

LATTICE BOLTZMANN SIMULATION OF POLYMER-SOLVENT SYSTEMS

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Received (received date)

Revised (revised date)

We investigate a new method for simulating polymer-solvent systems which combines a lattice Boltzmann approach for the fluid with a continuum molecular dynamics (MD) model for the polymer chain. The two parts are coupled by a friction force which is proportional to the difference of the monomer velocity and the fluid velocity at the monomer's position. The strength of the coupling can be tuned by a friction coefficient. Using this approach we examine the dynamics of one monomer immersed in the fluid, and by adding fluctuations to the fluid and the monomer, also the velocity autocorrelation function of one monomer. This results in the definition of an effective friction coefficient for the dynamics of the monomer. Furthermore we analyze the mapping of the model to an MD simulation, allowing to compare results obtained using the new method with MD.

Keywords: Lattice Boltzmann Simulation, Molecular Dynamics, Hydrodynamics, Polymer Solutions

1. Introduction

This paper is intended to describe a new method for simulating polymer-solvent systems by coupling a discrete simulation of the fluid to a continuous description of the polymer system. In using this model, the problem of hydrodynamic interaction in the dynamics of dilute and semi-dilute polymer solutions can be treated. The behavior of a single chain in a solvent, for example, has continuously attracted the attention of MD researchers,¹⁻³ mainly because the analytical theory relies on assumptions⁴⁻⁶ which can be tested using simulations. Furthermore, the problem of hydrodynamic screening in semi-dilute solutions still is a challenge for theory.⁷⁻⁹

Several algorithmic methods are in use for these kinds of problems, including Brownian Dynamics simulations,¹⁰⁻¹³ Molecular Dynamics,¹⁻³ and Dissipative Particle Dynamics.¹⁴ All of them have inherent strengths, but also some disadvantages: The first technique must face the problem that the algorithm scales as the cube of the number of particles, and the latter two simulate the solvent particles explicitly, leading to CPU intensive simulations of several thousand particles even for a single chain of, say, 30 monomers.

In this paper, we want to focus on the really necessary parts only, i.e. the hydro-

dynamics of the solvent and the (Brownian) motion of the polymer chains, thereby trying to keep the computational costs at a minimum. Therefore we simulate the polymer by using a well-established bead-spring continuum MD model,^{2, 15} and the fluid by using the lattice Boltzmann method.^{16–19} In the bead-spring model, the underlying chemistry of the polymer is coarse-grained, leading to an efficient method to simulate the scaling behavior of polymers. Especially for the dynamics it is the method of choice. On the other hand the lattice Boltzmann method offers the possibility of an easy and fast simulation of hydrodynamics, competitive with spectral or finite-difference methods.^{18, 19} Besides, it has the desirable property that fluctuations may be incorporated, which is fundamental for the simulation of Brownian motion.¹⁹ The coupling of the MD part for the polymer and the lattice Boltzmann method for the solvent is done via a friction ansatz, where we assume that the force exerted by the fluid on one monomer is proportional to the difference between the monomer velocity and the fluid velocity at the monomer's position. We are aware that this ansatz is not consistent with the no-slip boundary condition normally used for example in simulations of suspensions,^{19, 20} but on large length and time scales where the shape of the particle is not important, the correct behavior is reproduced. In fact, for the class of problems we are interested in, the relevant length and time scale are the radius of gyration and the Zimm time of the chain, respectively, which are long compared to monomer length and time scales.⁷ This ansatz should thus be sufficient to simulate the scaling behavior of the polymer. Actually, in the over-damped limit for the monomer motion, and the continuum limit for the fluid, our approach is identical to the Oono-Freed equations of motion,²¹ which are commonly used in polymer solution theory.

The remainder of the paper is organized as follows: In Sec. 2 we briefly describe the simulation technique, i.e. the lattice Boltzmann method, the polymer model and the coupling of both. In Sec. 3 we present preliminary tests by applying the method to the case of one monomer in a fluid, both with and without fluctuations. In particular, the method satisfies the fluctuation-dissipation theorem. Furthermore, we can define an effective friction coefficient, which is valuable for the mapping of MD simulations to the new method. We give a short conclusion and an outlook in Sec. 4.

