Variance Reduced Brownian Dynamics Simulations

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ABSTRACT: Brownian dynamics simulations can be made more efficient by incorporating the idea of importance sampling. By introducing and compensating a bias in favor of those configurations which mainly contribute to the average of a given quantity of interest, one can considerably reduce the variance of the stochastic simulation results. This idea can be applied to general stochastic differential equations of motion and is hence not restricted to Brownian dynamics simulations. The construction of variance reduced simulations requires an approximate understanding of the dynamics described by the underlying stochastic differential equation. The basic procedure is first developed in general and then illustrated for the example of a Hookean dumbbell solution in start up of steady shear flow. Particular emphasis is put on the development of a clearly structured formulation of the procedure which immediately allows for variance reduced simulations of nonlinear models. Possible applications in polymer kinetic theory and in the flow calculation of viscoelastic liquids are discussed.

I. Introduction

Brownian dynamics simulations of bead–spring models constitute a very powerful tool for understanding the dynamics of polymers in dilute solutions, both at equilibrium and in solutions undergoing flow.[1,2] Such simulations are based on the numerical integration of the equations of motion for the beads of a single chain molecule, where the presence of Brownian or random forces implies that the equations of motion are stochastic differential equations. Brownian dynamics simulations provide a straightforward and rigorous method for investigating nonlinear effects such as hydrodynamic interaction, excluded volume, or finite polymer extensibility.

The idea of Brownian dynamics is not limited to dilute solutions. In order to realize this, one should note that all kinetic theory models for the dynamics of polymers in solutions and melts are formulated in terms of diffusion equations for a suitable configurational distribution function.[3] Such diffusion equations, which are often referred to as Fokker–Planck equations, are equivalent to stochastic differential equations of motion.[2,4,5] More precisely, this equivalence can be formulated as follows: The polymer configurations are described by the variables occurring in the list of arguments of the configurational distribution function (except for time). The average of any function of the configurational variables can either be evaluated as an integral, weighted with the probability distribution obtained by solving the diffusion equation, or as an ensemble average for an ensemble of trajectories constructed from the stochastic differential equation (in this paper, we are interested only in averages and not in the individual trajectories). These averages are identical, including their time dependencies, even if the configurations at several different times are involved. This idea has been used, for example, to generalize the idea of Brownian dynamics simulations by writing down the stochastic differential equations of motion for reptation models.[6] Numerical integration of such stochastic differential equations leads to stochastic simulation techniques for the underlying models. For reptation models, the simulation algorithm is so simple that even complicated problems, such as recoil phenomena, can be treated without much effort.[7,8]

Simulation techniques are not only a very powerful tool for investigating kinetic theory models for various kinds of systems. Simulations of polymer dynamics have gained additional importance because they can be combined with the finite element method in order to calculate complex flows of polymeric liquids. The CONNFFESSIT idea (calculation of non-Newtonian flow: finite elements & stochastic simulation technique) makes it possible to calculate flows without using a constitutive equation—like in real liquids, the stresses are obtained directly from the configurations of a large number of polymer molecules in the system.[9] The momentum balance equation is solved together with the stochastic differential equations of motion for the molecules rather than together with a constitutive equation. Since complex flow calculations require the simulation of very large ensembles of polymer chains, the development of efficient simulation techniques is crucial for the successful implementation of the CONNFFESSIT idea.

The purpose of this paper is to describe and illustrate the idea behind variance reduced stochastic simulation techniques. A reduced variance implies that the simulation of a smaller ensemble is sufficient for reaching a given tolerance for the statistical error so that the simulations become more efficient. While the standard numerical integration schemes for stochastic differential equations—which we briefly review in the next section—are universal brute force methods, variance reduction requires deeper insight into the physical problem. An approximate description of the dynamics is required in order to construct a transformation of the stochastic differential equation which leads to rigorous results for the averages of interest and, moreover, to smaller statistical error bars. We develop a method of variance reduction based on the idea of importance sampling, and we then illustrate the ideas for the Hookean dumbbell model of dilute polymer solutions. A brief summary and outlook conclude the paper.

II. Stochastic Simulation Technique

The equations for the polymer dynamics in kinetic theory are usually diffusion equations (or Fokker–Planck

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equations) for the configurational distribution function. These equations are of the general form

$$\frac{\partial}{\partial t}p(t,\boldsymbol{q}) = -\frac{\partial}{\partial \boldsymbol{q}}\cdot[\boldsymbol{A}(t,\boldsymbol{q})p(t,\boldsymbol{q})] + \frac{1}{2}\frac{\partial}{\partial \boldsymbol{q}}\frac{\partial}{\partial \boldsymbol{q}}:[\boldsymbol{D}(t,\boldsymbol{q})p(t,\boldsymbol{q})] \tag{1}$$

where $\boldsymbol{q}$ and $\boldsymbol{A}(t,\boldsymbol{q})$ are d-dimensional column vectors representing the configuration vector and drift term, respectively, and $\boldsymbol{D}(t,\boldsymbol{q})$ is a positive semidefinite, symmetric $d \times d$ matrix representing the diffusion term. If we decompose $\boldsymbol{D}$ into the form $\boldsymbol{D} = \boldsymbol{B}\cdot\boldsymbol{B}^{\mathrm{T}}$, where $\boldsymbol{B}$ is a $d \times d'$ matrix, then the above Fokker–Planck equation is equivalent to the stochastic differential equation[2,4,5]

$$\mathrm{d}\boldsymbol{Q}(t) = \boldsymbol{A}(t,\boldsymbol{Q}(t))\,\mathrm{d}t + \boldsymbol{B}(t,\boldsymbol{Q}(t))\cdot\mathrm{d}\boldsymbol{W}(t) \tag{2}$$

where the stochastic process $\boldsymbol{W}(t)$ is a d'-dimensional Wiener process. For clarity, we here use different symbols for the time-dependent d-dimensional random variable $\boldsymbol{Q}(t)$ and the corresponding dummy variable $\boldsymbol{q}$ in the probability density at time t. Rather than giving rigorous definitions of the Wiener process and of the solution of the stochastic differential equation (2), which is a Markov process, we describe the discrete Euler approximation. We consider the time interval $[0, t_{\max}]$ and introduce a partition $0 = t_0 < t_1 \ldots < t_{n-1} < t_n = t_{\max}$. For a given initial condition, we construct the approximations $\boldsymbol{Q}_j$ to $\boldsymbol{Q}(t_j)$ by means of the iteration scheme

$$\boldsymbol{Q}_{j+1} = \boldsymbol{Q}_j + \boldsymbol{A}(t_j,\boldsymbol{Q}_j)\Delta t_j + \boldsymbol{B}(t_j,\boldsymbol{Q}_j)\cdot\boldsymbol{W}_j(\Delta t_j)^{1/2} \tag{3}$$

where $\Delta t_j = t_{j+1} - t_j$ and all the d'-dimensional column vectors $\boldsymbol{W}_j$ have independent Gaussian components with mean 0 and variance 1, that is,

$$\langle \boldsymbol{W}_j \rangle = \boldsymbol{0} \qquad \langle \boldsymbol{W}_j\boldsymbol{W}_k \rangle = \delta_{jk}\boldsymbol{1} \tag{4}$$

The Kronecker delta, δ_{jk}, implies that the Gaussian random vectors for different time steps are independent. The pointed brackets $\langle \ldots \rangle$ without subscript always denote expectations (or averages) over the underlying probability space.[2]

