Generalized event-chain Monte Carlo: Constructing rejection-free global-balance algorithms from infinitesimal steps

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In this article, we present an event-driven algorithm that generalizes the recent hard-sphere event-chain Monte Carlo method without introducing discretizations in time or in space. A factorization of the Metropolis filter and the concept of infinitesimal Monte Carlo moves are used to design a rejection-free Markov-chain Monte Carlo algorithm for particle systems with arbitrary pairwise interactions. The algorithm breaks detailed balance, but satisfies maximal global balance and performs better than the classic, local Metropolis algorithm in large systems. The new algorithm generates a continuum of samples of the stationary probability density. This allows us to compute the pressure and stress tensor as a byproduct of the simulation without any additional computations.

Keywords: Monte Carlo algorithms; particle systems; event-chain Monte Carlo; Markov chain lifting; global balance

I. INTRODUCTION

Markov-chain Monte Carlo (MCMC) methods in statistical physics have progressed far from the original local-move, detailed-balance Metropolis algorithm[3]. On the one hand, intricate non-local cluster moves have met with great success in lattice models[4,5]. To a lesser extent, continuum systems of hard spheres have in recent years also benefitted from non-local moves[6–9], building on earlier work[10–12]. On the other hand, extensions of the classic detailed balance condition have allowed to construct Markov chains that converge faster. These algorithms introduce persistence between subsequent moves and reduce the diffusive nature of the Markov chain on small and intermediate length and time scales. Notable examples are guided random walks[13], hybrid Monte Carlo[14,15] and overrelaxation[16]. The Markov chain lifting framework[17–19] unifies these concepts by augmenting the physical configuration space with auxiliary variables that resemble the momentum in Newtonian time evolution and in molecular dynamics (MD)[20]. Lifted Markov chains have already been applied to spin models[23,24], but not to continuum systems.

The present article draws on the above lines of research. As a main theoretical result, we introduce a factorized version of the Metropolis filter (acceptance rule) that is well suited for the simulation of N-particle systems with pair-potential interactions. Combined with the concept of infinitesimal Monte Carlo moves, this filter allows us to construct a rejection-free event-chain Monte Carlo (ECMC) algorithm that breaks detailed balance yet satisfies maximal global balance. This algorithm builds upon a recent insightful hybrid Monte Carlo scheme[15]. By virtue of infinitesimal displacements of particles, our algorithm produces a continuum of configurations that all sample the equilibrium distribution. Samples are obtained efficiently using an event-driven algorithm. For hard spheres, the events correspond to hard-sphere collisions, and the new algorithm reduces to the hard-sphere event-chain algorithm[6]. For general pair interactions, our algorithm replaces the hard-sphere collisions by pairwise collisions, whose collision distance is resampled after each event from to the pair potential. Finally, the continuum of ECMC samples permits to directly compute the pressure and the stress tensor in the NVT ensemble at no extra computational cost.

II. BALANCE CONDITIONS, FACTORIZED METROPOLIS FILTER

For an MCMC algorithm to converge to the stationary distribution, it must satisfy the global balance condition for the stationary flows $\varphi_{a\to b}$ from configuration a to b: the total flow into a configuration a must equal the total flow out of it,

$$\sum_b \varphi_{b\to a} = \sum_c \varphi_{a\to c} = \pi(a), \tag{1}$$

where $\pi(a)$ is the statistical weight of configuration a, e. g. given by a Boltzmann factor. The flow must also satisfy an ergodicity requirement[21]. The global balance, Eq. (1), is in particular satisfied by the *detailed balance* condition which equates the flows between any two configurations a and b:

$$\varphi_{a\to b} = \varphi_{b\to a} \tag{2}$$

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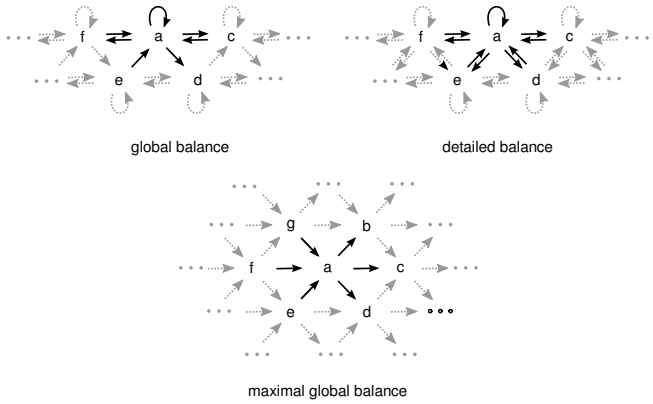

FIG. 1. Balance conditions for probability flow in Markov-chain Monte Carlo. Arrows represent stationary flows of equal magnitude. *Top left*: Global balance, a necessary condition for the convergence towards equilibrium. The total flow $\sum_c \varphi_{c\to a}$ into any configuration a must equal the total flow $\sum_c \varphi_{a\to c}$ out of it. The loops $\varphi_{a\to a}$, etc., correspond to rejected moves. *Top right*: Detailed balance: the net flow between any two configurations is zero, $\varphi_{a\to b} = \varphi_{b\to a}$. *Bottom*: Maximal global balance: $\varphi_{a\to b} > 0$ implies $\varphi_{b\to a} = 0$, the flow $\varphi_{a\to a}$ vanishes.