2. Simulation Technique

2.1. *The lattice Boltzmann method*

The lattice Boltzmann method is a discrete method to solve the Boltzmann equation on the computer, leading to the Navier-Stokes equations in the incompressible limit by means of a Chapman-Enskog expansion.²² The central quantity is $n_i(\mathbf{r}, t)$, the number of particles in a volume a^3 at the grid point \mathbf{r} at time t , which have the velocity $\mathbf{c}_i \frac{a}{\tau}$ ($i = 1, \dots, b$), where a is the lattice spacing, τ the time step and \mathbf{c}_i a vector leading to the i th neighbor on a grid with unit lattice constant. The evolution

equation for $n_i(\mathbf{r}, t)$ is the lattice Boltzmann equation¹⁶

$$n_i(\mathbf{r} + \mathbf{c}_i a, t + \tau) = n_i(\mathbf{r}, t) + \sum_{j=1}^b L_{ij} (n_j(\mathbf{r}, t) - n_j^{eq}(\rho, \mathbf{u})). \quad (1)$$

The matrix L_{ij} expresses the scattering between particle population i and j . Its eigenvalues can be determined from physical and numerical arguments, such that its explicit form is not necessary for the simulation algorithm.¹⁹ The local pseudo-equilibrium distribution $n_i^{eq}(\rho, \mathbf{u})$ depends on the density $\rho(\mathbf{r}, t) = \sum_i n_i(\mathbf{r}, t)\mu/a^3$ and fluid current $\mathbf{j}(\mathbf{r}, t) \equiv \rho\mathbf{u} = \sum_i n_i(\mathbf{r}, t)\mathbf{c}_i\mu/(\tau a^2)$ only. Here, μ is the mass of a fluid particle. The usual functional form for $n_i^{eq}(\rho, \mathbf{u})$ is assumed:²³

$$n_i^{eq}(\rho, \mathbf{u}) = \rho \left(A_q + B_q (\mathbf{c}_i \cdot \mathbf{u}) + C_q u^2 + D_q (\mathbf{c}_i \cdot \mathbf{u})^2 \right). \quad (2)$$

The coefficients A_q, B_q, C_q and D_q (which depend on the sub-lattice q , i.e. the magnitude of \mathbf{c}_i) are determined to reproduce the correct macroscopic hydrodynamic behavior. Explicit values are known for different lattices.²³

Here, we implement the 18-velocity model of Ref. 19, which corresponds to the D3Q18 model in the nomenclature of Ref. 23. The set of \mathbf{c}_i consists of the 6 nearest and 12 next-nearest neighbors on a simple cubic lattice. Via a Chapman-Enskog expansion one can show that this model leads to the Navier-Stokes equations in the limit of small Knudsen and low Mach numbers¹⁷, and derive a relation between the kinematic viscosity ν and the non-zero eigenvalue λ of L_{ij} belonging to the eigenvector $c_{i\alpha}c_{i\beta}$, ($\alpha, \beta = x, y, z, \alpha \neq \beta$),

$$\nu = -\frac{1}{6} \left(\frac{2}{\lambda} + 1 \right) \frac{a^2}{\tau}. \quad (3)$$

Fluctuations can be incorporated into the lattice Boltzmann method¹⁹ in the spirit of fluctuating hydrodynamics.²⁴ In order to conserve mass and momentum, one adds the fluctuations to fluxes of the conserved variables, i.e. the stress tensor. The lattice Boltzmann equation is therefore extended to

$$n_i(\mathbf{r} + \mathbf{c}_i a, t + \tau) = n_i(\mathbf{r}, t) + \sum_{j=1}^b L_{ij} (n_j(\mathbf{r}, t) - n_j^{eq}(\rho, \mathbf{u})) + n'_i(\mathbf{r}, t) \quad (4)$$

with the stochastic term

$$n'_i(\mathbf{r}, t) = -D_q \sigma'_{\alpha\beta} c_{i\alpha} c_{i\beta}. \quad (5)$$

