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Differences between fixed time step and kinetic Monte Carlo methods for biased diffusion

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ABSTRACT

We consider biased diffusion in a one-dimensional lattice and compare results obtained with fixed time step and kinetic Monte Carlo methods. Spurious dispersion and particle position correlation appear with the fixed time step Monte Carlo approach. The mentioned correlation increases with time. We demonstrate that the correct results, that correspond to a time step that tends to zero, are obtained using the kinetic Monte Carlo method. The conclusions also apply to biased diffusion in two or more dimensions and to random deposition.

1. Introduction

A crystalline surface, over which adsorbed particles diffuse, is usually modeled by a periodic potential as shown in Fig. 1(a). A particle in a potential well can jump to the neighboring wells. The transition rate is given by $v\exp[-E/(k_BT)]$, where v is a rate constant (usually of the order of 10^{12} s^{-1} [1]), E is the energy barrier, and T is temperature. If the particle is charged and an electric field is applied, the potential is modified as in Fig. 1(b). The transition rate is now bigger to the right than to the left. The difference between the two transition rates can be controlled with the temperature and the electric field intensity. The problem of biased diffusion, in different substrata, is analyzed in, for example, [2–6].

The numerical simulation of this simple physical system with a fixed time step Monte Carlo (MC) method introduces distortions and correlations that are not present in the kinetic MC method (for a review of the methods, see [7,8]). We will consider a system of N non-interacting particles with asymmetric or biased diffusion. We will obtain, analytically and numerically, the moments of position up to order 4 and the correlation between the positions of two particles for fixed time step and kinetic MC. The results will be compared with the ideal result that is obtained when the time step is infinitesimally small ($\Delta t \to 0$). Some simple examples where the mentioned distortions are relevant will be discussed.

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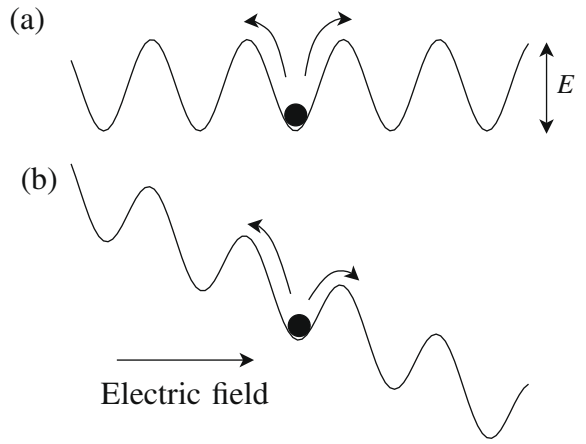

Fig. 1. (a) Adsorbed particle in a surface potential with energy barrier E. (b) Same potential for a charged particle with an applied electric field.

2. The methods

We consider a one-dimensional lattice with N non-interacting particles that can jump to the right or to the left with transition probabilities per unit time P and Q, respectively. In one MC step there are $2N$ possible processes, and one of these processes is chosen with the corresponding probability.

The difference between the fixed time step MC and kinetic MC methods is how time is increased after each step. For fixed time step MC, a small enough time step Δt is chosen so that the sum of the probabilities of all processes remains smaller or equal to 1, i.e. $N(P+Q)\Delta t \leqslant 1$. In each MC step, the probability that a particle jumps to the right or to the left is $p = P\Delta t$ or $q = Q\Delta t$, respectively; one of the $2N$ possible processes is chosen according to these probabilities and time is increased by Δt. Note that the probabilities p and q correspond to processes that can be split in two parts: one particle is chosen with uniform probability $1/N$ and this particle jumps to the left or to the right with probabilities Np and Nq.

In the kinetic MC method, the time increment is a stochastic variable that represents a Poisson process, with an exponential distribution that is given by $R\exp(-R\Delta t)$, where $R = N(P+Q)$ is the sum of the transition rates of all processes. To numerically accomplish this distribution, in each step the time increment is calculated with $\Delta t = -\ln(u)/R$, where u is a random number with a uniform distribution in the interval $(0,1]$ [9]. In each MC step, the probability that one particle jumps to the right or to the left is $p = P/R$ or $q = Q/R$, respectively. The sum of the probabilities of all processes is equal to one, so that in each MC step one possible process is always performed.

In both cases, the size of the jump (to the right or to the left) is Δx.

