the timescales involved. The fundamental time for a move from quantum theory is on the order of that of an inverse phonon frequency, about 10^{-13} sec, which is faster than the clock cycle of a computer. The time scale to understand in aging of materials or for geological materials is on the order of decades to centuries to the age of the earth, and consequently much longer than the timescale that a program can be run on a computer. Even if parallel computers can help to bridge this gap, they can at most gain on the order of the number of processor elements. We are left with the realization that to bridge these disparate time scales will require advanced algorithms and physical understanding of both the system and its intrinsic dynamics.

In this paper we study the theoretical efficiency of advanced dynamic algorithms for particle systems, in particular rejection-free Monte Carlo (RFMC) methods and Monte Carlo with Absorbing Markov Chain (MCAMC) [1,2] methods for a system of particles

in d dimensions. For simplicity we concentrate on a system containing only one impurity particle. This work builds on recent papers examining the efficiencies of RFMC methods for particle systems without disorder [3,4]. It is important to realize that both the RFMC and MCAMC algorithms simulate exactly the same dynamic as that of the original dynamic Monte Carlo procedure, so no approximations are involved but rather the dynamic is programmed using a different algorithm.

2. MODELS AND METHODS

2.1. Particle Models and Dynamic Monte Carlo

In this paper we study a system of particles in dimension d, with $1 \leq d \leq 2$. In the systems studied the particles move in continuous space via a dynamic Monte Carlo procedure. The dynamic Monte Carlo procedure is [4,5]: 1) Randomly pick one of the N particles; 2) A new position for the particle is chosen by uniformly picking a point in a d-dimensional hyperspherical volume of radius r_{choose}; 3) Reject the new position if it is outside of the cage formed by its nearest neighbor particles; 4) Evaluate the energy difference $\Delta E_j = E_{\text{new},j} - E_{\text{old},j}$ between the old and the new positions of the chosen particle j; 5) Decide whether or not the trial move is to be accepted, for example using a Metropolis criteria where the new move is accepted if $\Delta E_j \leq 0$ and otherwise accepted with a probability of $\exp\left(-\Delta E_j/T\right)$ with T the absolute temperature and where Boltzmann's constant $k_{\text{B}} = 1$; and 6) If the trial move is accepted particle j is moved to the new position, otherwise it stays in its old position. One pass through these six steps advances the time by one Monte Carlo step (mcs).

We use a potential between two particles a distance r apart given by [4] $U(r) = 0$ if $r \geq r_{\text{c}}$ for a given cut-off radius and $U_p(r) = (1/r^p) - (1/r_{\text{c}}^p)$ for $r \leq r_{\text{c}}$. Between particles of the same type we choose $p = 12$, since this is the repulsive portion of Lennard-Jones interactions. Between a normal particle and an impurity particle we choose $p_i = 4$. We have only one impurity particle in the simulation, and start initially with the system in a ground state. We perform the simulation in a fixed system volume, and hence at a fixed density ρ. Periodic boundary conditions are used in all cases.

2.2. Rejection-Free Dynamic Monte Carlo (RFMC)

For the RFMC algorithm the system changes its configuration at every algorithmic step. The time between these updates for the discrete-time RFMC algorithm in units of mcs is given by [1,2,4,6] $t_{\text{wait}} = 1 + \lfloor \ln(\tilde{r})/\ln(\Lambda) \rfloor$ where $\lfloor \cdots \rfloor$ is the integer part, $\tilde{r}$ is a random number uniformly distributed in $(0, 1]$, and Λ is the probability for the system to stay in its current state in one dynamic Monte Carlo attempt. Consequently, $\Lambda = \frac{1}{N} \sum_{j=1}^{N} \lambda_j$ since the probability that particle j is picked to attempt a move is $1/N$ and the probability that the dynamic Monte Carlo trial move will be rejected given that particle j is picked is given by λ_j. The average waiting time is given by $\langle t_{\text{wait}} \rangle = 1/(1 - \Lambda)$, and since this is the efficiency it can be quite large for $\Lambda \sim 1$.

2.3. Monte Carlo with Absorbing Markov Chains (MCAMC)

The idea of the MCAMC algorithm is to identify some fast degrees of freedom (here a particle that changes position more rapidly than others), and have an algorithm that calculates the waiting time between the current system configuration and the next system

configuration where a particle other than a fast particle has moved. Then one step of the algorithm advances time by m mcs, the time until the first 'slow' move occurs for a particular configuration. In discrete space, with s states in the 'fast' subspace given by an $s \times s$ matrix (the transient matrix) $\mathbf{T}$, the waiting time is found by the solution of the equation [1,2] $\vec{v}_{\text{initial}}^{\text{T}} \mathbf{T}^m \vec{e} < \tilde{r} \leq \vec{v}_{\text{initial}}^{\text{T}} \mathbf{T}^{m+1} \vec{e}$ for a uniform random number $\tilde{r}$ in $(0, 1]$. Here $\vec{e}$ is a vector of length s with all elements equal to unity, and the initial vector has all zero elements except for unity in the initial state of the system. For $s=1$ this reduces to the RFMC algorithm [6].