The random stress fluctuations σ' are assumed to have white noise behavior

$$\langle \sigma'_{\alpha\beta}(\mathbf{r}, t) \sigma'_{\gamma\delta}(\mathbf{r}', t') \rangle = A \delta_{\mathbf{r}\mathbf{r}'} \delta_{tt'} \left(\delta_{\alpha\gamma} \delta_{\beta\delta} + \delta_{\alpha\delta} \delta_{\beta\gamma} - \frac{2}{3} \delta_{\alpha\beta} \delta_{\gamma\delta} \right). \quad (6)$$

By solving a resulting discrete Langevin equation one can establish the following fluctuation-dissipation relation:¹⁹

$$A = \frac{2\eta k_B T \lambda^2}{a^3 \tau}. \quad (7)$$

2.2. The continuum model for the polymer

The polymer is assumed to be formed of monomers which interact through some potential V . For example, one can use a truncated Lennard-Jones potential for un-bonded and an additional FENE potential for bonded monomers.¹⁵

The equations of motion

$$\frac{d\mathbf{P}_i}{dt} = -\frac{\partial V}{\partial \mathbf{R}_i}, \quad \frac{d\mathbf{R}_i}{dt} = \frac{\mathbf{P}_i}{m} \quad (8)$$

can then be integrated numerically. We have chosen the velocity-Verlet algorithm for this purpose.²⁵ Additionally, the force on the monomer by the surrounding fluid must be taken into account, which is subject of the next section.

2.3. Coupling of monomer and solvent

The simplest approach for coupling the described models for the polymer and the solvent is to treat one monomer as a point particle. In analogy to the Stokes formula for a sphere in a viscous fluid, we assume the force on the monomer exerted by the fluid to be proportional to the difference of the velocity of the monomer \mathbf{V} and the fluid velocity \mathbf{u} at the monomer's position,

$$\mathbf{F}_{fl} = -\zeta [\mathbf{V} - \mathbf{u}(\mathbf{R}, t)]. \quad (9)$$

Here, ζ is a proportionality coefficient which we will refer to as the “bare” friction coefficient. This ansatz has also been used in the simulation of sedimentation.²⁶ In our simulation, we determine $\mathbf{u}(\mathbf{R}, t)$ by linear interpolation of the \mathbf{u} values from the nearest neighbor grid points. The opposite force is exerted on the fluid, i.e. the fluid in the cube a^3 “feels” a force density of $-\mathbf{F}_{fl}/a^3$, resulting in a momentum density exchange per time step of

$$-\mathbf{F}_{fl}/a^3 = \frac{\Delta \mathbf{j}}{\tau} = \sum_{i, \mathbf{r} \in nn} \Delta n_i(\mathbf{r}, t) \mathbf{c}_i \frac{\mu}{a^2 \tau^2}. \quad (10)$$

The last equation has to be satisfied for the change in the number of particles Δn_i of the nearest neighbor grid points (nn) in order to exchange the momentum density $\Delta \mathbf{j}$. Besides, one must also ensure mass conservation in the fluid. Still there is additional freedom for choosing the Δn_i .

We choose a rather simple procedure which involves, at each nearest-neighbor grid point, only the change of one of the n_i per spatial dimension. As an example, consider a particle sitting at (x, y, z) with $0 \leq x, y, z \leq a$. To exchange a certain amount Δj_x , the grid points $a(0, 0, 0)$ and $a(1, 0, 0)$ are updated according to

$$\Delta n_x^{(0,0,0)} = -\Delta n_{-x}^{(1,0,0)} = \frac{1}{2} \Delta j_x \frac{\tau a^2}{\mu} \left(1 - \frac{y}{a}\right) \left(1 - \frac{z}{a}\right), \quad (11)$$

where the index x in Δn_x corresponds to the velocity in $(1,0,0)$ direction, and $-x$ in the opposite direction. The last two factors assign the fraction $(1 - y/a)(1 - z/a)$

of Δj_x to the pair of grid points (linear interpolation). The other pairs and other spatial directions are treated analogously.

As mentioned before, the main drawback of this ansatz is that on small time and length scales it is not consistent with the no-slip boundary condition of rigid objects normally used in hydrodynamic theory. But on larger scales like the radius of gyration and the Zimm time of the polymer, the relaxation between fluid and monomer is established leading to the correct hydrodynamic scaling behavior, as will be seen in the following section. Therefore, by using this simple ansatz it should be possible to simulate the dynamic scaling behavior for polymers in a solvent.