For a given initial condition $\boldsymbol{Q}_0$, which is assumed to be stochastically independent from the Gaussian random variables $\boldsymbol{W}_j$, the approximation scheme (3) can readily be used for constructing random trajectories on a computer. It is very important to note that the function $\boldsymbol{B}$ in the diffusion term is evaluated with the configuration at time t_j. This choice is dictated by the assumed form of (1) and is known as the Itô approach. An important advantage of the Itô approach to stochastic differential equations is that the resulting equations are ideally suited for computer simulations. The price to pay is that the rules for transforming stochastic differential equations (the rules of Itô calculus) deviate from those of familiar deterministic calculus.[2,4,5]

For a given realization $\boldsymbol{q}_j$ of $\boldsymbol{Q}_j$, $\boldsymbol{Q}_{j+1}$ is a Gaussian random variable with average $\boldsymbol{q}_j + \boldsymbol{A}(t_j,\boldsymbol{q}_j)\,\Delta t_j$ and covariance matrix $\boldsymbol{D}(t_j,\boldsymbol{q}_j)\Delta t_j$, that is

$$p(\boldsymbol{Q}_{j+1} = \boldsymbol{q}_{j+1}|\boldsymbol{Q}_j = \boldsymbol{q}_j) = [(2\pi\Delta t_j)^d \det(\boldsymbol{D}(t_j,\boldsymbol{q}_j))]^{-1/2} \times \exp\left\{-\frac{1}{2\Delta t_j}[\boldsymbol{q}_{j+1} - \boldsymbol{q}_j - \boldsymbol{A}(t_j,\boldsymbol{q}_j)\Delta t_j]\cdot\boldsymbol{D}^{-1}(t_j,\boldsymbol{q}_j)\cdot[\boldsymbol{q}_{j+1} - \boldsymbol{q}_j - \boldsymbol{A}(t_j,\boldsymbol{q}_j)\Delta t_j]\right\} \tag{5}$$

Equation 3, which leads to the transition probabilities (5), constitutes the simplest integration scheme for stochastic differential equations. It can be shown that the discretization error for any average is linear in the maximum time step width. A comprehensive discussion of more sophisticated integration schemes can be found in ref 10.

III. Variance Reduction

The approach to variance reduction developed here was inspired by section 16.3 of ref 10. We generalize the ideas and the formulation so that they become directly applicable to a wide class of stochastic differential equations. Apparently, the variance reduction technique described in this paper has not previously been used in studying polymer dynamics. We believe that the idea of improving the efficiency of simulations by means of approximate solutions is so appealing, and the perspectives of the approach are so promising, that this approach should be systematically explored in polymer kinetic theory. We here want to describe the way to a successful application of variance reduced simulations based on stochastic differential equations.

A. Importance Sampling. In the usual approach, a direct simulation of a kinetic theory model defined in terms of a diffusion equation of the form (1) is based on the numerical integration scheme (3). We wish to develop a more efficient simulation technique. The variance reduction method pursued in the following is based on the same fundamental idea as Monte–Carlo integration schemes, namely importance sampling. We first explain the background of Monte–Carlo integration by considering the mean-square extension of Hookean dumbbells at equilibrium. We then develop the general ideas and formulas which eventually are made more and more explicit.

For a Hookean dumbbell at equilibrium, the Boltzmann distribution is

$$p_{\mathrm{eq}}(\boldsymbol{q}) = (2\pi k_{\mathrm{B}}T/H)^{-3/2}\exp\left\{-\frac{1}{2}\frac{H\boldsymbol{q}^2}{k_{\mathrm{B}}T}\right\} \tag{6}$$

where $\boldsymbol{q}$ is a a three-dimensional column vector representing the connector or configuration vector of the dumbbell, H is the Hookean spring constant, k_{B} is Boltzmann's constant, and T is the absolute temperature. In order to avoid unnecessarily lengthy equations, all distances are measured in units of $(k_{\mathrm{B}}T/H)^{1/2}$. This corresponds to setting $k_{\mathrm{B}}T/H = 1$; the appropriate factors can be reintroduced by means of dimensional analysis, and we do that whenever it is convenient. By evaluating the required Gaussian integral we obtain

$$\langle \boldsymbol{Q}^2 \rangle_{\mathrm{eq}} = \int \boldsymbol{q}^2 p_{\mathrm{eq}}(\boldsymbol{q})\,\mathrm{d}^3q = 3 \tag{7}$$

where, as before, we have used the symbol $\boldsymbol{Q}$ for a random variable and $\boldsymbol{q}$ for the dummy variable associated with $\boldsymbol{Q}$.

We now discuss various possible approaches to obtaining $\langle \boldsymbol{Q}^2 \rangle_{\mathrm{eq}}$ by simulation. In a first approach, we simulate N_{T} column vectors $\boldsymbol{q}$ which are distributed according to the three-dimensional distribution $p_{\mathrm{eq}}(\boldsymbol{q})$ (that is, we simulate three independent standard Gaussian random variables). Since, according to Wick's theorem for decomposing higher moments of a Gaussian distribution (see (2.8.4) of ref 4 or (6.11) of ref 5), the variance of the corresponding random variable $\boldsymbol{Q}^2$ is $\Theta_1 = 2\langle \boldsymbol{QQ} \rangle_{\mathrm{eq}}:\langle \boldsymbol{QQ} \rangle_{\mathrm{eq}} = 6$, the statistical error bar for the simulation is $(\Theta_1/N_{\mathrm{T}})^{1/2}$.

In a second approach, we exploit the observation that $p_{eq}(q)$ depends only on the length of q which leads to

$$\langle Q^2\rangle_{eq} = (2\pi)^{-1/2}\int q^4 \exp\left\{-\frac{1}{2}q^2\right\} dq = 3 \qquad (8)$$

We then need to simulate only one standard Gaussian random variable Q so that we save a factor of 3 in computer time per realization. However, again according to Wick's theorem, the variance is now $\Theta_2 = 96$, so that we need 16 times as many realizations as in the first simulation in order to obtain the same statstical error bars. We have clearly moved in the wrong direction by trying to cut down computer time by a factor of 3.

Why does the second simulation lead to such large statistical errors? The answer to this question lies in the following observation. The Gaussian distribution is peaked around 0. The majority of the generated configurations Q is hence around 0, and such configurations make only a very small contribution to the ensemble average of Q^4. The rare configurations with large $|Q|$ make a very large contribution to that average, and this is the source of the large statistical error bars. It would be much better to preferably generate those configurations which contribute most to the integral, and to suitably estimate the required integral from those biased configurations. This is the idea of importance sampling in Monte-Carlo integration, and this is the key to the variance reduced simulations developed below. For the simulation based on the column vector $\boldsymbol{Q}$ rather than the scalar quantity Q, configurations which contribute most to the integral are generated more frequently (even though the factor $\boldsymbol{Q}^2$ is still unfavorable).

In a final third approach, we try a simulation which is even better adapted to the integral in (8) by rewriting the integral as

$$(2\pi)^{-1/2}\int q^4 \exp\left\{-\frac{1}{2}q^2\right\} dq = \int x^4 f_c(x) p_{red}(x)\, dx \qquad (9)$$

with a correction factor

$$f_c(x) = (2\pi)^{-1/2}\exp\left\{-\frac{1}{2}x^2\right\}/p_{red}(x) \qquad (10)$$

If we simulate realizations of a random variable X with a distribution given by $p_{red}(x)$, the ensemble average of $X^4 f_c(X)$ yields the desired integral $\langle \boldsymbol{Q}^2\rangle_{eq}$. The optimal adaptation to the integral is obtained for the probability density

$$p_{red}(x) = \frac{1}{3}(2\pi)^{-1/2}x^4 \exp\left\{-\frac{1}{2}x^2\right\} \qquad (11)$$

Then, the simulation result for $X^4 f_c(X)$ is equal to 3, independent of the realization, and the variance Θ_3 is reduced to zero. Notice that the exact result is contained in the normalization factor of $p_{red}(x)$. Even if, in general, the optimal density $p_{red}(x)$ cannot be constructed, the above discussion shows how the simulation should be adapted to the quantity of interest, and how the resulting bias can be removed by means of the correction factor f_c.