(see Fig. 1). We will be concerned with algorithms satisfying *maximal global balance*, where flow between two configurations is unidirectional and flows from a to a (that is: rejections) are avoided: if $\varphi_{a\to b} > 0$, then $\varphi_{b\to a} = 0$. In this case, probability does flow back nonlocally from b to a. In the particle systems that we consider, this happens *via* the periodic boundary conditions.

MCMC methods commonly rely on the Metropolis algorithm, which enforces detailed balance of the flows between a and b as follows:

$$\varphi_{a\to b} = \mathcal{A}_{a\rightleftharpoons b} \min(\pi(a), \pi(b)), \tag{3}$$

In our algorithm, the a-priori probability $\mathcal{A}$ is symmetric and amounts to zero or a global constant that we drop for simplicity. Eq. (3) is manifestly symmetric in $\pi(a)$ and $\pi(b)$ so that, by construction, $\varphi_{a\to b} = \varphi_{b\to a}$. Since $\varphi_{a\to b} = \pi(a)p(a\to b)$, with p the acceptance probability, Eq. (3) is equivalent to the well-known Metropolis filter

$$p(a\to b) = \min\left(1, \frac{\pi(b)}{\pi(a)}\right) \tag{4}$$

that has been implemented in countless computer programs.

In statistical physics, the weight of a configuration a is often given by the Boltzmann factor $\pi(a) = \exp(-\beta E(a))$, where $E(a)$ is the energy of a and β is the inverse temperature, which we set to one for the majority of this article. Using the abbreviation

$$[x]^+ := \max(0, x) \quad (\geq 0), \tag{5}$$

we can write the Metropolis filter of Eq. (4) as

$$p(a\to b) = \min(1, \exp(-\Delta E)) = \exp\left(-[\Delta E]^+\right), \tag{6}$$

where $\Delta E = E(b) - E(a)$. This corresponds to the acceptance probability of a proposed move, whereas the rejection probability is $1 - p = 1 - \exp\left(-[\Delta E]^+\right)$.

We now consider an N-particle system with pair interactions $E = \sum_{\{i,j\}} E_{ij}$, where i and j, in our applications, label particles in D-dimensional space, but could also refer to spins or other degrees of freedom. The sum runs over all unordered pairs $\{i, j\}$ of particles. For such a system, the Metropolis filter has always been used as

$$\begin{aligned} p^{\mathrm{Met}}(a\to b) &= \min(1, \exp(-\sum_{\{i,j\}} \Delta E_{ij})) \\ &= \exp\Big(-[\sum_{\{i,j\}} \Delta E_{ij}]^+\Big). \end{aligned} \tag{7}$$

In the present article, however, we introduce a *factorized Metropolis filter*

$$\begin{aligned} p^{\mathrm{fact}}(a\to b) &= \prod_{\{i,j\}} \min(1, \exp(-\Delta E_{ij})) \\ &= \exp\Big(-\sum_{\{i,j\}} [\Delta E_{ij}]^+\Big) \end{aligned} \tag{8}$$

which also fulfills detailed balance by respecting the same flow symmetries as the standard Metropolis filter, as can be seen by applying the identity $\pi(a)/\pi(b) = \exp(\sum_{\{i,j\}} [\Delta E_{ij}]^+ - [-\Delta E_{ij}]^+)$ to Eq. (8). The conventional and the factorized Metropolis filter agree in the hard-sphere case[1] and (trivially) for $N = 2$. They differ whenever terms of opposite sign appear on the rhs of Eq. (8), i. e. for general interactions and $N > 2$. The factorization increases the rate of rejections in a detailed-balance MC algorithm. We find that for the soft-sphere interactions considered in this article, the rate of rejections is about 50% higher (soft spheres with $n = 12$, $\rho = 0.8 \ldots 1.2$, with a step size of 0.1 in units of the particle diameter). However, the factorization yields the acceptance probability as a product of independent pair interaction terms. This will be the key to derive a rejection-free lifted MCMC algorithm for general N-particle systems.

III. LIFTING: 1D SYSTEMS AND TWO PARTICLES IN A BOX

We now introduce the concept of Markov chain lifting in a simple setting, which we later generalize to interacting particle systems. We consider a one-dimensional discrete system with configurations a and stationary weights $\pi(a)$ (i. e., $a \in [\Delta, 2\Delta, \ldots, L\Delta]$). For moves sampled uniformly from $\{-\Delta, \Delta\}$, the standard Metropolis filter of

Eq. (4) satisfies detailed balance $\varphi_{a\to a+\Delta} = \varphi_{a+\Delta\to a}\ \forall a$. The stationary distribution π is sampled in the limit of infinite running times.

