Reaching large lengths and long times in polymer dynamics simulations

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A lattice model is presented for the simulation of dynamics in polymeric systems. Each polymer is represented as a chain of monomers, residing on a sequence of nearest-neighbor sites of a face-centered-cubic lattice. The polymers are self- and mutually avoiding walks: no lattice site is visited by more than one polymer, nor revisited by the same polymer after leaving it. The dynamics occurs through single-monomer displacements over one lattice spacing. To demonstrate the high computational efficiency of the model, we simulate a dense binary polymer mixture with repelling nearest-neighbor interactions between the two types of polymers, and observe the phase separation over a long period of time. The simulations consist of a total of 46,080 polymers, 100 monomers each, on a lattice with 13,824,000 sites, and an interaction strength of 0.1k_B T. In the final two decades of time, the domain-growth is found to be d(t) \sim t^{1/3}, as expected for a so-called “Model B” system.

I. INTRODUCTION

Lattice models are widely used as a theoretic tool to study polymer solutions. Most notably, Flory [1, 2] and Huggins [3, 4] introduced a mean-field theory of a particular simple model which has proven powerful enough to explain solubility properties and liquid-liquid phase separation of polymer mixtures. An extension to the model, describing the dynamics of such systems, has been introduced by Cahn and Hilliard [5, 6] and Hillert [7]. Some of the earliest computer simulations of lattice polymers were performed in 1962 by Verdier and Stockmayer [8], who studied the dynamics of polymers, modeled by self-avoiding walks on square and cubic lattices. Kolinski et al. [9, 10] added two-monomer moves to the dynamics and simulated a homopolymeric melt.

To describe the polymer dynamics in a melt with more detail than the random-walk approach, Carmesin introduced in 1988 the bond fluctuation model [11]. In the two-dimensional version of this model, each monomer occupies four (2 \times 2) lattice sites of a square lattice. Multiple occupation of lattice sites is not permitted. Monomers adjacent in the chain are connected by bonds with lengths between 2 and \sqrt{13}. The dynamics of the polymer chain consists of the displacement of single monomers to nearest-neighbor lattice sites, restricted by the constraints on bond length and excluded volume. The bond-fluctuation model was extended to three dimensions in 1991, by Deutsch and Binder [12]. This model has been used by several groups [13, 14] to simulate dense polymer melts.

In 1971, de Gennes [15] proposed that the main mechanism of polymer dynamics in gels and dense polymer mixtures is reptation, i.e., diffusion of stored length. To verify the theoretical predictions of de Gennes, Evans and Edwards [16] introduced the cage model. In this model, a single polymer is simulated as a random walk on a square or cubic lattice. In the limit of tight gels, the dynamics reduces to the re-orientation of pairs of connected segments pointing in opposite direction (“kinks”). The cage model has been used for the study of diffusion and relaxation times of a single polymer in a gel [16, 17, 18, 19], as well as for star polymers [20]. Extending the model to include excluded-volume effects or interactions between polymers is not easy.

An alternative model for a single reptating polymer was proposed by Rubinstein [21]. In this model, known as the repton model, the polymer is represented as a chain of monomers, residing on the sites of a square or cubic lattice. Monomers adjacent along the chain reside in either nearest-neighbor lattice sites, or in the same site. The latter case corresponds to the presence of stored length, a central concept in de Gennes’ description of entangled polymer dynamics. The dynamics of the polymer is strictly limited to reptation: the only kind of moves allowed to a monomer in the interior of the chain is that it can move to an adjacent lattice site, provided that one of its adjacent neighbors is already on the site to which it is moving, and the other is in the site it leaves. Per unit of time, each monomer attempts to move in each direction statistically once. The repton model is illustrated in Figure 1. The repton model has been studied extensively by one of us [22, 23].

The repton model can be extended to simulate polymers with self-avoiding walk statistics, as well as many interacting polymers; this is the route followed in this manuscript. In the next section, we discuss the extensions to the repton model which are required to describe equilibrium and dynamical properties of dense polymer solutions and melts. Since the dynamics of polymeric systems occurs on long time-scales and large length-scales, computational efficiency is of paramount importance. We therefore discuss in detail how the resulting extended repton model can be implemented with high computational efficiency, exploiting a technique known as multispin coding. At the end of this manuscript, we show the usefulness of our work by presenting simulation results on the phase separation of a dense binary polymer mixture.
FIG. 1: The repton model on a square lattice. The monomers in the polymer chain are connected by bonds of zero or unit length and each monomer is assigned to a face of the lattice, such that adjacent monomers in the chain occupy either the same or nearest-neighbor lattice sites. If a monomer is connected to one neighbor by a bond with zero length, and to its other neighbor by a bond with unit length, it can join its other neighbor. By doing so, the bond with zero length—a representation of "stored length"—diffuses along the chain. For instance, in the configuration depicted here, monomer 5 can join its neighbor 6. The ends of the polymer allow fluctuations of the total amount of stored length.