3. Validation of the Method

In what follows we will present simulations of a single monomer coupled to the fluid. The MD part was done, for simplicity, with a time step $\Delta t = \tau$. The monomer mass was chosen $m = \mu$.

3.1. Deterministic experiments

A simple test case for the method is the relaxation of a single particle having an initial velocity $V(0)$, say in x -direction, in the fluid, without any fluctuations. The corresponding velocity $V(t)$ of the particle as a function of time is shown in Figure 1.

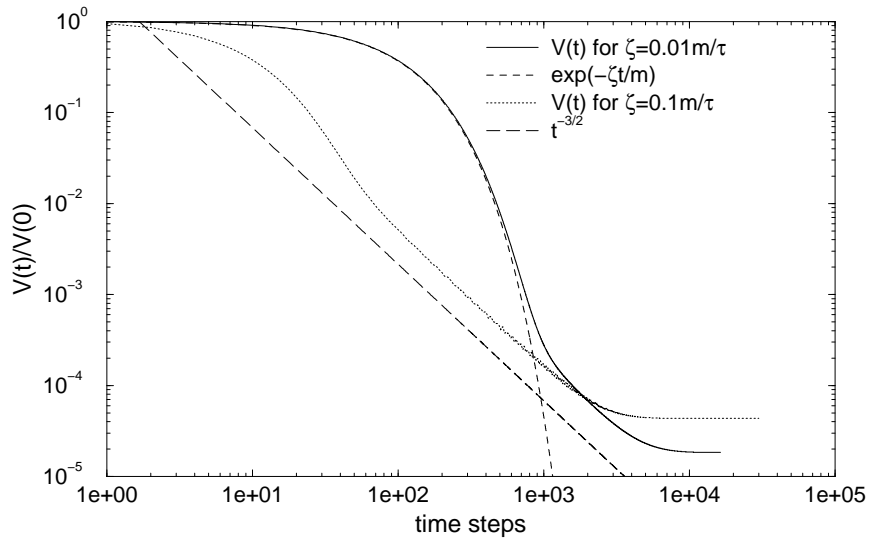


Figure 1: Velocity relaxation of an initially kicked particle in the fluid, simulated at $\lambda = -1.75$, and $\rho = 0.85\mu/a^3$. No fluctuations are added in this case. The expected asymptotic behavior is also shown in the plot.

For $\zeta = 0.01m/\tau$ one sees an exponential initial slope, as expected because the influence of the fluid at short times is negligible. After approx. 1000 time

steps the $t^{-3/2}$ behavior shows up, as predicted by hydrodynamic theory to be the long time behavior of such a system (“long time tail”).^{27, 28} Eventually, the finite size of the simulation box leads to flattening of the curve. Additionally, we also plotted the resulting curve for the higher friction coefficient $\zeta = 0.1m/\tau$, showing the much earlier onset of hydrodynamic behavior. But the problem with higher friction coefficients is a larger discretization error for the propagation of the particle, which is apparent in the jittering of the curve.

3.2. Stochastic experiments

To test the stochastic method we have measured the velocity autocorrelation function for a single particle in the fluctuating fluid. By the fluctuation-dissipation theorem (FDT) the curve should be identical to the deterministic curve of Figure 1 for the same bare friction coefficient. This is not the case, indicating that the simple coupling violates the FDT. This is seen more dramatically in Figure 2, where we plot the temperature of the particle $k_B T = m\langle v^2 \rangle / 3$ for various ζ . The temperature depends on the bare friction coefficient, which shows that the temperature is not well-defined in this case.