Guided by the ideas of our example of Hookean dumbbells at equilibrium, we can now develop the basic procedure for the construction of a variance reduced simulation for the stochastic differential equation (2). We first describe the general construction and we emphasize which conditional probability densities are required (in general, only approximate expressions are available). We then discuss the role of the approximations in variance reduced simulations.

Assume that the distribution of the initial d-dimensional random variable $\boldsymbol{Q}(t_0)$ at times t_0 is continuous with probability density $p^i(q)$. We wish to evaluate the average of a nonnegative function $h(\boldsymbol{Q}(t_{max}))$, where $\boldsymbol{Q}(t_{max})$ is obtained from the solution of the stochastic difference equation (2). For an optimal importance sampling we would like to simulate configurations with a probability density $p_{red}(q)$ proportional to

$$h(q)p^f(q)$$

where $p^f(q)$ denotes the probability density for the distribution of the random variable $\boldsymbol{Q}(t_{max})$,

$$p^f(q) = \int p(\boldsymbol{Q}(t_{max}) = q|\boldsymbol{Q}(t_0) = q_0)p^i(q_0)\, d^dq_0 \qquad (12)$$

Since the exact transition probability density for the solution of the underlying stochastic differential equation required in (12) is in general unknown, we need an approximation $p_{app}(\boldsymbol{Q}(t_{max}) = q|\boldsymbol{Q}(t_0) = q_0)$ as the *first basic input for variance reduced simulations*, and we define

$$p^f_{app}(q) \doteq \int p_{app}(\boldsymbol{Q}(t_{max}) = q|\boldsymbol{Q}(t_0) = q_0)p^i(q_0)\, d^dq_0 \qquad (13)$$

According to our previous considerations, we can introduce a correction factor

$$f_c(q) = \frac{p^f(q)}{p_{red}(q)} = \frac{p^f(q)}{p^f_{app}(q)}\frac{\langle h(\boldsymbol{Q}(t_{max}))\rangle_{app}}{h(q)} \qquad (14)$$

where $\langle h(\boldsymbol{Q}(t_{max}))\rangle_{app}$ is the approximate expression for the expectation of $h(\boldsymbol{Q}(t_{max}))$,

$$\langle h(\boldsymbol{Q}(t_{max}))\rangle_{app} \doteq \int h(q)p^f_{app}(q)\, d^dq$$

Even if we have an approximate expression $p^f_{app}(q)$ and we can simulate vectors according to that distribution, we still need to know the exact distribution $p^f(q)$ of $\boldsymbol{Q}(t_{max})$ in order to evaluate the correction factor $f_c(q)$. This problem is addressed in the next subsection.

B. Construction of Constrained Trajectories. We next want to estimate the exact probability density $p^f(q)$ appearing in the correction factor (14). The basic idea is to construct the required transition probability density in (12) from a sequence of transition probability densities at small time steps, as given in (5). We hence select a partition $t_0 < t_1 ... < t_{n-1} < t_n = t_{max}$ of the time range $T = [t_0, t_{max}]$, and we describe the construction of a variance reduced simulation which consists of the random variables $\tilde{\boldsymbol{Q}}_0$, $\tilde{\boldsymbol{Q}}_1$, ..., $\tilde{\boldsymbol{Q}}_n$ (we use different symboles $\tilde{\boldsymbol{Q}}_j$ and $\boldsymbol{Q}_j$ to clearly distinguish the variance reduced and direct simulation results for the exact solution $\boldsymbol{Q}(t_j)$). The construction of a trajectory proceeds in three steps:

1. Select a realization q_n of the final configuration $\tilde{\boldsymbol{Q}}_n$ according to a continuous distribution for which the density is proportional to $h(q)p^f_{app}(q)$.

2. Then, select a realization q_0 of the initial configuration $\tilde{\boldsymbol{Q}}_0$ according to a continuous distribution with density $p_{app}(\boldsymbol{Q}(t_0) = q_0|\boldsymbol{Q}(t_n) = q_n)$ where the function

$$p_{app}(\boldsymbol{Q}(t_0) = q|\boldsymbol{Q}(t_n) = q_n) \doteq p_{app}(\boldsymbol{Q}(t_n) = q_n|\boldsymbol{Q}(t_0) = q)\frac{p^i(q)}{p^f_{app}(q_n)}$$

is defined in terms of previously introduced probability densities.

3. Finally, for $j = 0, 1, ..., n-2$, select realizations $\boldsymbol{q}_{j+1}$ of the intermediate configurations $\boldsymbol{Q}_{j+1}$ according to the continuous distributions with densities

$$p_{\text{app}}(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j, \boldsymbol{Q}(t_n) = \boldsymbol{q}_n)$$

Note that the last step of this construction requires approximate conditional probability densities $p_{\text{app}}(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j, \boldsymbol{Q}(t_n) = \boldsymbol{q}_n)$ as the *second basic input for variance reduced simulations*. A very general idea for choosing these conditional probability densities in such a way that $\tilde{\boldsymbol{Q}}_{j+1}$ can be constructed very efficiently is suggested in the example of Hookean dumbbells in the subsequent section. Actually, the idea is to consider the random variable $\tilde{\boldsymbol{Q}}_{j+1}$ as the primary input rather than its probability density.

The reader should realize that the approximate probability densities required as basic inputs for variance reduced simulations are just sufficient for constructing the random variables $\tilde{\boldsymbol{Q}}_0, \tilde{\boldsymbol{Q}}_1, ..., \tilde{\boldsymbol{Q}}_n$; hence, no additional consistency conditions of the Chapman–Kolmogorov type[2,4,5] need to be verified. (This consistency condition for the family of transition probabilities of a Markov process states that if the transition probabilities for two successive small steps are combined, then the proper transition probability for the corresponding larger step must result.) The given initial distribution and the transition probability density $p_{\text{app}}(\boldsymbol{Q}(t_n) = \boldsymbol{q}_n|\boldsymbol{Q}(t_0) = \boldsymbol{q}_0)$ are necessary and sufficient for the simulation of $\tilde{\boldsymbol{Q}}_0$ and $\tilde{\boldsymbol{Q}}_n$. The conditional probability densities $p_{\text{app}}(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j, \boldsymbol{Q}(t_n) = \boldsymbol{q}_n)$ for $j = 0, 1, ..., n-2$ provide exactly the information needed to iteratively construct the intermediate random variables $\tilde{\boldsymbol{Q}}_1, \tilde{\boldsymbol{Q}}_2, ..., \tilde{\boldsymbol{Q}}_{n-1}$ if we assume that, like for the Markov process to be approximated, $\tilde{\boldsymbol{Q}}_{j+1}$ can depend on previous values only through $\tilde{\boldsymbol{Q}}_j$.