II. EXTENSIONS OF THE REPTON MODEL TO SIMULATE POLYMER MELTS

The repton model as proposed by Rubinstein simulates a single polymer obeying random-walk statistics, with dynamics limited to the diffusion of stored length. Two extensions to the repton model are needed to use it for the simulation of entangled chains: the chains have to obey volume exclusion and on the other hand, the constraint that disallows sideways motion has to be lifted.

The repton model describes "stored length" as two or more monomers on the same lattice site. Therefore, we cannot limit the polymers to self-avoiding-walk statistics by disallowing two monomers to occupy the same lattice site, without destroying the reptation dynamics that so successfully described polymer-diffusion in a gel. As reptation is also expected to play an important role in the melt, we should keep this kind of dynamics. The solution to this problem is to limit only the contours of the polymers to self-avoiding-walk statistics. Thus, multiple occupation of a lattice site is allowed only for two or more monomers which are adjacent along the chain. One convenient side-effect of this choice is that since the reptation moves in the interior of the chain do not affect the tube, they never cause a violation of volume exclusion.

Sideways motion is implemented by also allowing monomers to move by a single lattice spacing if this changes the tube of the polymer. Single-monomer moves work well on lattice structures that contain loops of three sites, like the triangular and face-centered-cubic (FCC) lattice; on lattices without such three-site loops, like the square and cubic lattices, the sideways movement can lead to lots of unwanted artifacts or, alternatively, must be implemented by allowing bonds to extend to next-nearest neighbors.

The repton model is often studied in its projected version, in which the state of the polymer is characterized by the bonds $s_i, i = 1 \ldots N - 1$, between the monomers $i = 1 \ldots N$, where $s_i \equiv x_{i+1} - x_i$ is equal to zero or ±1. Usually all allowed moves are attempted with the same (unit) rate, with the consequence that the three values for $s_i$ occur with equal probability. The density of stored length (probability that $s_i$ is zero) is then $1/3$. To obtain the same density of stored length in higher-dimensional repton models (before projection), the rates for moves which decrease the stored length should equal $2/z$ times those of the reverse moves, in which $z$ is the lattice coordination number: 4, 6 and 12 for the square, cubic or triangular, and FCC lattices, respectively. To conserve equilibrium properties, sideways moves which decrease the density of stored length should also be attempted with rates which are $2/z$ times the reverse moves.

Figure 2 shows model-polymers on a triangular lattice. In the upper polymer in the figure, reptation moves are possible for monomers 2, 4, 6, 9, 10, and 11; sideways moves can be made by monomers 2, 4, 6, 8, 9, 10, and 11. Monomers 3, 5, and 7 cannot move. The end-monomers 1 and 12 can move to any empty nearest-neighbor site. In the lower polymer, reptation moves are possible for...
monomers 3, 5, 6, 10, and 11; sideways moves can be made by monomers 3, 4, 5, 6, 7, 10, and 11. Monomers 2, 8, and 9 cannot move. The end-monomer 1 can move to any empty nearest-neighbor site, while end-monomer 12 can only move to the site occupied by monomer 11.

III. MULTISPIN CODING

Multispin coding is a programming technique which makes use of the low-level bit manipulation functions provided by the CPU of our computer, effectively doing many simple computations in parallel. These functions include logical operations like AND (∧), OR (∨) exclusive-or (XOR, ⊕), negation (NOT, ¬), as well as bitwise shifts (≪, integer multiplication by a power of 2; ≫, integer division by a power of 2); and arithmetic operations like addition and multiplication. For a general introduction to multispin coding we refer to Ref. 24, Chapter 15. Here, we will explain this technique analogous to Ref. 24, as applied to the projected repton model. Next we will discuss the implementation of this technique in our model for polymer melts.