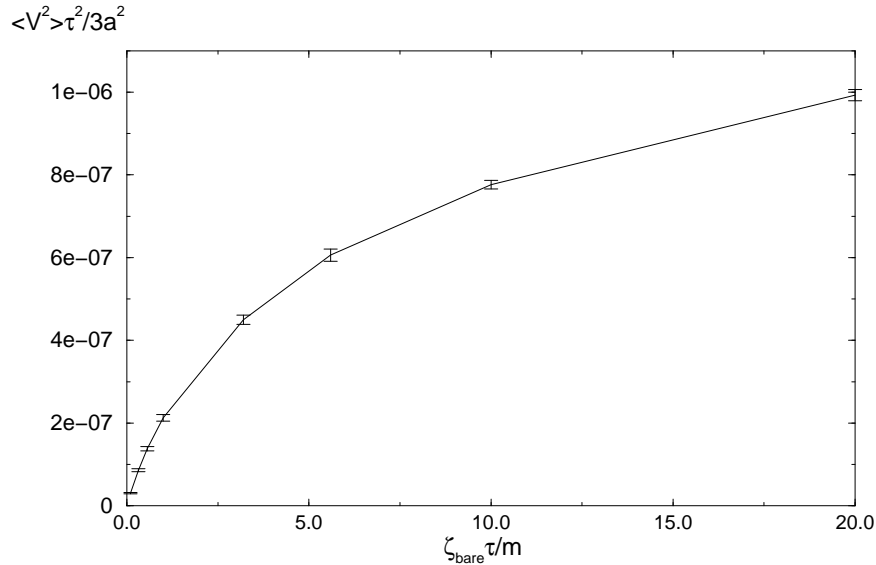


Figure 2: Measured temperature $k_B T = m\langle v^2 \rangle / 3$ of a particle in the fluctuating fluid, simulated at $\lambda = -1.75$, and $\rho = 0.85\mu/a^3$. The fluid had a temperature $k_B T = 10^{-5}\mu a^2/\tau^2$ based on the stochastic lattice Boltzmann approach, Eq. (7). The plot manifests that the temperature is not well-defined using the simple coupling.

The coupling (9), (10) must therefore be refined for stochastic dynamics. We will show by analytical considerations an extension of the coupling that satis-

fies fluctuation-dissipation. In analogy with Ref. 21 we start with the following Langevin-type equations for one particle and the fluid:

$$\frac{d\mathbf{R}}{dt} = \frac{\mathbf{P}}{m} \quad (12)$$

$$\frac{d\mathbf{P}}{dt} = \mathbf{F} - \frac{\zeta}{m} [\mathbf{P} - m\mathbf{u}(\mathbf{R})] + \mathbf{f} \quad (13)$$

$$\begin{aligned} \frac{\partial}{\partial t} \tilde{u}_\lambda(\mathbf{k}) &= -\nu k^2 \tilde{u}_\lambda(\mathbf{k}) + \gamma_\lambda(\mathbf{k}) \\ &+ \frac{1}{M} \left(\frac{\zeta}{m} [\mathbf{P} - m\mathbf{u}(\mathbf{R})] - \mathbf{f} \right) \cdot \mathbf{e}_\lambda(\mathbf{k}) \exp(i\mathbf{k}\mathbf{R}). \end{aligned} \quad (14)$$

The first two equations are the Langevin equations for a Brownian particle of mass m , the last is the Fourier-transformed fluctuating incompressible Navier-Stokes equation, both coupled via a friction term analogous to (9). However, in this continuum framework, the particle acts as a point source on the fluid, which via Fourier transformation yields the last term of Eq. (14). Note that also the stochastic force \mathbf{f} acting on the particle enters this term. $M = \rho V$ is the total mass of the fluid. The index $\lambda = 1, 2$ corresponds to the two transversal modes (the longitudinal is zero due to the assumption of incompressibility), $\mathbf{e}_\lambda(\mathbf{k})$ is a unit vector orthogonal to \mathbf{k} .

Explicitly, the Fourier transformations are written as

$$\mathbf{u}(\mathbf{r}) = \sum_{\mathbf{r}} \sum_{\lambda} \exp(-i\mathbf{k}\mathbf{r}) \mathbf{e}_\lambda(\mathbf{k}) \tilde{u}_\lambda(\mathbf{k}) \quad (15)$$

$$\tilde{u}_\lambda(\mathbf{k}) = \frac{1}{V} \int_V d^3r \exp(i\mathbf{k}\mathbf{r}) \mathbf{e}_\lambda(\mathbf{k}) \mathbf{u}(\mathbf{r}), \quad (16)$$

where $\mathbf{k} = \frac{2\pi}{L} \mathbf{n}$ ($\mathbf{n} \in \mathbb{Z}^3$) only assumes discrete values due to the finite linear extension L of the simulation cell.