In the spirit of (14) we introduce the correction factor

$$f_c(\boldsymbol{q}_0,\boldsymbol{q}_1,...,\boldsymbol{q}_n) = f_c'(\boldsymbol{q}_0,\boldsymbol{q}_1,...,\boldsymbol{q}_n)\frac{\langle h(\boldsymbol{Q}(t_{\max}))\rangle_{\text{app}}}{h(\boldsymbol{q}_n)} \quad (15)$$

where

$$f_c'(\boldsymbol{q}_0,\boldsymbol{q}_1,...,\boldsymbol{q}_n) = \frac{p(\boldsymbol{Q}(t_n) = \boldsymbol{q}_n|\boldsymbol{Q}(t_{n-1}) = \boldsymbol{q}_{n-1})}{p_{\text{app}}(\boldsymbol{Q}(t_n) = \boldsymbol{q}_n|\boldsymbol{Q}(t_0) = \boldsymbol{q}_0)} \times \prod_{j=0}^{n-2}\frac{p(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j)}{p_{\text{app}}(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j, \boldsymbol{Q}(t_n) = \boldsymbol{q}_n)} \quad (16)$$

The factor f_c' is the ratio of the exact and the approximate probability density for finding the values $\boldsymbol{q}_0, \boldsymbol{q}_1, ..., \boldsymbol{q}_n$ at the times $t_0, t_1, ..., t_n$. If the transition probability densities $p_{\text{app}}(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j, \boldsymbol{Q}(t_n) = \boldsymbol{q}_n)$ are given by the exact Markovian results,

$$p_{\text{app}}(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j, \boldsymbol{Q}(t_n) = \boldsymbol{q}_n) = \frac{p(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j)p(\boldsymbol{Q}(t_n) = \boldsymbol{q}_n|\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1})}{p(\boldsymbol{Q}(t_n) = \boldsymbol{q}_n|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j)}$$

then the factor f_c' is equal to

$$p(\boldsymbol{Q}(t_n) = \boldsymbol{q}_n|\boldsymbol{Q}(t_0) = \boldsymbol{q}_0)/p_{\text{app}}(\boldsymbol{Q}(t_n) = \boldsymbol{q}_n|\boldsymbol{Q}(t_0) = \boldsymbol{q}_0)$$

and the correction factor f_c is very similar to the result in (14); after averaging the $\boldsymbol{q}_0$ dependence of f_c with the conditional probability density of $\tilde{\boldsymbol{Q}}_0$ given that $\tilde{\boldsymbol{Q}}_n = \boldsymbol{q}_n$ we indeed recover (14).

We now have the correction factor f_c in a form which, once the required approximate transition probability densities are given, can be used in simulations. In (15) and (16), only the exact small-step transition probabilities $p(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j)$ are required as an additional ingredient. If we choose sufficiently small time steps, any desired accuracy can be achieved for these transition probabilities by evaluating them by means of suitable numerical integration schemes. For example, for the Euler scheme, the exact transition probability density $p(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j)$ is replaced by the Gaussian probability density $p(\boldsymbol{Q}_{j+1} = \boldsymbol{q}_{j+1}|\boldsymbol{Q}_j = \boldsymbol{q}_j)$ given in (5). The occurrence of the exact transition probabilities in the correction factor is the reason why trajectories need to be considered. For the evaluation of averages considered in the next subsection we first assume that we know the exact small-step transition probabilities before we discuss the small systematic errors introduced by evaluating them for numerical integration schemes.

C. Evaluation of Averages. Since the correction factor compensates the bias introduced in the simulation we obtain

$$\langle h(\boldsymbol{Q}(t_n))\rangle = \langle f_c(\tilde{\boldsymbol{Q}}_0,\tilde{\boldsymbol{Q}}_1,...,\tilde{\boldsymbol{Q}}_n)h(\tilde{\boldsymbol{Q}}_n)\rangle \quad (17)$$

so that $f_c h(\tilde{\boldsymbol{Q}}_n)$ is indeed an estimate for the average of a function h at time $t_n = t_{\max}$. Equation 17 can be rewritten in the alternative form

$$\langle h(\boldsymbol{Q}(t_n))\rangle = \langle f_c'(\tilde{\boldsymbol{Q}}_0,\tilde{\boldsymbol{Q}}_1,...,\tilde{\boldsymbol{Q}}_n)\rangle\langle h(\boldsymbol{Q}(t_n))\rangle_{\text{app}} \quad (18)$$

The representation (18) clearly shows that we achieved our aim: $f_c'(\tilde{\boldsymbol{Q}}_0,\tilde{\boldsymbol{Q}}_1,...,\tilde{\boldsymbol{Q}}_n)\langle h(\boldsymbol{Q}(t_n))\rangle_{\text{app}}$ constitutes an estimate for the desired average $\langle h(\boldsymbol{Q}(t_n))\rangle$, and if all approximate transition probabilities coincide with the exact ones, then $f_c' = 1$ and the variance of the estimator is reduced to zero.

Since we now have information about the distribution at the intermediate times t_j, we obtain the following identity for an arbitrary function g,

$$\langle g(\boldsymbol{Q}(t_0),\boldsymbol{Q}(t_1),...,\boldsymbol{Q}(t_n))\rangle = \langle f_c(\tilde{\boldsymbol{Q}}_0,\tilde{\boldsymbol{Q}}_1,...,\tilde{\boldsymbol{Q}}_n)g(\tilde{\boldsymbol{Q}}_0,\tilde{\boldsymbol{Q}}_1,...,\tilde{\boldsymbol{Q}}_n)\rangle \quad (19)$$

Thus we have an alternative estimation for the average of an arbitrary function g. Note that the results (17) and (19) are exact regardless of the accuracy of the required approximate transition probabilities. However, the variance reduction crucially depends on the quality of the approximation. In order to see this we evaluate the variance of the estimator,

$$\text{Var}\,[f_c(\tilde{\boldsymbol{Q}}_0,\tilde{\boldsymbol{Q}}_1,...,\tilde{\boldsymbol{Q}}_n)g(\tilde{\boldsymbol{Q}}_0,\tilde{\boldsymbol{Q}}_1,...,\tilde{\boldsymbol{Q}}_n)] = \langle h(\boldsymbol{Q}(t_{\max}))\rangle_{\text{app}} \times \left\langle f_c(\boldsymbol{Q}(t_0),\boldsymbol{Q}(t_1),...,\boldsymbol{Q}(t_n))\frac{g^2(\boldsymbol{Q}(t_0),\boldsymbol{Q}(t_1),...,\boldsymbol{Q}(t_n))}{h(\boldsymbol{Q}(t_n))}\right\rangle - \langle g(\boldsymbol{Q}(t_0),\boldsymbol{Q}(t_1),...,\boldsymbol{Q}(t_n))\rangle^2 \quad (20)$$

Two requirements are crucial for a successful variance reduction. First, the ratio of the exact and the approximate probability density f_c' introduced in (16) should be close to 1. A good approximation to the trajectories of the underlying stochastic differential equation is hence required. In choosing such an approximation one should keep in mind that one needs to be able to simulate the

random variables $\tilde{\boldsymbol{Q}}_j$ based on the corresponding approximate distribution. The second requirement is that the essential configuration dependence of the function g should be like that of h (for an appropriate importance sampling). If g depends only on $\boldsymbol{Q}(t_n)$ and g is nonnegative, we may choose $h = g$. Even if the exact transition probabilities are known, the variance can only be reduced to zero if g is either only positive or only negative. In other situations, one can carry out separate simulations for the positive and negative parts of the quantity of interest.[10]

In practice, the short-time transition probabilities are obtained from an approximate integration scheme for the underlying stochastic differential equation. However, all the arguments leading to the rigorous identities (17) and (19) remain valid if the configurations $\boldsymbol{Q}(t_j)$ are replaced by the time-discretized approximate solution $\boldsymbol{Q}_j$ constructed by means of the corresponding integration scheme. In summary, we are now in a position where we can try to design simulations which, although they suffer from the usual systematic errors due to time discretization (linear in time step for the Euler scheme), have smaller statistical error bars. According to the remarks of this paragraph, the development of higher-order variance reduced schemes is possible by using more refined expressions for the small-step transition probabilities occurring in the correction factor.