Lifting[17–19], in this example, consists in duplicating each configuration a with a momentum-like variable into two configurations $a_\pm = (a, h = \pm 1)$. The lifting variable determines the next proposed move, which would in ordinary Metropolis MC be sampled from a prior distribution: For a_+, only the particle move $a \to a + \Delta$ is proposed, and for a_-, only $a \to a - \Delta$. For flow balance, we introduce *lifting moves* $a_+ \to a_-$ and $a_- \to a_+$ which take effect if the particle move is rejected, as summarized in the diagram,

$$\begin{array}{ccccccccc} \cdots & \longrightarrow & (a-\Delta)_+ & \xrightarrow{\varphi_0} & a_+ & \xrightarrow{\varphi_1} & (a+\Delta)_+ & \longrightarrow & \cdots \\ & & \uparrow\downarrow & & \uparrow\downarrow & & \uparrow\downarrow & & \\ & & {\scriptstyle [\varphi_1-\varphi_0]^+} & & {\scriptstyle [\varphi_0-\varphi_1]^+} & & & & \\ \cdots & \longleftarrow & (a-\Delta)_- & \xleftarrow[\varphi_0]{} & a_- & \xleftarrow[\varphi_1]{} & (a+\Delta)_- & \longleftarrow & \cdots \end{array} \tag{9}$$

where the flows φ_0 and φ_1 are given by

$$\varphi_0 = \min[\pi(a), \pi(a-\Delta)], \tag{10}$$

$$\varphi_1 = \min[\pi(a), \pi(a+\Delta)]. \tag{11}$$

We take the weights of the lifted configurations to be the same as the weights of the original configurations, $\pi(a_\pm) = \pi(a)$, adjusting the constant of normalization. This rejection-free MCMC algorithm satisfies maximal global balance, as only one of the two flows $\varphi_{a_+\to a_-}$ and $\varphi_{a_-\to a_+}$ can be non-zero. In the physical variables a, however, rejections are still present.

We now consider uniform stationary probabilities ($\pi(a) = \text{const}$) and impose hard-wall boundary conditions in our one-dimensional discrete model. The lifting flows are non-zero only for a rightmoving particle at $a = L\Delta$, and for a leftmoving particle at $a = \Delta$. For these configurations, the lifting flow equals the entire incoming flow, and the particle reverses its direction. One can show that the lifted algorithm visits all sites in $\mathcal{O}(L)$ steps, rather than in $\mathcal{O}(L^2)$ steps for the Metropolis algorithm.[2]

As demonstrated in Fig. 2, the discrete one-dimensional system with hard walls corresponds to two D-dimensional hard spheres that are constrained to move only, say, along the x direction, in a box with periodic boundary conditions. The new lifting variable i now indicates the moving sphere, and the hard wall turns into a no-overlap condition for the two spheres. Although the spheres only move towards the right, the algorithm satisfies maximal global balance due to flows across the periodic boundaries. Ergodicity for the unconstrained two-sphere problem in a D-dimensional box is achieved by resampling, after a fixed number of steps, the moving particle $i \in \{1, 2\}$ and the direction of motion $\boldsymbol{\Delta} \in \{+\boldsymbol{e}_\mathrm{x}\Delta, +\boldsymbol{e}_\mathrm{y}\Delta\}$ (for the example of two hard disks in a periodic box). The sequence of moves between resampling is referred to in the following as an *event chain*.

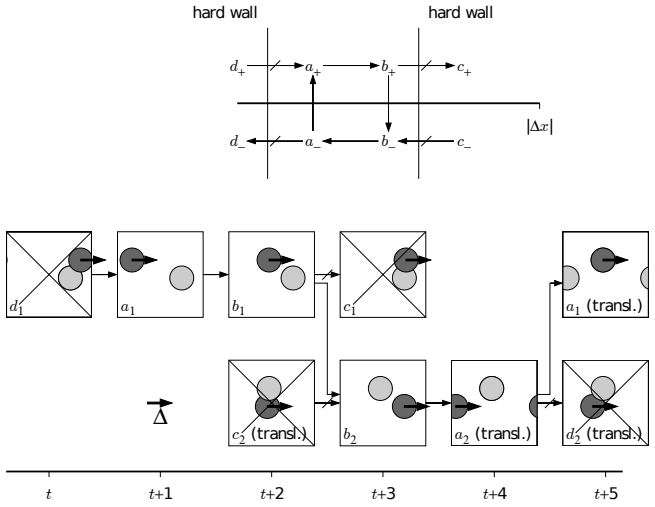

FIG. 2. *Upper*: Discrete one-dimensional system ($L = 2$) with constant probabilities $\pi(a) = \pi(b)$ and hard-wall conditions $\pi(c) = \pi(d) = 0$. The lifting variable $h = \pm 1$ corresponds to the direction of motion of the particle. *Lower*: Equivalent lifting algorithm for two hard spheres with finite displacement $\boldsymbol{\Delta}$ (the values of the lifting variable ± 1 are replaced by $\{1, 2\}$). The forbidden particle moves $b_1 \to c_1$ and $a_2 \to d_2$ trigger lifting moves. Maximal global balance is satisfied by moving in the $+x$ direction only. The equivalence between one-dimensional motion and two-particle dynamics in a constrained direction carries over to arbitrary pair potentials.

IV. INFINITESIMAL MOVES AND EVENT-CHAIN ALGORITHM FOR INTERACTING MANYPARTICLE SYSTEMS

We now extend the discussion of Section III to N-particle systems, first for hard spheres, and then for particles with arbitrary pairwise potentials. The idea to indicate the moving particle and its 'momentum' by lifting variables generalizes trivially to the N-particle case. Special care is, however, required to preserve global balance, and we show that the factorized Metropolis filter can be used to implement maximal global balance in the infinitesimal-move limit. We then implement this scheme efficiently in an event-driven MCMC algorithm.