For \mathbf{f} and γ , the usual continuum fluctuation-dissipation relations for the *uncoupled* system are assumed:

$$\langle f_\alpha(t) f_\beta(t') \rangle = 2k_B T \zeta \delta(t-t') \delta_{\alpha\beta} \quad (17)$$

$$\langle \gamma_\lambda^*(\mathbf{k}, t) \gamma_\mu(\mathbf{q}, t') \rangle = 2 \frac{k_B T}{M} \nu k^2 \delta_{\mathbf{k}\mathbf{q}} \delta_{\lambda\mu} \delta(t-t'). \quad (18)$$

The Fokker-Planck equation for the system (12)–(14) of stochastic equations can be determined and it can be shown that the equilibrium distribution

$$\mathcal{P} \propto \exp \left(-\frac{P^2}{2mk_B T} - \frac{M}{2k_B T} \sum_{\mathbf{k}, \lambda} |\tilde{u}_\lambda(\mathbf{k})|^2 \right) \quad (19)$$

is a stationary solution of the Fokker-Planck equation. Therefore the FDT holds for (12)–(14).

Concerning the simulation method, these analytical considerations indicate that, for stochastic dynamics, the equations of motion (8), (9) for the monomers should

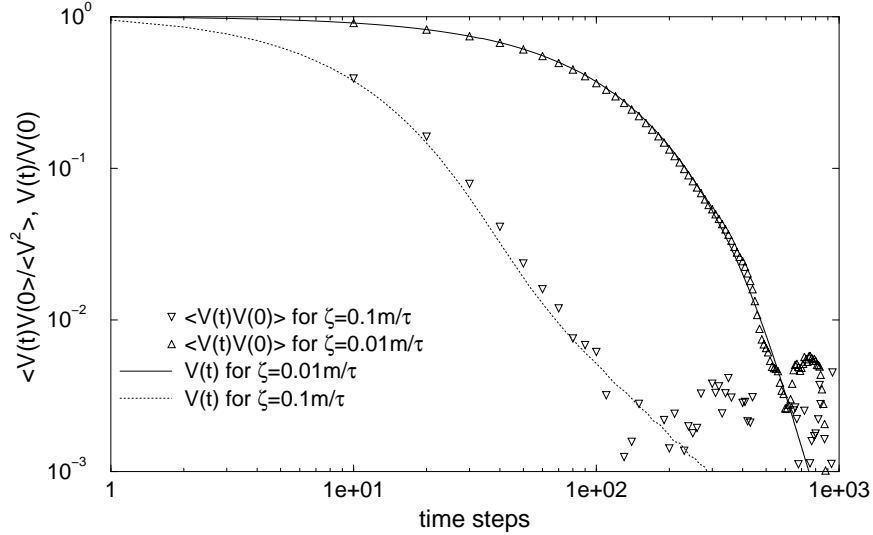


Figure 3: Velocity autocorrelation function compared to velocity relaxation of a kicked particle, simulated at $\lambda = -1.75$, and $\rho = 0.85\mu/a^3$. In the stochastic case $k_B T = 10^{-5}\mu a^2/\tau^2$. The plot shows that the fluctuation-dissipation theorem holds for our simulation.

be extended by the stochastic term \mathbf{f} , and the momentum density exchange in the fluid should include this term also, i.e.

$$\frac{\Delta \mathbf{j}}{\tau} = -(\mathbf{F}_{fl} + \mathbf{f})/a^3. \quad (20)$$

Note that this implies overall momentum conservation of fluid and particle.

Using the modified stochastic equations, the velocity autocorrelation function is identical to the velocity relaxation of the kicked particle (Figure 3), therefore the FDT now holds.