IV. Example: Hookean Dumbbells

Here we explicitly construct a variance reduced simulation for Hookean dumbbells in the start up of steady shear flow. In order to avoid unnecessarily lengthy equations we measure not only all distances in units of $(k_BT/H)^{1/2}$ but also all times in units of the fundamental dumbbell relaxation time $\lambda_H = \zeta/(4H)$, where ζ is the bead friction coefficient. This corresponds to setting $\lambda_H = k_BT/H = 1$; the appropriate factors can be reintroduced by means of dimensional analysis, and we do that in the final results. For this example, the stochastic differential equation (2) for $d = 3$ has the more explicit form

$$d\boldsymbol{Q}(t) = \left[\boldsymbol{\kappa}\cdot\boldsymbol{Q}(t) - \frac{1}{2}\boldsymbol{Q}(t)\right]dt + d\boldsymbol{W}(t) \tag{21}$$

where the only nonvanishing component of the transposed velocity gradient tensor $\boldsymbol{\kappa}$ is the shear rate, $\dot{\gamma} = \kappa_{12}$.

A. Required Conditional Probabilities. The first basic input required for a variance reduced simulation is the transition probability density

$$p_{app}(\boldsymbol{Q}(t_{max}) = \boldsymbol{q}|\boldsymbol{Q}(0) = \boldsymbol{q}_0) = p_\Theta(\boldsymbol{q} - e^{-t_{max}/2}(1 + \boldsymbol{\kappa}t_{max})\cdot\boldsymbol{q}_0) \tag{22}$$

where the most general centered Gaussian probability density

$$p_\Theta(\boldsymbol{q}) = [(2\pi)^3 \det(\Theta)]^{-1/2} \exp\left\{-\frac{1}{2}\boldsymbol{q}\cdot\Theta^{-1}\cdot\boldsymbol{q}\right\} \tag{23}$$

is determined by the covariance matrix

$$\Theta = (1 - e^{-t_{max}})\,1 + [1 - (1 + t_{max})e^{-t_{max}}](\boldsymbol{\kappa} + \boldsymbol{\kappa}^T) + 2\left[1 - \left(1 + t_{max} + \frac{1}{2}t_{max}^2\right)e^{-t_{max}}\right]\boldsymbol{\kappa}\cdot\boldsymbol{\kappa}^T \tag{24}$$

This transition probability follows from the closed-form solution of the linear stochastic differential equation for the Rouse model. For equilibrium initial conditions at $t_0 = 0$, we obtain the probability density required for the construction of $\tilde{\boldsymbol{Q}}_n$ at $t_n = t_{max}$ from (13),

$$p^f_{app}(\boldsymbol{q}) = p_{\Theta_f}(\boldsymbol{q}) \tag{25}$$

with

$$\Theta_f = 1 + (1 - e^{-t_{max}})(\boldsymbol{\kappa} + \boldsymbol{\kappa}^T) + 2[1 - (1 + t_{max})e^{-t_{max}}]\,\boldsymbol{\kappa}\cdot\boldsymbol{\kappa}^T \tag{26}$$

In other words, the probability density (25) allows us to do the first step of the procedure outlined in the beginning of section III.B. For the second step, we determine the conditional probability density required for the construction of $\tilde{\boldsymbol{Q}}_0$ for given $\tilde{\boldsymbol{Q}}_n$ which is the Gaussian distribution

$$p_{app}(\boldsymbol{Q}(0) = \boldsymbol{q}|\boldsymbol{Q}(t_{max}) = \boldsymbol{q}_n) = p_{\Theta_i}(\boldsymbol{q} - e^{-t_{max}/2}(1 + \boldsymbol{\kappa}^T t_{max})\cdot\Theta_f^{-1}\cdot\boldsymbol{q}_n) \tag{27}$$

with

$$\Theta_i = (1 + \boldsymbol{\kappa}^T t_{max})\cdot\Theta_f^{-1}\cdot\Theta\cdot(1 - \boldsymbol{\kappa}^T t_{max}) \tag{28}$$

The distributions (25) and (27) allow us the simulation of first $\tilde{\boldsymbol{Q}}_n$ and then $\tilde{\boldsymbol{Q}}_0$. These distributions correspond to the exact solution for Hookean dumbbells in the start up of steady shear flow; for small time steps, they deviate slightly from the time-discrete solution which enters the correction factor in (16) through the factors $p(\boldsymbol{Q}_{j+1} = \boldsymbol{q}_{j+1}|\boldsymbol{Q}_j = \boldsymbol{q}_j)$. The variance will hence not be reduced to zero. In nonlinear problems, however, the increase in the variance of the estimator due to the approximate transition probability $p_{app}(\boldsymbol{Q}(t_{max}) = \boldsymbol{q}|\boldsymbol{Q}(0) = \boldsymbol{q}_0)$ is certainly much larger. We once more emphasize that the variance reduction crucially depends on the quality of available approximations. Gaussian approximations are particularly convenient in carrying out variance reduced simulations. If the parameters of the Gaussian distribution are calculated by numerical solution of ordinary differential equations, these equations may be discretized with the same time step as used in the simulations.

For the third and final step of the procedure outlined in the beginning of section III.B we need to describe an iterative procedure for constructing $\tilde{\boldsymbol{Q}}_{j+1}$ for given values of $\tilde{\boldsymbol{Q}}_j$ and $\tilde{\boldsymbol{Q}}_n$ for $j = 0, 1, \ldots, n-2$. Rather than specifying the conditional probability densities $p_{app}(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j, \boldsymbol{Q}(t_n) = \boldsymbol{q}_n)$ we prefer to directly handle the random variables $\tilde{\boldsymbol{Q}}_{j+1}$; this is in the spirit of using random variables instead of their distributions or stochastic differential equations instead of diffusion equations. Loosely speaking, we need to carry out a time step starting at $\tilde{\boldsymbol{Q}}_j$, and we have to keep in mind that the trajectory should end at $\tilde{\boldsymbol{Q}}_n$. In the Rouse model we can calculate the exact distribution of $\tilde{\boldsymbol{Q}}_{j+1}$. Guided by the exact solution, we here suggest a more general procedure,

$$\tilde{\boldsymbol{Q}}_{j+1} = [1 - \epsilon(t_j,t_{j+1},t_n)]\cdot\left\{\tilde{\boldsymbol{Q}}_j + \left[\boldsymbol{\kappa}\cdot\tilde{\boldsymbol{Q}}_j - \frac{1}{2}\tilde{\boldsymbol{Q}}_j\right]\Delta t_j\right\} + \epsilon(t_j,t_{j+1},t_n)\cdot\boldsymbol{\Phi}_{app}(t_{j+1},t_n)\cdot\tilde{\boldsymbol{Q}}_n + ([1 - \epsilon(t_j,t_{j+1},t_n)]\Delta t_j)^{1/2}\cdot\boldsymbol{W}_j \tag{29}$$