3. Continuous time random walk

The one-dimensional system with N non-interacting particles can be visualized as one particle that performs a random walk in an N-dimensional system. For this system, the Continuous Time Random Walk (CTRW) method [10,11] can be applied to obtain the Laplace and Fourier transform of the probability density $n(\mathbf{x},t)$, that gives the probability to find the particle in position $\mathbf{x} = (x_1, x_2, \ldots, x_N)$ at time t. The method is a generalization of the random walk, in which time appears as a continuous variable. It can be applied to both, fixed time step MC and kinetic MC.

The complete solution of the CTRW problem is given by the Montroll–Weiss equation [12] for the characteristic function, i.e. the Fourier and Laplace transform of $n(\mathbf{x},t)$:

$$\hat{\tilde{n}}(\mathbf{k},s) = \frac{1-\hat{\psi}(s)}{s}\frac{1}{1-\hat{\psi}(s)\tilde{f}(\mathbf{k})}, \tag{1}$$

where $\hat{\psi}(s)$ is the Laplace transform of the waiting time distribution $\psi(t)$ (the probability density of a time increment of duration t between two successive steps); and $\tilde{f}(\mathbf{k})$, called the structure function of the random walk, is the Fourier transform of $f(\mathbf{x})$, the probability density of a jump of size $\mathbf{x}$.

The main difference between fixed time step and kinetic MC appears in the waiting time distribution. For fixed time step MC, this distribution is a Dirac delta,

$$\psi_F(t) = \delta(t-\Delta t), \quad \hat{\psi}_F(s) = e^{-s\Delta t}, \tag{2}$$

while for kinetic MC, the distribution is exponential:

$$\psi_K(t) = R\exp(-Rt), \quad \hat{\psi}_K(s) = R/(s+R). \tag{3}$$

We use subindices F and K to refer to fixed time step or kinetic MC, respectively.

It is convenient to define a density of transition rates of jumps of size $\mathbf{x}$ as $g(\mathbf{x}) = \sum_{i=1}^{M} P_i \delta(\mathbf{x} - \Delta\mathbf{x}_i)$, where P_i are the transition rates of the M possible processes and $\Delta\mathbf{x}_i$ are the corresponding jump sizes. (We are considering a finite number of processes and a discrete density $g(\mathbf{x})$.) The density $g(\mathbf{x})$ is not normalized and its integral is equal to the sum of transition rates $R = \sum_{i=1}^{M} P_i$. The way in which $g(\mathbf{x})$ is normalized to obtain the probability density $f(\mathbf{x})$ is different for fixed time step or kinetic MC. For fixed time step MC, the time step Δt must be small enough such that $R\Delta t \leqslant 1$, and we have

$$f_F(\mathbf{x}) = g(\mathbf{x})\,\Delta t + (1 - R\Delta t)\delta(\mathbf{x}), \tag{4}$$

where the last term represents the case in which non of the M possible processes is performed. On the other hand, in one step of the kinetic MC method, one of the M possible processes is always performed and the normalization is

$$f_K(\mathbf{x}) = g(\mathbf{x})/R. \tag{5}$$

Eqs. (4) and (5) coincide when the time step takes its maximum possible value: $\Delta t = 1/R$. In particular, for our case of N particles with biased diffusion, we have

$$\begin{aligned} g(\mathbf{x}) &= \sum_{i=1}^{N} [P\delta(x_i - \Delta x) + Q\delta(x_i + \Delta x)] \prod_{j \neq i} \delta(x_j), \\ \tilde{g}(\mathbf{k}) &= \sum_{i=1}^{N} [Pe^{-ik_i\Delta x} + Qe^{ik_i\Delta x}]. \end{aligned} \tag{6}$$

Introducing Eqs. (2) and (4) for fixed time step MC, or Eqs. (3) and (5) for kinetic MC, in Eq. (1), we obtain:

$$\hat{\tilde{n}}_F(\mathbf{k}, s) = \frac{1}{s + [R - \tilde{g}(\mathbf{k})]\frac{s\Delta t}{\exp(s\Delta t) - 1}}, \tag{7}$$

$$\hat{\tilde{n}}_K(\mathbf{k}, s) = \frac{1}{s + R - \tilde{g}(\mathbf{k})}. \tag{8}$$