Before spending time devising and programming an actual MCAMC algorithm for particle systems in continuous space, the efficiency of such an algorithm should be estimated. We can obtain this efficiency in a RFMC or dynamic Monte Carlo simulation by measuring the average time $\langle t_{\text{wait}} \rangle_{\text{MCAMC}}$ required for a 'slow' particle to move. This is the method used in the current paper.

3. THEORETICAL RESULTS

3.1. Rejection-Free Monte Carlo (RFMC)

We have previously predicted that for homogeneous d-dimensional particle systems the asymptotic average waiting time for the RFMC algorithm at high densities and low temperatures is [4] $\langle t_{\text{wait}} \rangle \propto r_{\text{choose}}^d \, T^{-\frac{d}{2}} \, \rho^{\frac{p+2}{2}}$. This result was found by noting that the integral for λ_j (which are all the same for a homogeneous system in the ground state) is strongly peaked for low temperatures and hence a Laplace saddle-point integration approximation can be used. This gives the RFMC algorithmic efficiency to be [4]

$$\langle t_{\text{wait}} \rangle \; = \; [\langle \exp\left(-\Delta E/T\right) \rangle]^{-1} \; \propto \; r_{\text{choose}}^d \, T^{-\frac{d}{2}} \, \sqrt{\left| \det \tilde{\mathbf{A}} \right|} \tag{1}$$

where the elements of the $d \times d$ matrix are [4] $\tilde{A}_{k\ell} = -T \, \frac{\partial^2}{\partial x_k \partial x_l} \ln\left[P(\vec{x})\right]\big|_{\vec{x}=\vec{x}_0}$ where $\vec{x}_0$ is the equilibrium position and $P(\vec{x})$ is the integrand in the integral that gives $1 - \lambda_j$. For our two-body interactions, which depend only on the distance a between the nearest-neighbor particles, $A_{\ell\ell} \sim a^{-p-2}$ and off diagonal elements are zero. Since for a homogeneous system in the ground state $\rho \propto a^{-d}$ one has that $\left|\det\tilde{\mathbf{A}}\right|^{\frac{1}{2}} \sim \left[a^{-p-2}\right]^{\frac{d}{2}} \sim \rho^{\frac{p+2}{2}}$.

For our case with a single impurity the same analysis holds, but now rather than having to perform one integral to obtain all λ_j, we have to perform a slightly different integral for each particle j. Each integral for a λ_j may have a different local environment for the system in the ground state and the impurity particle has a different two-body interaction than that between non-impurity particles. Since every λ_j will have the temperature enter into the integral the same way, we obtain the prediction that asymptotically at low temperatures the efficiency will be given by $\langle t_{\text{wait}} \rangle \propto T^{-\frac{d}{2}}$ even for the case of a system with disorder. For the disordered case the RFMC algorithmic efficiency is

$$\langle t_{\text{wait}} \rangle \; \propto \; r_{\text{choose}}^d \, T^{-\frac{d}{2}} \, N \, \left[\sum_{j=1}^N \left(\det\left| \tilde{\mathbf{A}}_j \right| \right)^{-\frac{1}{2}} \right]^{-1} . \tag{2}$$

3.2. Monte Carlo with Absorbing Markov Chains (MCAMC)

Even for a single impurity particle the efficiency of MCAMC is complicated. Under certain assumptions [7] the ratio of the efficiency of the MCAMC algorithm to that of the

RFMC algorithm can be shown to be

$$R_{\text{MCAMC}} = \frac{\langle t_{\text{wait}} \rangle_{\text{MCAMC}}}{\langle t_{\text{wait}} \rangle} \propto 1 + \frac{1}{N} \sqrt{\frac{\det \left| \tilde{\mathbf{A}} \right|}{\det \left| \tilde{\mathbf{A}}_{\text{impurity}} \right|}}. \tag{3}$$

Thus the MCAMC efficiency compared to the RFMC efficiency is independent of the temperature. This is in marked contrast to the case of discrete state systems [1].

4. SIMULATION RESULTS

To perform the simulations we found that relaxation to the ground state from an initial state was very slow. Therefore we performed a quenching procedure by cooling the system slowly from high temperature. Once the system was quenched we measured the efficiency of the RFMC method by measuring $\langle t_{\text{wait}} \rangle$ and measured the efficiency of the MCAMC method by measuring $\langle t_{\text{wait}} \rangle_{\text{MCAMC}}$. We studied one- and two-dimensional systems, and 10^6 independent samples are averaged at each temperature.