A. repton model

In the projected repton model, the state of the chain with monomers \( i = 1 \ldots N \) is specified by the set of their coordinates \( \{x_1, \ldots, x_N\} \). Monomers which are adjacent along the chain have to reside in either the same lattice site, or nearest neighbors, with the consequence that the step \( s_i \equiv x_{i+1} - x_i \) can only take the values \( \pm 1 \) or 0. An alternative way to store the polymer state is therefore to store the coordinate \( x_1 \) of the first monomer, and to store for step \( i = 1 \ldots N - 1 \) two arrays of bits \( (A_i, B_i) \) that can take the combination \( (0, 0) \) if \( s_i = 0 \), \( (0, 1) \) if \( s_i = 1 \), and \( (1, 0) \) if \( s_i = -1 \).

In one unit of time, each monomer attempts to move in each direction statistically once. Thus, \( 2N \) elementary moves, i.e. one monomer attempting to move in one direction, should be attempted per unit of time.

It is a property of the repton model, that if monomer \( i \) in the interior of the chain can join its neighbor \( i + 1 \), it cannot join its neighbor \( i - 1 \), and vice versa. We exploit this by attempting these two elementary moves simultaneously. The first and last monomer can move to both directions, if they are located in the same site as their neighbor along the chain, so this trick does not apply for these two monomers; they are therefore selected with a double probability.

In detail, each monomer in the interior of the chain is selected with a probability \( 1/(N + 2) \). If the selected monomer is located in the same site as exactly one of its two adjacent neighbors, it will join the other neighbor. The bits that contain information on whether a move is possible or not, are \( (A_i, B_i) \) and \( (A_j, B_j) \) with \( j = i - 1 \). A move is possible if one of the two pairs is \( (0, 0) \) and the other is not. If the move is carried out, \( (A_i, B_i) \) and \( (A_j, B_j) \) are exchanged. A sequence of logical operations that achieves this is:

\[
\begin{align*}
y &= (A_i \lor B_i) \oplus (A_j \lor B_j) \\
M_A &= (A_i \lor A_j) \land y \\
M_B &= (B_i \lor B_j) \land y \\
A_i' &= A_i \land M_A \\
B_j' &= B_j \land M_B \\
A_j' &= A_j \land M_A \\
B_i' &= B_i \land M_B
\end{align*}
\]

The first and last monomers are selected, each, with a probability \( 2/(N + 2) \); twice the probability to select a specific monomer in the interior of the chain. After the selection of an end monomer, the intended direction is also randomly selected, with 50% probability to be in the positive or negative direction. The rates at which elementary moves are attempted is thus equal for interior monomers and those at the ends. If the first or last monomer is selected, similar statements suffice to update \( (A_1, B_1) \) or \( (A_{N-1}, B_{N-1}) \), respectively. The following statements can be used to update the end-monomer 1, trying to displace it in the negative or positive direction based on the value of \( r_1 \).

\[
\begin{align*}
A_1' &= \neg (B_1 \lor r_1) \\
B_1' &= (\neg A_1) \land r_1
\end{align*}
\]

However, in case the first monomer is selected, also its coordinate \( x_1 \) needs to be updated. Since this coordinate is an integer number that can take a wide range of values, its update is not implemented in a multispin coding fashion. Luckily, the probability to select the first monomer decreases with increasing polymer length.

The motivation for multispin coding lies in its efficiency. The simulation of the dynamics of 64 polymers involves \( 128N \) elementary moves per time unit. With multispin coding, it involves that the above sequence of logical operations for interior monomers should be performed statistically \( N - 2 \) times, which involves only 11 logical operations, 4 loads, and 4 stores. On a fast workstation, this takes 41 ns of cpu-time, or 0.32 ns per elementary move. For updating the last monomers, even less logical operations suffice. For the first monomers, however, the update of the 64 values of \( x_1 \) cannot be achieved as efficiently, and a loop over all 64 polymers is inevitable, with the consequence that the simulation of the first monomer requires 5.8 ns cpu-time per elementary move.

The multispin implementation outlined above will thus perform 64 simulations in parallel. These simulations are however correlated, since they share the sequence of selected monomers. In fact, if at some point two simulations are in identical polymer configurations, they will stay in identical configurations ever after; in long simulations of small systems, this “locking” will inevitably happen. Complete locking is avoided by an uncorrelated choice in the directions in which end monomers attempt to move, using a 64-bit random bit pattern rather than a binary choice between all-up or all-down. If desired, more
TABLE I: The twelve vectors pointing to nearest neighbors of a FCC lattice, expressed in combinations of $\hat{t}$, $\hat{u}$, $\hat{v}$ and $\hat{w} = -(\hat{t} + \hat{u} + \hat{v})$.