It is assumed that the fixed time step MC exactly reproduces the behavior of the physical system only in the limit $\Delta t \to 0$. By comparison of Eqs. (7) and (8) we can demonstrate that this limit corresponds to the kinetic MC:

$$\lim_{\Delta t \to 0} \hat{\tilde{n}}_F(\mathbf{k}, s) = \hat{\tilde{n}}_K(\mathbf{k}, s). \tag{9}$$

Therefore, for this system of non-interacting particles, the kinetic MC method generates the exact characteristic function. All moments and correlations can be obtained by differentiation of the characteristic function,

$$\langle x_1 x_2 \cdots x_m \rangle = i^m \left. \frac{\partial^m \tilde{n}(\mathbf{k}, t)}{\partial k_1 \partial k_2 \cdots \partial k_m} \right|_{\mathbf{k}=0}. \tag{10}$$

(The sign of the imaginary number i in (10) depends on the convention of signs of the Fourier transform; we are using a minus sign in the exponential of the forward transform, but others use the opposite convention as, for example, [10].)

Another method to obtain the moments and correlations is presented in Appendix A. This method has the advantage that does not use Laplace transform (the inverse Laplace transform of $\hat{\tilde{n}}_F(\mathbf{k}, s)$ is not straightforward to obtain).

For simplicity, we will consider that the jump step is $\Delta x = 1$.

4. Moments

The mean value of position of one of the N particles (that starts from position $x = 0$ for $t = 0$) is the same for both methods: $\langle x \rangle_F = \langle x \rangle_K = \bar{x} = (P - Q)\,t$. But, for other moments, the fixed time step MC method introduces errors proportional to Δt. The centered moments, for fixed time step or kinetic MC, are

$$\Delta x^m|_{F,K} = \langle (x - \bar{x})^m \rangle_{F,K}. \tag{11}$$

For order 2 we have

$$\begin{aligned} \Delta x^2|_K &= \sigma^2 = (P + Q)t, \\ \Delta x^2|_F &= (P + Q)t - (P - Q)^2 t\Delta t, \end{aligned} \tag{12}$$

where σ is the dispersion of the exact result given by the kinetic MC method. The difference between both moments increases linearly with time, but the moment magnitude also increases with time. It is convenient to define a normalized difference between moments as follows:

$$D_m = \frac{\Delta x^m|_F - \Delta x^m|_K}{\sigma^m}. \tag{13}$$

This definition is equivalent to a scaling of the space coordinate so that the probability distribution of particles has a constant width. We get, for the moment difference up to order 4,

$$D_2 = -\frac{(P-Q)^2}{P+Q}\Delta t, \tag{14}$$

$$D_3 = \frac{2(P-Q)^3\Delta t - 3(P^2-Q^2)}{(P+Q)^{3/2}\sqrt{t}}\Delta t, \tag{15}$$

$$D_4 = \{3(t-2\Delta t)[(P-Q)^4\Delta t - 2(P-Q)^2(P+Q)] + 2PQ - 7Q^2 - 7P^2\}[(P+Q)^2 t]^{-1}\Delta t. \tag{16}$$

In the limit $t \to \infty$, the difference between odd moments tends to 0, since the same moments tend to 0 because the probability distributions tend to a symmetric Gaussian. But the widths of the Gaussians are different, and a difference between even moments remain even for $t \to \infty$. For the difference of order 4 moments we have $\lim_{t\to\infty} D_4 = 3[(P-Q)^4\Delta t - 2(P-Q)^2(P+Q)](P+Q)^{-2}\Delta t$.

Fig. 2 shows D_2, D_3 and D_4 against time for diffusion of one particle with $P = 3/8, Q = 1/8$ and $\Delta t = 1$, for fixed time step and kinetic MC methods. In MC simulations of diffusion processes, a time interval equal to $1/(P+Q)$ is usually considered as the time needed for all particles to have, on average, one chance to jump. In experiments, this time varies in a broad range that depends on the values of the temperature and the energy barrier. If N particles are present in the system, the time increment after one MC step (in which only one particle can jump) is taken as $\Delta t = 1/[N(P+Q)]$, so that for a time increment equal to $1/(P+Q)$ all particles had a chance to jump. This choice is arbitrary in the sense that even for only one particle we can chose $\Delta t = 1/[N(P+Q)]$ with N large, but the simulation would be quite inefficient since during most of the computing time the particle would stay at its site. In our simulations, we considered diffusion of only one particle with Δt large in order to magnify the distortions introduced by the fixed time step MC, but for N particles the same distortions are still present, the only difference is that the vertical axis of Fig. 2 must be approximately scaled by a factor $1/N$.