Lifted configurations are now specified by the N hard-sphere centers $(\boldsymbol{r}_1, \ldots, \boldsymbol{r}_N)$, the moving sphere i and its direction of motion $\boldsymbol{\Delta}$. For concreteness, we focus on the positive x direction, $\boldsymbol{\Delta} = +\boldsymbol{e}_\mathrm{x}\Delta$, as before. A particle move is

$$a_i = (\boldsymbol{r}_1, \ldots, \boldsymbol{r}_i, \ldots, \boldsymbol{r}_N) \to b_i = (\boldsymbol{r}_1, \ldots, \boldsymbol{r}_i + \boldsymbol{\Delta}, \ldots, \boldsymbol{r}_N). \tag{12}$$

This algorithm violates global balance because it generates configurations with multiple overlaps, see Fig. 3. In the presence of a multiple overlap, it is impossible to define flows that satisfy the global balance condition. Multiple overlaps vanish, and maximal global balance is recovered, for infinitesimal moves $|\boldsymbol{\Delta}| \to 0$: In that limit, the factorized Metropolis filter identifies a unique colli-

sion partner, with probability one, since no two particles are at the same distance from the moving particle. The collision partner then inherits the lifting variable and moves forward in the next step. By a succession of infinitesimal steps that add up to a finite chain displacement ℓ, this reproduces the hard-sphere event-chain algorithm[6]. Of course, the infinitesimal-move algorithm is not implemented naively through a fine discretization, but rather by identifying the next lifting event, and then advancing the moving disk to contact.

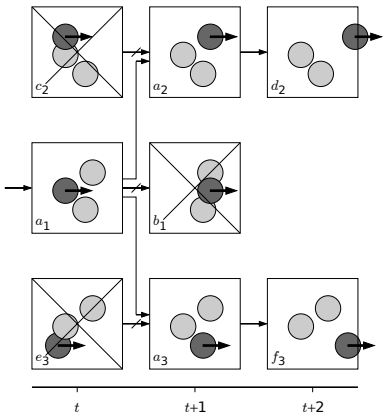

FIG. 3. Multiple overlaps for hard disks (weight π = const for the non-overlapping physical configurations a, d, f). The violation of the global balance condition is caused by the multiple overlap in configuration b (overlap of disk 1 with both disks 2 and 3): the flow into all legal configurations a, d, and f must be equal, while the illegal (crossed-out) configurations b, c, and e generate zero flow. The multiple overlap disappears, and global balance is again satisfied, in the limit $|\mathbf{\Delta}| \to 0$.

We now generalize the infinitesimal-move event-chain algorithm to arbitrary pair potentials, using the factorized Metropolis filter, Eq. (8). For general interactions, the energy change between configurations a_i and b_i which differ by an infinitesimal displacement $\mathrm{d}x_i$ of particle i is

$$\mathrm{d}E = E(b) - E(a) = \sum_{j(\neq i)} \frac{\partial E_{ij}(\boldsymbol{r}_j - \boldsymbol{r}_i)}{\partial x_i} \mathrm{d}x_i = \sum_{j(\neq i)} \mathrm{d}E_{ij}, \tag{13}$$

where $\mathrm{d}E_{ij}$ is the pairwise energy change, and E_{ij} the pair potential. According to the factorized Metropolis filter, the move is rejected with probability

$$1 - p^{\mathrm{fact}}(a_i \to b_i) = 1 - \exp\Big(-\sum_{j(\neq i)} [\mathrm{d}E_{ij}]^+\Big) = \sum_{j(\neq i)} [\mathrm{d}E_{ij}]^+. \tag{14}$$

Remarkably, for infinitesimal displacements, the rejection probability is a sum of pair terms, while the individual terms $[\mathrm{d}E_{ij}]^+$ normally neither add up to the total energy change $\mathrm{d}E$ nor to $[\mathrm{d}E]^+$. We use the terms in Eq. (14) as the probabilities for lifting moves

$$p^{\mathrm{lift}}(a_i \to a_j) = [\mathrm{d}E_{ij}]^+ \quad \forall j \neq i, \tag{15}$$

and obtain a rejection-free, maximal global balance MCMC algorithm. Fig. 4 illustrates that the total flow into the configuration a_i equals the total flow out of configuration a_i, satisfying the global balance condition Eq. (1). Explicitly, the lifting flows in the example of Fig. 4 are:

$$\varphi_{a_2 \to a_1} = \pi(a)[\mathrm{d}E_{21}]^+$$
$$\varphi_{a_3 \to a_1} = \pi(a)[\mathrm{d}E_{31}]^+$$

and the particle move flow,

$$\varphi_{b_1 \to a_1} = \varphi^{\mathrm{fact}}_{a \to b} = \pi(a)(1 - [\mathrm{d}E_{21}]^+ - [\mathrm{d}E_{31}]^+)$$

Indeed, Eqs. (14) and (15) define a rejection-free *infinitesimal* MCMC algorithm with maximal global balance.