for $j = 0, 1, \ldots, n-2$, where $[1 - \epsilon(t_j,t_{j+1},t_n)]^{1/2}$ represents some matrix σ for which $\sigma\cdot\sigma^T = 1 - \epsilon(t_j,t_{j+1},t_n)$. This is a generalization of the Euler scheme (3), which is recovered for $\epsilon(t_j,t_{j+1},t_n) = 0$. The idea behind introducing $\epsilon(t_j,t_{j+1},t_n)$ is to reduce the influence of the drift and diffusion terms when the given final value $\tilde{\boldsymbol{Q}}_n$ is approached and, at the same time, to switch on the influence of the final value. The quantity $\boldsymbol{\Phi}_{app}(t_{j+1},t_n)$ is introduced in order to

appropriately guide the trajectory to its final value. Reasonable forms of these terms can be determined as follows. If in a suitable approximation the expectation at time t_n, denoted by $\boldsymbol{q}_n$, depends linearly on the initial condition $\boldsymbol{q}_{j+1}$ at time t_{j+1}, then we read off the matrix $\Phi_{\text{app}}(t_{j+1},t_n)$ from the linear relationship $\boldsymbol{q}_{j+1} = \Phi_{\text{app}}(t_{j+1},t_n)\cdot\boldsymbol{q}_n$. The quantity $\epsilon(t_j,t_{j+1},t_n)$ can be constructed from the variance of the process at t_n if it starts with a deterministic initial condition at t_{j+1}, denoted by $\Theta(t_{j+1},t_n)$ (it is assumed that the variance $\Theta(t_{j+1},t_n)$ is independent of the initial value, at least to a satisfactory approximation). We then suggest the following choice,

$$\epsilon(t_j,t_{j+1},t_n) = (t_{j+1}-t_j)[(t_{j+1}-t_j)\mathbf{1} + \Phi_{\text{app}}(t_{j+1},t_n)\cdot\Theta(t_{j+1},t_n)\cdot\Phi_{\text{app}}^{\text{T}}(t_{j+1},t_n)]^{-1} \quad (30)$$

where, in general, $(t_{j+1}-t_j)$ should be multiplied by the diffusion tensor which here is $4k_BT/\zeta = k_BT/(H\lambda_H) = 1$ for the units chosen. Note that $\epsilon(t_j,t_{j+1},t_n)$ is small except for the last few time steps for which the denominator, like the numerator, is of the order of Δt_j. This suggests that for constructing the intermediate positions it may be sufficient to have good approximations for short-time transitions rather than for the more complicated conditional probability densities $p_{\text{app}}(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j, \boldsymbol{Q}(t_n) = \boldsymbol{q}_n)$.

In principle, generalizations in which the diffusion tensor depends on $\tilde{\boldsymbol{Q}}_j$ and the auxiliary quantities $\Theta(t_{j+1},t_n)$ and $\Phi_{\text{app}}(t_{j+1},t_n)$ depend on $\tilde{\boldsymbol{Q}}_{j+1}$ are possible. However, if these matrices are independent of configuration, all the coefficients in (29) can be evaluated before the simulation is started, and the variance reduced simulation is not much more time consuming than a standard Euler integration scheme. Notice also that the iteration step of a variance reduced simulation is very similar to what is done in standard stochastic simulations (like Brownian dynamics).

For the Hookean dumbbell problem in shear flow we have

$$\Phi_{\text{app}}(t_{j+1},t_n) = e^{(t_n-t_{j+1})/2}\,[1-\boldsymbol{k}(t_n-t_{j+1})] \quad (31)$$

and

$$\Phi_{\text{app}}(t_{j+1},t_n)\cdot\Theta(t_{j+1},t_n)\cdot\Phi_{\text{app}}^{\text{T}}(t_{j+1},t_n) = (e^{t_n-t_{j+1}}-1)\mathbf{1} + \{e^{t_n-t_{j+1}}[1-(t_n-t_{j+1})]-1\}(\kappa+\kappa^{\text{T}}) + 2\left\{e^{t_n-t_{j+1}}\left[1-(t_n-t_{j+1})+\frac{1}{2}(t_n-t_{j+1})^2\right]-1\right\}\kappa\cdot\kappa^{\text{T}} \quad (32)$$

We now have a complete iterative scheme for variance reduced simulations.

B. Details of the Simulation. Here, some details on the concrete realization of the simulation are given. We wish to reduce the variance of the mean-square dumbbell extension at $t_n = t_{\max}$. In the first step, $\tilde{\boldsymbol{Q}}_n$ is sampled according to the distribution

$$q^2 p_{\Theta_f}(\boldsymbol{q})/\text{Tr}(\Theta_f)$$

obtained from (25) and (26). Realizations $\boldsymbol{q}_n$ of $\tilde{\boldsymbol{Q}}_n$ are constructed from five Gaussian random variables with mean 0 and variance 1, W_j^f, $j = 1, \ldots, 5$, by a suitable transformation. To that end we diagonalize the covariance matrix Θ_f,

$$\Omega_{\Theta_f}^{\text{T}}\cdot\Theta_f\cdot\Omega_{\Theta_f} = \begin{pmatrix} \Theta_f^{(1)} & 0 & 0 \\ 0 & \Theta_f^{(2)} & 0 \\ 0 & 0 & \Theta_f^{(3)} \end{pmatrix}$$

where Ω_{Θ_f} is an orthogonal matrix. We first construct an auxiliary three-dimensional column vector $\mathbf{X}$. With probability $\Theta_f^{(k)}/\text{Tr}(\Theta_f)$ we select one of the three space components k and define $X_k = \text{sgn}(W_1^f)[(W_1^f)^2 + (W_2^f)^2 + (W_3^f)^2]^{1/2}$, and the remaining two components are set equal to W_4^f and W_5^f. It can be verified that

$$\sum_{k=1}^{3}(\Omega_{\Theta_f})_{jk}(\Theta_f^{(k)})^{1/2}X_k$$

constitutes a realization of the components of $\tilde{\boldsymbol{Q}}_n$. The result,

$$p_{\text{app}}^f(\boldsymbol{q}_n) = \frac{1}{[(2\pi)^3\det(\Theta_f)]^{1/2}}\exp\left\{-\frac{1}{2}\sum_{j=1}^{5}(W_j^f)^2\right\} \quad (33)$$

for the realization $\boldsymbol{q}_n$ constructed from the random numbers W_j^f will be useful in evaluating the correction factor f_c.

According to (27), the configuration at $t = 0$ can subsequently be realized as

$$\tilde{\boldsymbol{Q}}_0 = e^{-t_{\max}/2}(1+\kappa^{\text{T}}t_{\max})\cdot\Theta_f^{-1}\cdot\tilde{\boldsymbol{Q}}_n + \Theta_i^{1/2}\cdot\boldsymbol{W}^i, \quad (34)$$

where the three components of $\boldsymbol{W}^i$ are further independent Gaussian random variables and $\Theta_i^{1/2}$ represents some matrix σ with $\sigma\cdot\sigma^{\text{T}} = \Theta_i$. Notice that for the realization $\boldsymbol{q}_0$ of $\tilde{\boldsymbol{Q}}_0$ constructed from the random vector $\boldsymbol{W}^i$ one has

$$p_{\text{app}}(\boldsymbol{Q}(0) = \boldsymbol{q}_0|\boldsymbol{Q}(t_n) = \boldsymbol{q}_n) = \frac{1}{[(2\pi)^3\det(\Theta_i)]^{1/2}}\exp\left\{-\frac{1}{2}(\mathbf{W}^i)^2\right\} \quad (35)$$

Finally, all the intermediate configurations can be constructed from (29), where we assume a constant time step Δt. For these intermediate steps we have

$$p_{\text{app}}(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j, \boldsymbol{Q}(t_n) = \boldsymbol{q}_n) = \frac{1}{[(2\pi\Delta t)^3\det[1-\epsilon(t_j,t_{j+1},t_n)]]^{1/2}}\exp\left\{-\frac{1}{2}\boldsymbol{W}_j^2\right\} \quad (36)$$

for the realization $\boldsymbol{q}_{j+1}$ of $\tilde{\boldsymbol{Q}}_{j+1}$ constructed from the random vector $\boldsymbol{W}_j$ by means of (29).