| vector in $\mathbb{Z}^4$ | $\mathbb{Z}^3$ |
|-------------------------|---------------|
| $(-1, 1, 0, 0)$         | $(-1, 1, 0)$  |
| $(0, -1, 1, 0)$         | $(0, -1, 1)$  |
| $(0, 0, -1, 1)$         | $(1, 1, 0)$   |
| $(1, 0, 0, -1)$         | $(0, -1, 1)$  |
| $(-1, 0, 1, 0)$         | $(-1, 0, 1)$  |
| $(0, 1, 0, -1)$         | $(-1, 0, 1)$  |
| $(0, -1, 1, 0)$         | $(1, 0, 1)$   |
| $(1, 0, 1, 0)$          | $(0, 1, 0)$   |
| $(-1, 0, 0, 1)$         | $(-1, 0, 1)$  |
| $(0, 0, 1, -1)$         | $(0, 1, 1)$   |
| $(0, 1, -1, 0)$         | $(0, 1, 0)$   |
| $(1, -1, 0, 0)$         | $(1, -1, 0)$  |

decoupling can be obtained at the expense of a lower efficiency, by rejecting a fraction of the allowed moves, also using a 64-bit random bit pattern. An important remark is, however, that as long as each of the 64 simulations are correct in themselves, one obtains 64 unbiased results; one should just be careful to assign significance to the spread in those 64 simulations.

B. model for polymer melts

While monomers in the projected repton model live on a one-dimensional lattice, the monomers in the model that we propose for polymer melts live on a FCC lattice. It is helpful to note that the three-dimensional hyperplane, located in a four-dimensional hypercubic space through the origin and with perpendicularly oriented vectors $(1, 1, 1, 1)$, is such an FCC lattice. Stated differently, the set of points $\vec{x} = (a, b, c, d)$ with integer-valued coordinates, constrained to $a + b + c + d = 0$, forms a FCC lattice. The twelve vectors pointing to nearest-neighbor sites are $\vec{t} = (-1, 1, 0, 0)$, $\vec{u} = (0, -1, 1, 0)$, $\vec{v} = (0, 0, -1, 1)$, $\vec{w} = -(\vec{t} + \vec{u} + \vec{v}) = (1, 0, 0, -1)$, and some of their combinations, as listed in Table I. The vector $(0, 0, 0, 0)$ is used as the representation for a zero-length bond.

As in the projected repton model, the state of a polymer on our FCC lattice can be specified by the location $\vec{x}_1$ of the first monomer, plus additionally the direction in which adjacent neighbors are located. We choose for the polymer melt model to store the four directional bits in a single word: bits $k$, $k+16$, $k+32$ and $k+48$ of the 64-bit word $D_i$ indicate the vector pointing from monomer $i$ to monomer $i+1$. Thus, sixteen polymers are updated simultaneously.

The coordinates of the $i^{th}$ monomer in polymer $k$ can then be retrieved by summing over all words $D_i$ bits $k$, $k+16$, $k+32$ and $k+48$, yielding respectively the numbers $a$, $b$, $c$ and $d$; the monomer position is then $\vec{x}_i = a\vec{t} + b\vec{u} + c\vec{v} + d\vec{w}$. Note that these summations require only $3i$ operations (a right shift of $k$ bits, masking the direction bits, and adding to the sum), since the summation in the different bits can be done in a single operation. Since in the polymer melt model we need the coordinates frequently (every time we attempt a possible sideways move), and since our polymers are often several hundred monomers long, we do not keep track explicitly of only the position $\vec{x}_1$ of the first monomer, but also of the last monomer as well a few other monomers along the chain, such that the distance along the chain to a monomer with known position is always less than 15. Of course, to retrieve the position $\vec{x}_i$, we start from the nearest monomer with known position in either direction along the polymer.

The implementation proceeds analogous to the projected repton model. Also here, if an interior monomer can move in one direction, its move in the other direction is blocked; this can be exploited as in the repton model, by combining two elementary moves. The precise operations are:

$$y_0 = D_i \lor (D_i \gg 32)$$
$$y_1 = y_0 \lor (y_0 \gg 16)$$
$$z_0 = D_j \lor (D_j \gg 32)$$
$$z_1 = z_0 \lor (z_0 \gg 16)$$
$$m_0 = (y_1 \oplus z_1) \land (2^{16} - 1)$$
$$m_1 = (D_i \oplus D_j) \land (M \ast m_0)$$
$$D'_i = D_i \oplus m_1$$
$$D'_j = D_j \oplus m_1$$

Here, $A \ll k$ stands for shifting the word $A$ over $k$ bits to the left, and $M = 2^0 + 2^1 + 2^2 + 2^3$ is a constant, used to duplicate the low 16 bits in the higher bits of the word. Thus, with only 15 operations, 2 loads, and 2 stores, we have performed 32 elementary moves. On a fast workstation, the above implementation requires 1.25 ns cpu-time per elementary move.