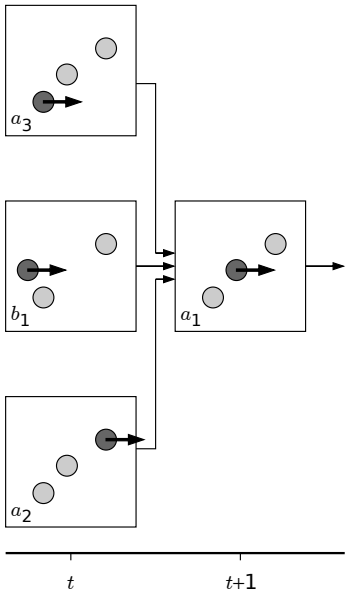

FIG. 4. Maximal global balance for N particles with arbitrary pair interactions (infinitesimal step, factorized Metropolis filter of Eq. (8)). Flow into a_1 is due to $N-1$ lifting moves (here, for $N = 3$, $a_2 \to a_1$ and $a_3 \to a_1$) and to a particle move (here, $b_1 \to a_1$). For infinitesimal steps, the flow into a_1 equals $\pi(a)$ (see text), thus balancing the flow $\pi(a)$ out of a_1 and satisfying global balance, Eq. (1).

In order to implement this algorithm efficiently, we choose an event-based approach. As mentioned, the factorized Metropolis filter ensures that no two lifting events can occur in the same infinitesimal timestep. Thus, every interaction of the moving particle with another can be treated independently of other interactions. Further, for fixed partner j, the lifting probabilites at successive timesteps are independent, and they vanish if the pair potential decreases during displacement. Following the BKL algorithm[15,25], we determine the displacement until the first lifting move occurs by sampling a uniform

random number Υ_{ij} from $(0,1]$ which determines the admissible energy increase until lifting, $E^*_{ij} = -\ln \Upsilon_{ij}$. The displacement until lifting s_{ij} is then found from

$$E^*_{ij} = \int_0^{E^*_{ij}} [\mathrm{d}E_{ij}]^+ = \int_0^{s_{ij}} \left[\frac{\partial E_{ij}(\boldsymbol{r}_j - \boldsymbol{r}_i - s\boldsymbol{e}_\mathrm{x})}{\partial s}\right]^+ \mathrm{d}s. \tag{16}$$

If this equation lacks a solution due to the shape of the interaction potential, or due to a large thermal excitation E^*_{ij}, no lifting event is generated, $s_{ij} = \infty$. While solving Eq. (16) can be nontrivial in general, we give a fast method for the most usual pair potentials below. The smallest of the $N-1$ independent $\{s_{ij}\}_{j\neq i}$ determines the lifting move $i \to j^*$ which will actually take place, advancing the moving disk by $\min_{j\neq i}(s_{ij}) = s_{ij^*}$ in the $+\boldsymbol{e}_\mathrm{x}$ direction and changing the lifting variable to j^*. We also make sure that the total displacement in a single event chain equals the chain displacement ℓ, which usually requires truncating the final event. After the end of the chain, the lifting variables are resampled. Each chain thus consists in an infinite succession of infinitesimal moves that add up to the chain displacement ℓ. Alternatively, we could introduce a small constant probability for terminating a chain in each infinitesimal move. This would effectively lead to exponentially distributed random ℓ, and is also a valid MCMC algorithm.

We conclude with some practical remarks on solving Eq. (16) for model potentials that occur in practice. Many model potentials are central potentials, $E_{ij}(\boldsymbol{r}_{ij}) = E_{ij}(r_{ij})$. If the pair potential consists of several terms, e. g. attractive and repulsive terms, it may be convenient to treat them separately by a further factorization of the Metropolis filter, and decompose the lifting probabilities, $[\mathrm{d}E^{\mathrm{attr}}_{ij}]^+ + [\mathrm{d}E^{\mathrm{rep}}_{ij}]^+$, where E^{attr} and E^{rep} are the attractive and repulsive parts of the pair potential. This decomposition can lead, however, to a higher event rate than the full potential. For instance, the Lennard-Jones potential reduces to two soft-sphere interactions, one attractive and one repulsive, and the mean free path between events is reduced at most by half in comparison to the case without decomposition. In return, the decomposition greatly simplifies the Monte Carlo program.

We will thus focus here on the case of soft-sphere potentials which are monotonous. A lifting move can only be generated if the moving particle is in the rising part of the pair potential. In this case, solving Eq. (16) amounts to sampling the energy increase $E^*_{ij} = -\log \Upsilon_{ij}$, with Υ_{ij} a uniform random number from $(0,1]$, which fixes the interaction energy $E^{\mathrm{lift}}_{ij} = E_{ij}(r_{ij}) + E^*_{ij}$, and thus the interparticle distance $r^{\mathrm{lift}}_{ij} = E^{-1}_{ij}(E^{\mathrm{lift}}_{ij})$ at the lifting move. (If E^{lift} exceeds any possible value of the interaction potential, there is no lifting move generated.) The admissible displacement s_{ij} for the i,j particle pair is then the positive root of $r^{\mathrm{lift}}_{ij} = |\boldsymbol{r}_j - \boldsymbol{r}_i - s_{ij}\boldsymbol{e}_\mathrm{x}|$. Again, if no such root exists, no lifting move is generated, and particle i will pass particle j. Using this method, and the decomposition into attractive and repulsive terms, Eq. (16) is thus easily computable for a large range of potentials.

In systems with periodic boundary conditions, for very long chains, a particle can pass by the same collision partner more than once: The pair potential no longer is monotonous. This is most easily avoided by tuning the chain displacement so that the moving sphere can only interact with one periodic image of the other spheres or by introducing a lifting move of the moving sphere with itself after a displacement of half the box. This move does not change the statistics of the following events. After it, the next event is computed as usual.