Finally, we use the Euler approximation (5) for the exact transition probabilities,

$$p(\boldsymbol{Q}(t_{j+1}) = \boldsymbol{q}_{j+1}|\boldsymbol{Q}(t_j) = \boldsymbol{q}_j) = p_{\Delta t}\left(\boldsymbol{q}_{j+1}-\boldsymbol{q}_j-\left[\kappa\cdot\boldsymbol{q}_j-\frac{1}{2}\boldsymbol{q}_j\right]\Delta t\right) \quad (37)$$

By combining the results (33) and (35)–(37), we obtain for the square of the correction factor

$$f_c^2(\tilde{\mathbf{Q}}_0,\tilde{\mathbf{Q}}_1,...,\tilde{\mathbf{Q}}_n) = \frac{\det(\Theta_i)\,\det(\Theta_f)}{(\Delta t)^3} \times \prod_{j=0}^{n-2} \det[1-\epsilon(t_j,t_{j+1},t_n)]\,\exp\left\{\sum_{j=1}^{5}(W_j^f)^2 + (\mathbf{W}^i)^2 + \sum_{j=0}^{n-2}\mathbf{W}_j^2 - \tilde{\mathbf{Q}}_0^2 - \frac{1}{\Delta t}\sum_{j=0}^{n-1}\left[\tilde{\mathbf{Q}}_{j+1} - \tilde{\mathbf{Q}}_j - \left[\kappa\cdot\tilde{\mathbf{Q}}_j - \frac{1}{2}\tilde{\mathbf{Q}}_j\right]\Delta t\right]^2\right\} \quad (38)$$

C. Results. In order to check the variance reduction achieved by the ideas presented above, we have performed direct and variance reduced simulations for Hookean dumbbells in the start up of steady shear flow with $\lambda_H\dot{\gamma} = 2$. The maximum time was chosen as $t_{max} = 5\lambda_H$. In Figure 1, the exact results for the mean-square size and for the polymer contribution to the viscosity are shown (the dumbbell number density used in the Figures is assumed to be n_p). At small t, the time dependence of the viscosity is linear whereas the mean-square size has a quadratic time dependence. The approach to the steady state values is more rapid for the viscosity than for the size. Actually, the exact solution for the discrete time step[11] used in the simulations, $\Delta t = 0.01\lambda_H$, was used for checking the simulation results. The agreement was found to be excellent, within the very small statistical error bars.

Figures 2 and 3 show the results for the statistical error bars in our simulations of 30 000 blocks of 1024 trajectories ($N_T = 30.72 \times 10^6$). Blocks of trajectories were used so that identical operations could be performed on all the trajectories of a block in the innermost loops of the computer program, thus achieving excellent vectorization of the programs on a Cray Y-MP computer. The simulation of a single block of 1024 trajectories over 500 time steps required 0.64 and 0.72 s of CPU time on a Cray Y-MP computer for the direct and variance reduced simulations, respectively.

The dramatic effect of variance reduction for $\langle \mathbf{Q}^2(t)\rangle$ near t_{max}, where the variance of the estimator for $\langle \mathbf{Q}^2(t_{max})\rangle$ was minimized, is obvious in Figure 2. Even in the biased variance reduced simulation, all material properties at all times can be estimated; however, the statistical error bars for short times are larger than in a direct simulation (this is the consequence of an inappropriate bias). For large t_{max}, the variance of an ideal variance reduced estimator ($f_c \to 1$ for $\Delta t \to 0$) for $(H/k_BT)\langle \mathbf{Q}^2(0)\rangle$ is

$$15\left(2c + \frac{1}{c}\right)R_F\left(1, 1 + \frac{\lambda_H\dot{\gamma}}{c}, 1 - \frac{\lambda_H\dot{\gamma}}{c}\right) - 9$$

where $c = [1 + (\lambda_H\dot{\gamma})^2]^{1/2}$ and R_F is Carlson's standard elliptic integral of the first kind (see section 6.11 of ref 12). For $\lambda_H\dot{\gamma} = 2$, this leads to an increase of the error bars by a factor of about 3.5 at $t = 0$ if the variance is optimized at large t_{max}. The factor observed in simulations with $\Delta t = 0.01\lambda_H$ and $t_{max} = 5\lambda_H$ is 3.3, and as one can see in Figure 2, this factor decreases with increasing time.

Even though the variance reduced simulation is designed for one particular quantity at one particular time, the statistical error bars for other quantities may also be reduced. Since the stress tensor is quadratic in the connector vector, we may expect smaller error bars also for the viscometric functions near t_{max} when the variance of the estimator for $\langle \mathbf{Q}^2(t_{max})\rangle$ is minimized. A detailed comparison of the error bars for the polymer contribution to the viscosity, η_p, and the normal stress differences, Ψ_1 and Ψ_2, at $t = t_{max}$ is given in Table 1. Indeed, the error bars for the viscosity and first normal stress coefficient are reduced by factors of 2.0 and 3.7, respectively. The error bar for the second normal stress coefficient is roughly

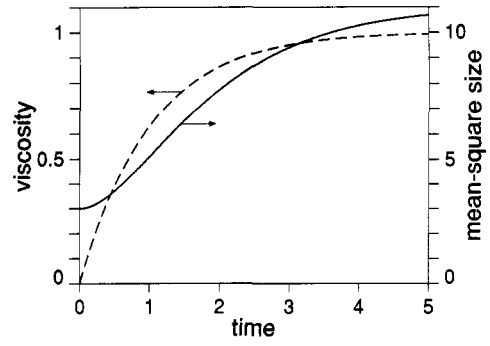

Figure 1. Analytic results for the mean-square size and for the polymer contribution to the viscosity for Hookean dumbbells in the start up of steady shear flow with $\lambda_H\dot{\gamma} = 2$. The units for time, mean-square size, and viscosity are λ_H, k_BT/H, and $n_pk_BT\lambda_H$, respectively.

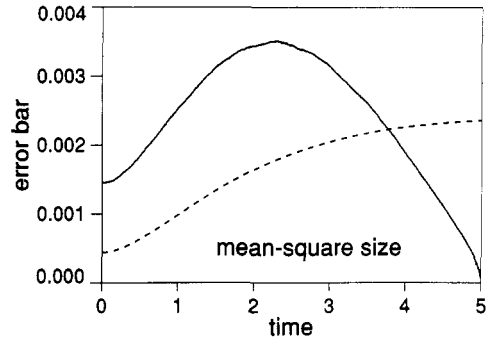

Figure 2. Comparison of the statistical error bars for the mean-square size of a Hookean dumbbell in the start up of steady shear flow with $\lambda_H\dot{\gamma} = 2$ in direct (dashed curve) and variance reduced (continuous curve) simulations. The units for time and error bar are λ_H and k_BT/H, respectively.