In the case of non-biased or symmetric diffusion, $P = Q$, the fixed time step MC performs better than for biased diffusion, since $D_2^s = D_3^s = 0$. There is a difference between moments of order 4: $D_4^s = -3\Delta t/t$, but it tends to 0 for $t \to \infty$.

The errors introduced in the moments by the fixed time step MC can be corrected with a change of the transition rates. We define the transition rates P' and Q' such that, using fixed time step MC, they reproduce the first two moments that are obtained with kinetic MC with transition rates P and Q, i.e.:

$$(P-Q)t = (P'-Q')t, \tag{17}$$

$$(P+Q)t = (P'+Q')t - (P'-Q')^2 t\Delta t. \tag{18}$$

Solving Eqs. (17) and (18) we get

$$\begin{aligned} P' &= P + (P-Q)^2\Delta t/2, \\ Q' &= Q + (P-Q)^2\Delta t/2. \end{aligned} \tag{19}$$

With this modification of the transition rates for fixed time step MC, we have $D_2' = 0$ and obtain

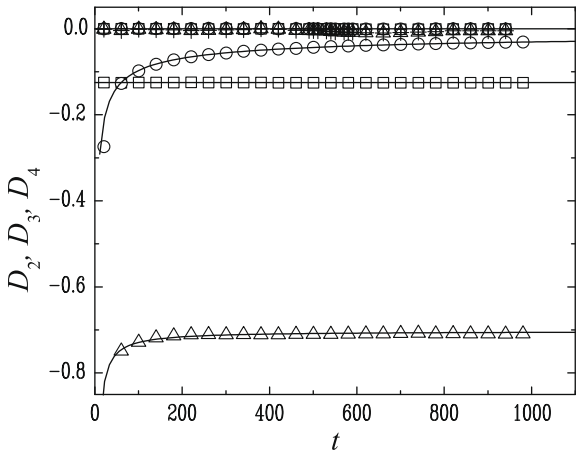

Fig. 2. Numerical results of $D_2(\square), D_3(\circ)$ and $D_4(\triangle)$ against time for $P = 3/8, Q = 1/8$ and $N = 1$. The curves represent the analytical results of Eqs. (14)–(16). We have chosen a time scale such that $\Delta t = 1, p = P$ and $q = Q$ for fixed time step MC. Number of samples: 10^7. The numerical results represented by symbols $\boxplus, \oplus$ and $\triangleq$, around zero, correspond to D_2, D_3 and D_4 with the fixed time step moments replaced by the analytical result obtained for the limit $\Delta t \to 0$; this is a confirmation of the correct behaviour of the kinetic MC results.

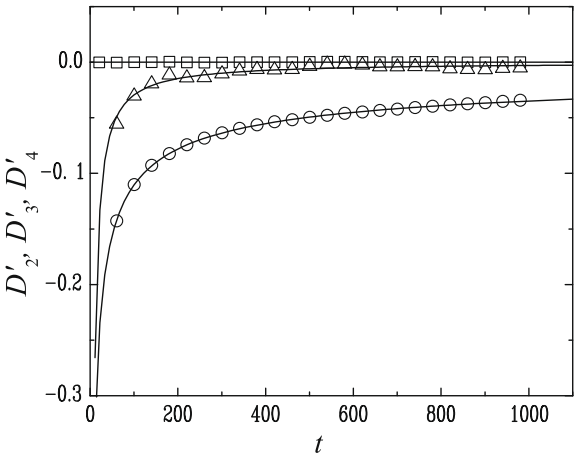

Fig. 3. $D'_2(\square), D'_3(\circ)$ and $D'_4(\triangle)$ against time for $P = 3/8, Q = 1/8, N = 1$ and $\Delta t = 1$, that correspond to the modified transition rates $P' = 13/32$ and $Q' = 5/32$. Dots are numerical results and the curves correspond to Eqs. (20) and (21). Number of samples: 10^7.