V. SPEEDUP WITH RESPECT TO METROPOLIS MONTE CARLO

We now compare the performance of the generalized ECMC algorithm with Metropolis Monte Carlo (MMC). As an application, we consider a two-dimensional system of N particles interacting with a truncated pairwise power-law potential, $E_{ij}(r) = \tilde{E}(\min(r, r_\mathrm{c}))$, with $\tilde{E}(r) = \epsilon(\sigma/r)^n$ and $r_\mathrm{c} = 1.8\sigma$, σ being the particle diameter. This potential includes important physical interactions such as the dipole interaction in magnetic colloids[26], hard disks ($n \to \infty$) and Lennard-Jones particles, once decomposed into a repulsive soft-sphere interaction ($n = 12$) and an attractive one ($n = 6$). In comparison to MMC, the ECMC algorithm uses more random numbers, one per interaction term, whereas Metropolis uses one per step. In our implementations, however, the main computational workload is the evaluation of the potentials, not the generation of random numbers (using the Mersenne Twister). One event of the ECMC algorithm is thus implemented in the same amount of time as one attempted step of MMC. ($3.2 \cdot 10^9$ steps per hour in MMC, $1.5 \cdot 10^9$ events per hour in ECMC). We compare the performance of the algorithms in terms of the CPU time used (see Fig. 5 for details).

As estimate of the relative performance of the algorithms we consider as in other recent work[6,8,9] the autocorrelation time τ_6 of the global orientational order parameter Ψ_6,

$$\Psi_6 = \frac{1}{N} \cdot \sum_{i,j} \frac{A_{ij}}{A_i} \exp(6\mathrm{i}\theta_{ij}) \tag{17}$$

where θ_{ij} is the angle of the bond vector between particles i and j against a fixed axis and A_{ij}/A_i the contribution of particle j to particle i's Voronoi cell perimeter[27,28]. With Ψ_6 being a global observable, we assume that its autocorrelation time τ_6 is representative for the mixing time for dense liquid states close to the freezing point, located at $\rho\sigma^2 = 1$ for $n = 12$, and $\rho\sigma^2 \approx 0.89$ for harder disks with $n = 1024$, where $\rho\sigma^2$ is the dimensionless density, $N\sigma^2/V$.

We find that in terms of CPU time, in small systems, ECMC mixes a few times quicker than MMC.

We tuned both algorithms to their optimal parameters (see Fig. 5 for details). Speedup, defined as the ratio $\tau_6(\mathrm{MMC})/\tau_6(\mathrm{ECMC})$, is, in the region of study, not found to be a strong function of density (Fig. 5, bottom row). For increasing system size, however, the speedup increases (Fig. 5, top row). An increase of speedup with system size has also been found for hard-sphere systems, where it approaches two orders of magnitude in large systems of 10^6 particles[8]. We thus expect that the generalized ECMC algorithm has similar characteristics with respect to system size as the hard-sphere ECMC algorithm. For very large systems, the MMC algorithm does not equilibrate within the allotted simulation time: The distribution of $|\Psi_6|$ is not yet stationary, even though the simulation time exceeds the τ_6 by a factor of 100. Thus, we have not determined τ_6 for these systems.

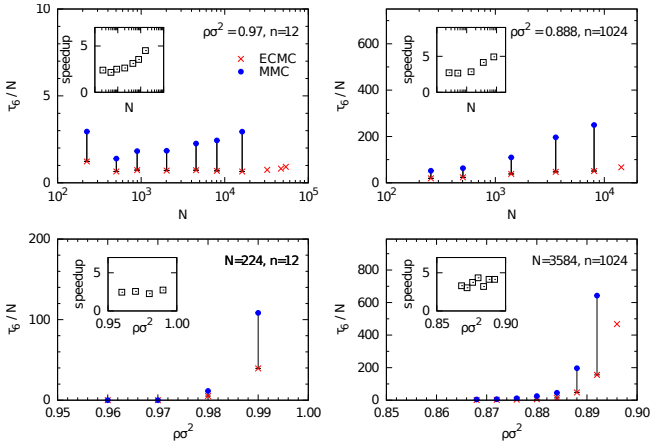

FIG. 5. Autocorrelation times τ_6 of the Ψ_6 orientational order parameter for event-chain Monte Carlo (ECMC) and Metropolis Monte Carlo (MMC), for soft-disk systems of N particles at inverse temperature $\beta = 1$. τ_6 is measured in arbitrary though comparable amounts of CPU time: One unit of CPU time for ECMC is a displacement of $T = 100N\sigma$, chain displacement $\ell = 0.025\sqrt{N}\sigma$, spanning thus about half the system volume. Event chains in $+x$ and $+y$ alternate every $\Delta T = N\sigma/2$ of displacement. One unit of CPU time for Metropolis consists of $1000N$ moves, where a move is the attempt to displace a particle by a random vector sampled from a disk of radius 0.16σ.