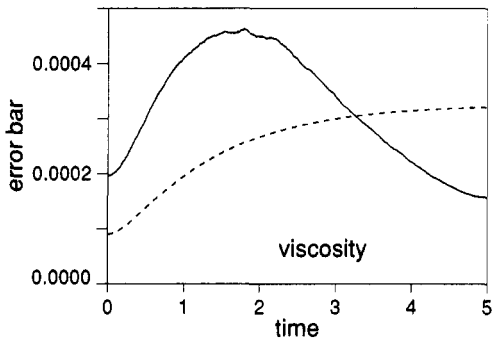

Figure 3. Comparison of the statistical error bars for the polymer contribution to the viscosity for Hookean dumbbells in the start up of steady shear flow with $\lambda_H\dot{\gamma} = 2$ in direct (dashed curve) and variance reduced (continuous curve) simulations. The units for time and error bar are λ_H and $n_pk_BT\lambda_H$, respectively.

the same in the direct and variance reduced simulations. The statistical error bars for the viscosity at all times are compared in Figure 3. They deviate only by a factor of order 2, where the direct simulation is better for short times and the variance reduced simulation is superior near t_{max}.

The variance reduction achieved for $\langle \mathbf{Q}^2(t_{max})\rangle$ corresponds to a decrease of the statistical error bars by a factor of 37. In order to obtain the same error bar as in a direct simulation, a factor of some 1200 in CPU time can be saved (where the longer run time for the variance reduced simulation has been taken into account).

The above construction of variance reduced simulations for Hookean dumbbells uses transition probabilities obtained from the exact solution. In general, approximate expressions for the transition probabilities are required, and Gaussian approximations are particularly convenient

Table 1. Detailed Comparison of Statistical Error Bars for Hookean Dumbbells in the Start Up of Steady Shear Flow at $t_{max} = 5\lambda_H$ with $\lambda_H\dot{\gamma} = 2$

	average	error bar in variance reduced	error bar in direct simulation
$\frac{H}{k_BT}\langle Q^2\rangle$	10.704 941	0.000 064	0.002 361
$\frac{\eta_p}{n_p k_B T \lambda_H}$	0.993 134	0.000 157	0.000 321
$\frac{\Psi_1}{n_p k_B T \lambda_H^2}$	1.924 525	0.000 144	0.000 529
$\frac{\Psi_2}{n_p k_B T \lambda_H^2}$	−0.000 039	0.000 097	0.000 090

for carrying out the simulations. Since there exist excellent approximations for models with hydrodynamic interaction,[13-15] the method is particularly promising for such models. In any case, the idea of using approximations in order to improve simulations is very attractive. It makes the development of good approximation schemes even more rewarding. In addition to a good approximation one can obtain the exact results by means of simulations with smaller statistical error bars; in particular, one can better judge the quality of the approximation by means of the more precise simulation results.

V. Summary and Discussion

We have reviewed how stochastic simulation techniques for kinetic theory models can be developed by rewriting diffusion equations as stochastic differential equations and by then applying numerical integration schemes. The most popular of these simulations are Brownian dynamics simulations for polymers in dilute solutions, but stochastic simulations for reptation models and other systems have also been carried out.

As a new idea, we have presented a basic procedure for constructing more efficient simulations. A reduction of the statistical error bars is achieved by suitably transforming the time evolution of the system, where the construction of the transformation requires an approximate understanding of the dynamics of the system. The development of the transformation is guided by the idea of importance sampling which is also the starting point for Monte-Carlo simulations.

A variance reduced simulation proceeds by first selecting the (biased) final configuration, then the initial configuration, and finally all the intermediate configurations. The construction of the final and initial configurations is particularly simple when the time period of interest is so long that assuming independent initial and final configurations is a good approximation. Then, the initial configuration is sampled according to the given initial distribution. The distribution of the independent final configuration is constructed from an approximate theory. Alternatively, if the solution may be expected to be reasonably well approximated within a given class of distributions, for example Gaussian, then the free parameters can be estimated from a quick preliminary simulation (without variance reduction); even if the error bars are rather large, the introduction of a bias in a reasonable direction may lead to considerably reduced error bars.

After the final and initial configurations are chosen, the construction of variance reduced simulations requires an approximate theory of the conditional distribution of configurations at intermediate times for given previous and final configurations. We suggest a tractable approximation which can be applied in very general situations in order to handle this most complicated building block of a variance reduced simulation. The suggested procedure for constructing intermediate configurations is very similar to numerical integration schemes for stochastic differential equations. For the general stochastic differential equation (2) we suggest a scheme of the form

$$\tilde{\boldsymbol{Q}}_{j+1} = [1 - \epsilon(t_j,t_{j+1},t_n)]\cdot\{\tilde{\boldsymbol{Q}}_j + \boldsymbol{A}(t_j,\tilde{\boldsymbol{Q}}_j)\Delta t_j\} + \epsilon(t_j,t_{j+1},t_n)\cdot\boldsymbol{\Phi}_{app}(t_{j+1},t_n)\cdot\tilde{\boldsymbol{Q}}_n + ([1 - \epsilon(t_j,t_{j+1},t_n)]\Delta t_j)^{1/2}\cdot\boldsymbol{B}(t_j,\tilde{\boldsymbol{Q}}_j)\cdot\boldsymbol{W}_j \quad (39)$$

where the proper choice of the quantities $\epsilon(t_j,t_{j+1},t_n)$ and $\boldsymbol{\Phi}_{app}(t_{j+1},t_n)$ guided by approximate solutions is the crucial task in constructing variance reduced simulations.

Variance reduced simulations are optimized for the evaluation of particular averages. We have shown how other averages can also be evaluated, even averages at different times.

After describing the abstract ideas, we apply them to the simulation of Hookean dumbbells in the start up of steady shear flow. Not surprisingly, dramatic variance reductions can be achieved for this exactly solvable model. Work on variance reduced simulations for Hookean dumbbells with hydrodynamic interaction is in progress. As a first test we have calculated the mean-square size of dumbbells with hydrodynamic interaction in shear flow at $t_{max} = 10\lambda_H$ (the result is then very close to the steady-state value). For our preliminary tests we use the following construction of a variance reduced simulation: the initial and final distributions are assumed to be independent, the final distribution is assumed to be Gaussian with a covariance matrix obtained from a quick direct simulation, and the quantities $\epsilon(t_j,t_{j+1},t_n)$ and $\boldsymbol{\Phi}_{app}(t_{j+1},t_n)$ given in (30)–(32) are introduced into the simulation algorithm for dumbbells in the presence of hydrodynamic interaction without any modification. With this very simple construction, a variance reduction of roughly 2 was achieved; the required computer time is thus reduced by a factor of order 4.

A great deal of further work is needed before variance reduced simulations can be established as a standard tool in polymer kinetic theory. First of all, nontrivial models need to be simulated (in ongoing work, we have chosen hydrodynamic interaction as a crucial test case). The use of non-Gaussian random variables (which can be generated more efficiently) in variance reduced simulations remains to be explored (the handling of the correction factor will then be considerably different). Higher-order schemes (allowing for larger time steps) should be developed. Once the tool of variance reduced simulations is fully developed it should be combined with the finite element method to calculate the flow of viscoelastic liquids, that is via CONNFFESSIT. Then, the required approximate understanding of the dynamics may be obtained from conventional flow calculations so that a fruitful blending of classical flow calculations and CONNFFESSIT might result.

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