$$D'_3 = -\frac{3(P^2 - Q^2) + (P - Q)^3 \Delta t}{(P + Q)^{3/2}\sqrt{t}} \Delta t, \tag{20}$$

$$D'_4 = 3\frac{(P - Q)^4 \Delta t^2 + 2(P - Q)^2 (P + Q)\Delta t - 2(P^2 + Q^2)}{(P + Q)^2 t} \Delta t. \tag{21}$$

As expected for all odd moments, D'_3 tends to 0 when $t \to \infty$. The main difference with respect to Eqs. (15) and (16) is that now D'_4 tends to 0 when $t \to \infty$. Therefore, for t large, the first four moments approach the correct value using fixed time step MC with transition rates modified according to (19), and it is expected that the same happens for higher order moments since the probability distribution tends to a Gaussian that is completely determined by its mean value and dispersion. Fig. 3 shows numerical and analytical results for D'_2, D'_3 and D'_4 with the modified transition rates.

Let us stress that there is another problem that the modification of the transition rates can not avoid: the correlations.

5. Correlations

The correlation between fluctuations of position around the mean value of two particles is $C_{K,F} = \langle (x_1 - \bar{x}_1)(x_2 - \bar{x}_2) \rangle_{K,F}$. The analytical results for kinetic and fixed time step MC are

$$\begin{aligned} C_K &= 0, \\ C_F &= -(P - Q)^2 t \Delta t. \end{aligned} \tag{22}$$

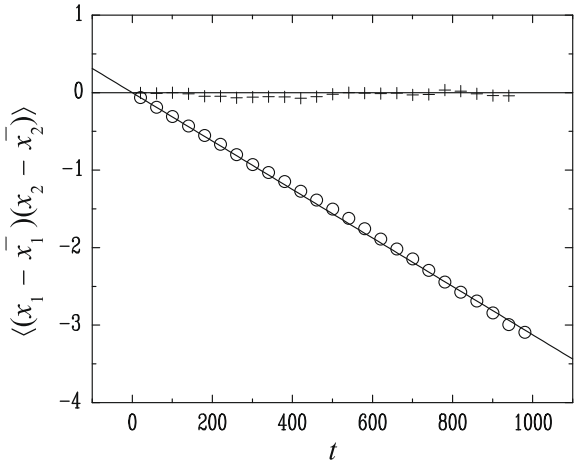

Fig. 4. Correlation between position of two particles for fixed time step MC (circles) and for kinetic MC (crosses) against time (numerical and analytical) for $P = 3/8$ y $Q = 1/8, N = 20$ and $\Delta t = 1/20$. Number of samples: 10^5.

The fixed time step MC method again introduces a distortion, now in the form of a spurious correlation. As happened with the second order moment, the distortion is proportional to Δt and is different from zero for biased diffusion, i.e. $P \neq Q$.

The correlation increases with time, therefore, even for small values of Δt, for a long enough time there will be a large value of the correlation.

Fig. 4 shows numerical and analytical results of C_F and C_K against time for a one-dimensional system with 20 particles.

Note that the modified transition rates, P' and Q', introduced in the previous section, do not change the value of the correlation C_F, since it depends on the difference $P - Q$ and, according to Eq. (17), this difference does not change.

Let us now present an illustrative example where the correlations in the fixed time step MC method appear in a very simple way. We consider a system of $N = 2$ particles, particle 1 and particle 2, with $P > 0$ and $Q = 0$. The particles start at $t = 0$ from position $x_1 = x_2 = 0$. In the fixed time step MC simulation, we chose Δt such that $p_1 = p_2 = P\Delta t = 1/2 (q_1 = q_2 = 0)$. Then, at each MC step, always one particle jumps to the right. After $n = 2k (k = 1, 2, 3, \ldots)$ MC steps, the average position of both particles is $\bar{x}_1 = \bar{x}_2 = k$, and $t = 2k\Delta t$. Let us now consider only one sample composed by these two particles. Let $m_1 (m_2)$ be the number of jumps performed by particle 1 (particle 2). Then, we have $m_1 + m_2 = n, x_1 = m_1$ and $x_2 = m_2$, at time t. There are three possibilities: (a) $m_1 > k$ and $m_2 < k$, (b) $m_1 < k$ and $m_2 > k$, and (c) $m_1 = m_2 = k$. For the first two cases, $C_{12} = (x_1 - \bar{x}_1)(x_2 - \bar{x}_2)$ is negative. Only for case (c) we have $C_{12} = 0$. The correlation C_F is the mean value of C_{12}, then obviously $C_F < 0$. The same result can be obtained after $n = 2k + 1 (k = 0, 1, 2 \ldots)$ MC steps.