Figure 1 shows the results for a single impurity particle with p_i=4, p=12, N=100, and ρ=2. For d=1 with a single impurity all nearest-neighbor distances a values are equal except for two. The measured values for the nearest neighbor distances at T=1 were $a_{\text{impurity}} = 0.137(1)$ and $a = 0.507(2)$. From these values the efficiency ratio R_{MCAMC} from Eq. (3) is predicted to be almost unity. For our parameter values the MCAMC algorithm is not much more efficient than is the RFMC algorithm. The behavior of the efficiency on the temperature is $\sim T^{-\frac{d}{2}}$ as predicted, and is shown in Fig. 1. We find that both the RFMC and MCAMC efficiencies for the single impurity case at this density are comparable to that for the homogeneous case. This is because of compensation effects that are present in d=1 but absent in higher dimensions [7].

For d=2 simulation results are shown in Fig. (2) for the efficiencies versus the temperature. The simulations were performed with N=100 and ρ=4.0. Again it is seen that asymptotically the efficiency goes as $T^{-\frac{d}{2}}$. Furthermore, the asymptotic efficiency ratio R_{MCAMC} is approximately independent of T. Unlike the d=1 case, the MCAMC algorithm significantly outperforms the RFMC for the inhomogeneous lattice. This is because due to 'cage' effects $a \approx a_{\text{impurity}}$. From Eq. (3) for our parameters the predicted ratio is $R_{\text{MCAMC}} \approx 21$, and the ratio of the intercepts of the two lines in Fig. (2) is about 19. For ρ=8 the predicted value would be $R_{\text{MCAMC}} \approx 320$.

5. CONCLUSIONS AND DISCUSSION

We have studied the efficiencies of the rejection-free Monte Carlo (RFMC) and the Monte Carlo with Absorbing Markov Chains (MCAMC) algorithms for particle systems with an impurity particle. We have compared our theoretical predictions of the asymptotic efficiency to that from simulations in d=1 and d=2. We find that even with impurities both the RFMC and MCAMC algorithms have efficiencies that grow as the temperature is lowered as $T^{-\frac{d}{2}}$. We also find that the ratio of the MCAMC to the RFMC asymptotic efficiencies is approximately independent of T. For d=1 there is very little gain in efficiency by having the impurity particle put into a MCAMC algorithm, as opposed to using the simpler RFMC algorithm. In contrast, in d=2, due to the 'cage' effect, the MCAMC
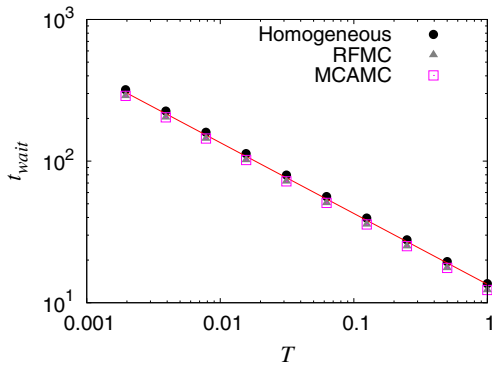
Figure 1. The efficiencies of RFMC and MCAMC for $d=1$ is shown as a function of temperature T. This is for $\rho = 2.0$ and $N = 100$. The solid line has slope $-\frac{1}{2}$. The disordered system has efficiencies of both the RFMC and MCAMC algorithms that are almost the same as those for RFMC for the homogeneous system. The units for t_{wait} are mcs per particle.
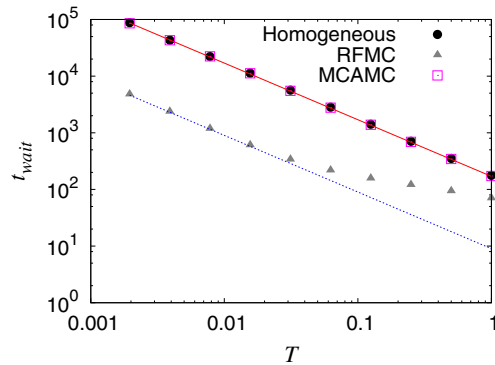
Figure 2. The efficiencies of RFMC and MCAMC for $d=2$ is shown as a function of temperature T. This is for $\rho = 4.0$ and $N = 100$. The lines have slope -1. The efficiencies of the MCAMC algorithm for the disordered system are almost the same as those for the RFMC for the homogeneous system. The units for t_{wait} are mcs per particle.

algorithm can be orders of magnitude more efficient than the RFMC algorithm. Further studies of the efficiencies in $d=3$ and for *ab initio* dynamic Monte Carlo [8] would be of interest. Actual implementations of the MCAMC algorithm requires further development, but this study shows that the algorithm could be a big improvement over the RFMC algorithm for $d \geq 2$.

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