VI. DIRECT PRESSURE COMPUTATION

In order to obtain the equation of state in the NVT ensemble for the particle system under study, the pressure P must be computed. Usually, the pressure is obtained using the virial theorem (see Sec. 2.2 of[30]), either by averaging the virial, or by integration of the product of the static pair correlation function $g(r)$ and the pair potential $E_{ij}(r)$. Direct averaging is not possible for hard-sphere interactions, since the potential is singular. It is thus required to compute a discrete approximation of $g(r)$ and extrapolate it to the contact value to obtain P. Even for non-singular steep potentials, the approach via $g(r)$ is bothersome, since the dominant contributions to P come from close pairs (for the family of power-law potentials, $r \approx \sigma$), which is poorly sampled in the canonical ensemble. Finally, evaluation of the virial during the simulation implies extra computation for evaluating the forces. By contrast, in hard-sphere event-driven molecular dynamics the virial pressure is directly related to the collision rate, which is a trivial byproduct of the computation[20]:

$$\beta P = \rho - \frac{\beta\rho m}{2T_{\mathrm{sim}}N} \sum_{\text{collisions}} b_{ij}, \tag{18}$$

where $\rho = N/V$ is the particle number density, T_{sim} the total simulation time, m the mass of a particle, and $b_{ij} = (\boldsymbol{r}_i - \boldsymbol{r}_j)(\boldsymbol{v}_i - \boldsymbol{v}_j)$, with $\boldsymbol{r}_{i,j}, \boldsymbol{v}_{i,j}$ the positions and velocities of the colliding particles. In the following we show that in ECMC, the rate of lifting moves is, just like the collision rate in event-driven MD, directly related to the pressure. We give an elementary derivation independent of the virial theorem for the soft-particle case. The results are, however, also valid for hard particles and can be derived using arguments by Speedy[29] which connect the pressure to the stochastic geometry of the admissible configurations.

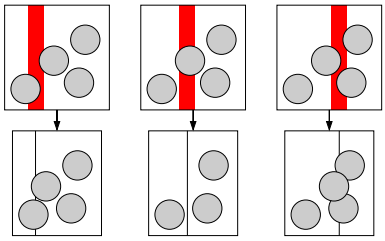

FIG. 6. Virtual rift volume changes by random removal of an infinitesimal strip from a hard-sphere configuration. *Left*: A successful removal, *Center*: Elimination of a particle (ideal gas pressure), *Right*: Generation of an overlap (excess pressure). The left and right cases become indistinct for soft interactions.

In order to compute the pressure $\beta P = \partial \ln Z/\partial V$, we consider virtual *rift volume changes* effected by removing a randomly located strip of size $\mathrm{d}L_x \times L_y$ from the system (see Fig. 6). By considering all positions of the strip, this procedure yields all N-particle configurations in the smaller simulation box, and thus the new partition function $Z(V + \mathrm{d}V)$. For isotropic systems, we recover the virial expression,

$$\beta P = \rho + \frac{1}{V} \left\langle \sum_{\{i,j\}} (x_j - x_i) \frac{\beta \partial E_{ij}(\boldsymbol{r}_i - \boldsymbol{r}_j)}{\partial x_i} \right\rangle, \tag{19}$$

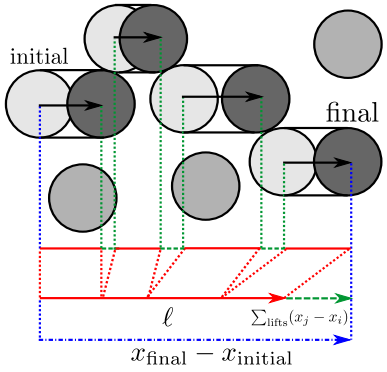

FIG. 7. Direct computation of the pressure: The excess pressure is derived from the ratio of excess displacement (green dashed lines, $\sum_{\text{lifts}}(x_j - x_i)$) and the chain displacement ℓ (red solid line). For isotropic systems, only the distance between the final and initial particle $x_{\text{final}} - x_{\text{initial}}$ (blue dash-dot arrow) has to be recorded.

n	N	$\rho\sigma^2$	virial pressure	ECMC pressure
12	2^{14}	0.990	14.4369 ± 0.0058	14.4267 ± 0.0038
48	2^{14}	0.860	8.753 ± 0.011	8.7565 ± 0.0023
48	2^{14}	0.888	9.441 ± 0.025	9.429 ± 0.027
1024	2^{14}	0.888	9.174 ± 0.028	9.1679 ± 0.0026
∞ (HS)	2^{16}	0.888	9.1667 ± 0.0073	9.1723 ± 0.0064
12, 6 (LJ)	2^{14}	0.888	1.44833 ± 0.00031	1.447623 ± 0.000045

TABLE I. Comparison of pressure computed using the virial expression and from excess displacement in ECMC Eq. (20), for repulsive soft and hard sphere (HS) interactions, and for the Lennard-Jones (LJ) potential, at $\beta = 1$. Pressures and densities are nondimensionalized, $\beta P\sigma^2$ and $\rho\sigma^2$. The deviations given are standard errors from 10 independent simulations each. For LJ, the potential was decomposed into attractive and repulsive parts.