6. Examples

In this section, we present some simple models of physical systems where the choice of the fixed time step MC method introduces errors that are not present in the kinetic MC method.

The most simple example is Brownian motion in one dimension with a drift velocity v_d. The drift velocity is correctly obtained by fixed time step or kinetic MC, and its value is $v_d = (P - Q)$ (we are using $\Delta x = 1$). The diffusion coefficient is related to the second order moment by $\Delta x^2|_{F,K} = 2D_{F,K}\, t$. According to Eq. (12), there is a difference between the diffusion coefficients obtained with both MC methods: $D_F - D_K = -(P - Q)^2 \Delta t/2$. Fig. 5 shows the one particle probability density against position for time $t = 500 (\Delta t = 1, P = 0.9, Q = 0.1)$ for kinetic and fixed time step MC. The difference between the widths of the Gaussian curves is directly related to the error introduced in the diffusion coefficient by the fixed time step MC method.

For one particle in a two dimensional system, the spurious correlations of the fixed time step MC also produces distortions in the particle probability density. Fig. 6 shows the particle density in grey scale for a sequence of times and for fixed time step MC. The x and y components of the movement have the same transition rates $(P_x = P_y = 0.7, Q_x = Q_y = 0.05)$ so, in average, the particle moves along the diagonal $y = x$. Due to the negative correlation between fluctuations of coordinates x and y [see Eq. (22)], the particle density is elongated along a direction perpendicular to the diagonal. This distortion disappears when the kinetic MC method is used, as shown in Fig. 7.

Another example is a model of surface growth known as random deposition [13]. The model consists in a discrete one-dimensional lattice in which, at each step of the dynamics, one site is randomly chosen and one particle falls over it. Each time a particle falls over a site, its height $h(i, t)$ is increased by one. The original definition of the model considers that, after each step, time is increased by a fixed time increment $\Delta t = 1/L$, where L is the number of sites. The model is equivalent to biased diffusion with $P = 1$ and $Q = 0$. The width of the interface w^2 is given by the centered second order moment, $w^2 = t(1 - 1/L)$. The result shows a dependence of the width of the interface on the size of the system, that has no physical

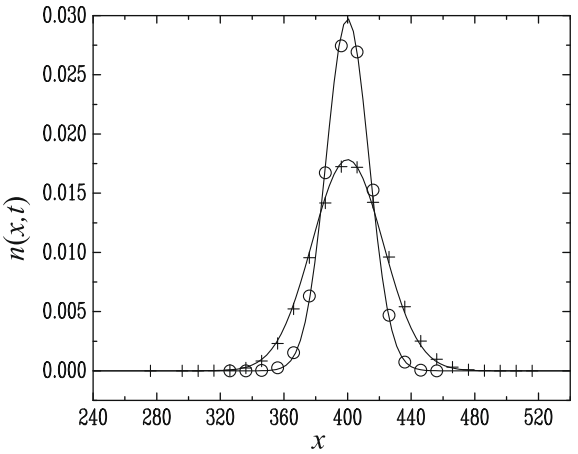

Fig. 5. Particle probability density $n(x, t)$ against position x, for $P = 0.9, Q = 0.1, N = 1, \Delta t = 1$ and for time $t = 500$. The points correspond to the numerical results (∘: fixed time step MC, +: kinetic Monte Carlo), and the curves are Gaussian functions with mean value $(P - Q)\, t$ and dispersions given by Eqs. (12). Number of samples: 10^6.