3.3. Mapping to an MD simulation

In order to compare results of the new method with MD simulations using explicit solvent particles, it is desirable to determine the physical input values for the new method from results of MD simulations, i.e. the kinematic viscosity ν , the density ρ , the temperature T and the “bare” friction coefficient ζ must be mapped. ν , ρ and T are readily extracted from an MD simulation, the mapping of ζ is more complicated. We suggest the following method: In the MD simulation the diffusion coefficient D can be measured, either via the mean square displacement or the Green-Kubo relation, respectively,

$$D = \lim_{t \rightarrow \infty} \frac{\langle (\Delta \mathbf{r})^2 \rangle}{6t} = \frac{1}{3} \int_0^\infty \langle \mathbf{v}(t) \cdot \mathbf{v}(0) \rangle dt. \quad (21)$$

The “effective” friction coefficient is then calculated by the Einstein relation $\zeta = k_B T / D$.²⁵

For this purpose, one has to vary ζ over a wide range. Since the discretization error of the MD part is governed by the product $\zeta_{\text{bare}} \tau$, see Eq. (9), large friction coefficients require small time steps. In order to facilitate this, we introduce an “absolute” unit system and measure the time step τ in units of the absolute time unit τ_0 . Inspired by Lennard-Jones MD simulations, where the typical time step is roughly $10^{-2} \tau_{LJ}$ (τ_{LJ} being the natural time scale for these simulations), we choose $\tau = 10^{-2} \tau_0$ for small frictions, and $\tau = 10^{-3} \tau_0$ for larger ones, while keeping ν constant, $\nu = 2.4 a^2 / \tau_0$.

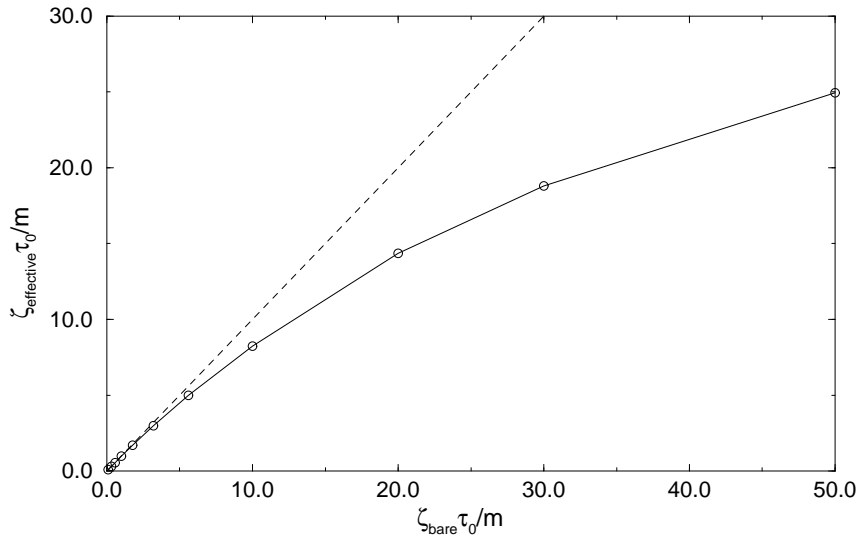


Figure 4: Relation between the bare friction coefficient and the effective friction coefficient. The dashed line corresponds to $\zeta_{\text{effective}} = \zeta_{\text{bare}}$.

We determine the “effective” friction coefficient via the Green-Kubo relation, Eq. (21). As we know that the velocity autocorrelation function is identical to the response velocity function of an initially kicked particle, we can use the deterministic functions of Figure 1 instead of the velocity autocorrelation function, which would need a much larger numerical effort. Performing the integral for several ζ yields a relation between the “bare” and the “effective” ζ which allows to obtain the former (which is the input value we seek) from the latter. The relation between $\zeta_{\text{effective}}$ and ζ_{bare} is shown in Figure 4. To summarize, we can determine the physical input values for the new method from results of MD simulations.

4. Conclusion and Outlook

We have presented a simple simulation method for polymer-solvent systems which

focuses on the essentials to simulate scaling behavior for polymers. The coupling of the monomers to the fluid is done by a simple friction ansatz. We get promising results for single particle properties: The FDT holds and the long time tail is observed. Additionally, we have shown how to map the new method to an MD simulation, allowing to compare results from both.

In the future, the simulation of a single chain in a solvent will be done, and the detailed comparison of the results with an existing MD simulation² by using the described procedure. This also allows to compare the efficiency of the new method against MD. Further optimizations are possible by tuning a and τ with respect to the typical length and time scales of the polymer systems.

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