where $\langle\cdot\rangle$ is the canonical average. The first term of the rhs in Eq. (19) is due to particles located in the removed strip, which lead to illegal configurations with less than N particles (Fig. 6, center). This term yields the ideal-gas pressure. The non-ideal contribution to the pressure results from changes in the Boltzmann weight due to compressed bonds, with $(x_j - x_i)$ accounting for the probability of a bond traversing the removed strip. In hard spheres, this term is produced by particle overlap (Fig. 6, right panel). Replacing the canonical average in Eq. (19) by an average in the lifted canonical ensemble, $\langle\cdot\rangle_k := N^{-1}\sum_k\langle\cdot\rangle$, where k is the lifting variable, one of the sums collapses and yields a factor of N; we recover the probabilities for a lifting move from $i \to j$ and $j \to i$. An ECMC simulation will reproduce the lifted canonical average and thus yields an unbiased estimator of the pressure. Summing up all the lifting events in a chain (see Fig. 7), we obtain

$$\beta P = \rho \cdot \left\langle \frac{x_{\text{final}} - x_{\text{initial}}}{\ell} \right\rangle_{\text{chains}}, \tag{20}$$

where $\langle\cdot\rangle_{\text{chains}}$ is the average over event chains, x_{initial} is the position of the first particle before the effects of the chain, and x_{final} the position of the last particle after, adjusted for periodic boundaries if necessary. Thus, it suffices to know the beginning and end of event chains to compute the pressure. Explicitly,

$$x_{\text{final}} - x_{\text{initial}} = \ell + \sum_{\text{lifts}}(x_j - x_i) \tag{21}$$

where x_i and x_j are the positions of the moving particle i and of the hit particle j, respectively, *at lifting*, see Fig. 7. In the ideal gas, there are no lifting moves and Eq. (20) reduces to the ideal gas pressure. The excess displacement $(x_j - x_i)$ can be negative for an interaction potential with attractive components, such as Lennard-Jones. If the potential is decomposed into attractive and repulsive parts as outlined in Section IV, individual excess displacements for the two potentials also add up to the correct pressure. As evidenced by Table I, the results obtained from ECMC via Eq. (20) agree with the conventional virial approach. Since no extra computation is required, the procedure via the excess displacement in ECMC is more efficient than the virial approach, in particular for steep potentials.

Finally, one might be interested in anisotropic systems where the collision rates can depend on the direction of the event chains. In this case, the derivation presented for longitudinal rifts (removing strips normal to the chain direction) is supplemented with an analogous result for transverse rifts (removing strips aligned with the event chain), which leads in D dimensions to the full pressure,

$$\beta P = \rho + \left\langle \frac{\rho}{D\ell} \sum_{\text{lifts}} \frac{(\boldsymbol{r}_j - \boldsymbol{r}_i)^2}{x_j - x_i} \right\rangle_{\text{chains}}, \tag{22}$$

where x is the coordinate parallel to the chain direction. More generally, the full stress tensor τ can be computed as an average of the dyadic product of the interparticle distance $\boldsymbol{r}_{ij} = \boldsymbol{r}_j - \boldsymbol{r}_i$ at collision:

$$\beta\tau = -\rho\mathbf{1} - \left\langle \frac{\rho}{\ell} \sum_{\text{lifts}} \frac{\boldsymbol{r}_{ij}\boldsymbol{r}_{ij}^{\text{t}}}{x_j - x_i} \right\rangle_{\text{chains}}, \tag{23}$$

where $\mathbf{1}$ is the identity matrix in D dimensions.

CONCLUSION

In the present article, we have generalized the event-chain Monte Carlo algorithm from hard spheres to particle systems interacting with arbitrary pair potentials, such as Lennard-Jones liquids or soft disks. The resulting

algorithm is faster than conventional Metropolis Monte Carlo, with the gap in performance increasing with the system size. It is based on the lifting concept, and relies on a new factorization of the Metropolis filter, applied to infinitesimal Monte Carlo moves, to achieve maximal global balance. The infinitesimal moves are implemented efficiently in an event-based algorithm using the BKL approach. The algorithm generates a continuum of samples of the equilibrium distribution. This has allowed us to derive the pressure and the stress tensor in the NVT ensemble directly from the simulation without any additional computation. Even though presented in periodic boundary conditions, the algorithm also applies to non-periodic systems, by introducing chains in the $-x$ and $-y$ direction to render it ergodic.

Infinitesimal moves permit to apply the framework of lifted Markov chains to the interacting particles problem, since they define uniquely the next event, while satisfying global balance. By subdivision into infinitesimal moves, both the original hard-sphere event-chain algorithm[6], and the hybrid MC algorithm of Peters and de With[15] are revealed to be lifting algorithms. Lifting improves mixing in large, strongly correlated systems, since clusters of particles are displaced in a cooperative way. It is thus applicable to packing problems, glassy systems, etc. particularly, as its dynamics are fundamentally different from the MMC or MD case. As the original event-chain algorithm, it can be parallelized[9]. We expect the algorithm to extend to complex fluids, with particles possessing internal degrees of freedom, to path integral (quantum) Monte Carlo, and other sampling problems.

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[1]The standard Metropolis filter and its factorized variant agree also for discretized interaction potentials with infinitesimal moves, as considered in Ref[15].

[2]As it stands, the lifting algorithm is deterministic. To make it aperiodic, small lifting flows, and also small on-site probabilities must be added to Eq. (9). This can be done[18] preserving the mixing time $\propto L$ for the lifting algorithm, whereas local MCMC algorithms mix in $\propto L^2$ steps. This mathematical result has been considerably generalized[22].

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