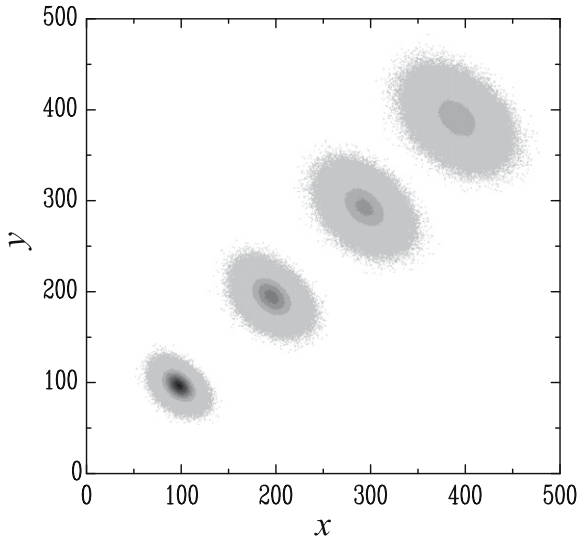

Fig. 6. Probability density of one particle in a two dimensional system for fixed time step MC. The sequence, from left-bottom to right-top, correspond to times $t = 150, 300, 450$ and 600. The transition rates of the movement in the x and y coordinates are $P_x = P_y = 0.7$ and $Q_x = Q_y = 0.05$. The density appears elongated along the direction perpendicular to the diagonal $y = x$. $\Delta t = 1/2$ and the number of samples: 10^6. (White = 0, black = 0.0022).

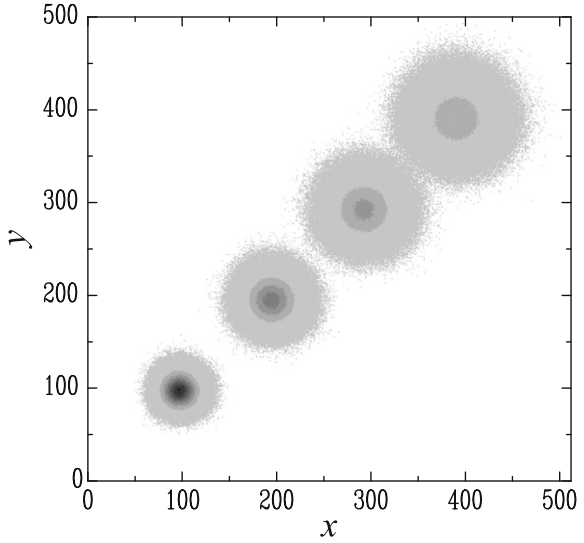

Fig. 7. Probability density of a particle in a two dimensional system for kinetic MC. The distortions of the fixed time stem MC method are not present now. Parameters as in Fig. 6. (White = 0, black = 0.0014).

meaning. On the other hand, the definition of the model is intended to produce an *uncorrelated* interface, since each site behaves independently of the rest of the system. But this is not the case, since a negative correlation, given by $C_F = -t/L$, appears between the heights of the columns in any pair of sites. Again, all these distortions disappear using the stochastic time step of the kinetic MC method.

7. Conclusions

We analyzed biased diffusion of N non-interacting particles in a one dimensional lattice simulated using the methods of fixed time step MC and kinetic MC. The fixed time step MC method introduces distortions, in the values of moments and correlations, that, in the limit of small time step Δt, are proportional to Δt. Let us note that all terms proportional to Δt in the moments or correlations represent an artifact of the method, since Δt is an arbitrary parameter that has no counterpart in the physical system that is modeled, and the correct result is only obtained in the ideal limit $\Delta t \to 0$.

We have demonstrated, using the Montroll–Weiss equation for Continuous Time Random Walk, that the evolution of the particle probability density, $n(\mathbf{x},t)$, with kinetic MC is equivalent to the evolution with fixed time step MC in the limit $\Delta t \to 0$. This means that kinetic MC reproduces the correct dynamics that for fixed time step MC would require an infinite computational time.

The errors that the fixed time step MC (with finite Δt) introduces in moments can be eliminated, in the limit $t \to \infty$, with a simple modification of the transition rates. The new transition rates, P' and Q', are defined so that they produce the same (undistorted) mean value and the correct second order moment. Since for $t \to \infty$ the particle density tends to a Gaussian completely determined by its mean value and dispersion, the errors in higher order moments vanish for large times.

Nevertheless, distortions in correlations can not be avoided with the modified transition rates. Since we are considering non-interacting particles, correlations should be zero, but the fixed time step MC introduces a spurious non zero correlation between fluctuations of position of two particles. Correlations calculated with kinetic MC are zero.

It has to be noted that, in most of the previous works on biased diffusion simulated with fixed time step MC, the errors of the method are absorbed by the statistical error since a small value of Δt and a large number of particles are, in general, used.

In conclusion, simulation of biased diffusion with fixed time step MC introduces distortions in moments and correlations that can produce errors in, for example, the calculation of diffusion coefficients or particle density profiles. The distortions are not present in the kinetic MC method with stochastic time step.

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Appendix A

In this appendix, we present an alternative method for the calculation of correlations and moments of N non-interacting particles with biased diffusion in a one-dimensional lattice.

The vector $\mathbf{x} = (x_1, \ldots, x_N)$ represents the position of the N particles. At $t = 0$ we have $\mathbf{x} = \mathbf{0}$, so, the initial probability density is $n(\mathbf{x}, 0) = \delta(\mathbf{x})$. We define the auxiliary function $F_{F,K}(\mathbf{z}, t)$ as follows:

$$F_{F,K}(\mathbf{z},t) = \int d\mathbf{x} n_{F,K}(\mathbf{x},t) z_1^{x_1} z_2^{x_2} \cdots z_N^{x_N}, \tag{23}$$

where, as before, subindices F and K stand for fixed time step or kinetic MC. It can be shown that all moments and correlations can be obtained via the equation

$$\langle x_1^{j_1} x_2^{j_2} \cdots x_N^{j_N} \rangle_{F,K} = \prod_{i=1}^{N} \left(z_i \frac{\partial}{\partial z_i} \right)^{j_i} F_{F,K}(\mathbf{z},t) \Big|_{\mathbf{z}=(1,\ldots,1)}. \tag{24}$$

We consider first the case of fixed time step MC. In order to obtain an expression for $F_F(\mathbf{z}, t)$, we use the Master equation for $n_F(\mathbf{x}, t)$ (for simplicity, we will omit subindex F in n) considering fixed time increments of size Δt:

$$n(\mathbf{x}, t + \Delta t) = (1 - R\Delta t)\, n(\mathbf{x}, t) + P \Delta t \sum_{i=1}^{N} n(x_1, \ldots x_i - \Delta x, \ldots x_N; t) + Q \Delta t \sum_{i=1}^{N} n(x_1, \ldots x_i + \Delta x, \ldots x_N; t). \tag{25}$$

Introducing Eq. (25) in Eq. (23), we obtain

$$\begin{aligned} F_F(\mathbf{z}, t + \Delta t) = {} & (1 - R\Delta t) \int d\mathbf{x}\, n(\mathbf{x},t) z_1^{x_1} \cdots z_N^{x_N} + P \Delta t \sum_{i=1}^{N} \int d\mathbf{x}\, n(\mathbf{x},t) z_1^{x_1} \cdots z_i^{x_i + \Delta x} \cdots z_N^{x_N} \\ & + Q \Delta t \sum_{i=1}^{N} \int d\mathbf{x}\, n(\mathbf{x},t) z_1^{x_1} \cdots z_i^{x_i - \Delta x} \cdots z_N^{x_N}, \end{aligned} \tag{26}$$

where a change of the integration variable was performed in the last two terms. The following recursion formula is obtained:

$$F_F(\mathbf{z}, t + \Delta t) = \left(1 - R\Delta t + P \Delta t \sum_{i=1}^{N} z_i^{\Delta x} + Q \Delta t \sum_{i=1}^{N} z_i^{-\Delta x} \right) F(\mathbf{z}, t). \tag{27}$$

This formula and the initial condition $F_F(\mathbf{z}, 0) = 1$, allow us to obtain the following expression for F_F:

$$F_F(\mathbf{z}, t) = \left(1 - R\Delta t + P \Delta t \sum_{i=1}^{N} z_i + Q \Delta t \sum_{i=1}^{N} z_i^{-1} \right)^{t/\Delta t}, \tag{28}$$

where $t/\Delta t$ is the number of fixed size time steps and, for simplicity, we have used $\Delta x = 1$.

The auxiliary function for kinetic MC can be obtained knowing that it is equivalent to fixed time step MC in the limit $\Delta t \to 0$, as was demonstrated in Eq. (9). In this limit, we get

$$F_K(\mathbf{z}, t) = \exp\left[\left(-R + P\sum_{i=1}^{N} z_i + Q\sum_{i=1}^{N} z_i^{-1}\right)t\right]. \tag{29}$$

Using the auxiliary functions of Eqs. (28) and (29), we can obtain all the analytical results of moments and correlations shown in the previous pages.

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