As in the projected repton model, if the first monomers are displaced, whose positions are tracked, these positions have to be updated in a loop over the 16 polymers. We succeeded in implementing moves of the first monomers in 82 ns cpu-time per elementary move. Other monomers whose position is tracked require roughly the same computational effort. Displacement of the first and last monomers is attempted at twice the rate of the other monomers, for the same reason as in the projected repton model.

Besides the reptation moves, the dynamics consists of sideways moves. If a sideways move is attempted on monomer $i$, the relevant bit patterns are those indicating the direction from monomer $i-1$ to $i$ and from $i$ to $i+1$. These bit patterns, as listed in Table II, are all numbers in the range 0 to 15, except for the values 5, 10, and 15 that do not occur; the number 0 denotes stored length, while the other 12 numbers denote bonds to the twelve nearest-neighbor sites. For every combination $(D_i, D_j)$ of those bit patterns, we have pre-computed the lists of all bit pat-
terns $D_i'$ and $D_j'$ after a sideways move. Depending on the combination $(D_i; D_j)$, at most four different sideways moves can be proposed. We therefore have precomputed four such lists for $D_i'$ and for $D_j'$. In one step, we first select randomly the monomer number $i$ and the list number $k$; then we attempt a sideways move of monomer $i$ to the position determined by the $k$th list; finally, if this move does not lead to overlapping monomers, it is accepted. This check for overlap requires computing the position of monomers, which requires computing the distance to the nearest tracked monomer. In our implementation, in which this distance is at most 15 monomers, the total cpu-time required per such step equals 90 ns.

Not all moves are attempted with the same frequency. Since the long-time dynamics is determined by reptation, as argued by de Gennes, the time scale is set such that reptation moves in the interior of the polymers are attempted with unit rate. Sideways moves which do not increase the amount of stored length are attempted with some rate $r_s$; the most natural choice for this rate would be unit rate once more, but because of the much higher computational cost for these moves, we often chose some value of $r_s < 1$. As discussed above, consistency with the density of stored length of the projected repton model demands that moves in which the amount of stored length is increased are attempted with a rate of $2r_s/z$. Since the most mobile monomers hop away to other sites with a total rate of $2r_s$, and since 16 monomers are potentially moved in each step, $r_s/8$ steps as described above should be performed per monomer and per unit of time.

Moves in which the first or last monomer joins its neighbor along the chain can be viewed partly as reptation moves, partly as sideways movement. On these grounds, we have decided to attempt these moves with rate $1 + r_s$. Consistency with respect to the density of stored length requires that moves in which the first or last monomer leaves its neighbor along the chain in a specific direction are attempted with a rate of $2(1 + r_s)/z$.

IV. APPLICATION: PHASE SEPARATION OF A BINARY POLYMER MIXTURE

To illustrate the efficiency of the above computational approach, we performed a simulation of the phase separation of a binary polymer mixture with polymer types $A$ and $B$. The $A$ and $B$ polymers interact with a short-range repulsion, described by the Hamiltonian

$$H = J \sum_{\langle \vec{r}, \vec{r}' \rangle} \delta(\sigma_{\vec{r}}, A) \delta(\sigma_{\vec{r}'}, B) + \delta(\sigma_{\vec{r}}, B) \delta(\sigma_{\vec{r}'}, A),$$

where the summation runs over all pairs of nearest-neighbor sites, and $\sigma(\vec{r})$ is $A$, $B$, or 0 if site $\vec{r}$ is occupied by a polymer of type $A$, $B$ or empty, respectively. The repulsion between $A$ and $B$ polymers provides the driving force for the phase separation.

We simulated a system containing in total 46,080 polymers of length $L = 100$ on a lattice of $N = 13,824,000$ sites, at inverse temperature $\beta J = 0.1$. The system evolves in time through reptation moves, at unit rate, in combination with sideways moves at a rate of $r_s = 1/30$. Figure 3 shows two-dimensional slices of the three-dimensional system at times $t = 0$, $t = 4.7 \cdot 10^5$, and $t = 3.7 \cdot 10^6$ MC time units.

At various times, we determine the two-point correlation function

$$g_{AB}(\vec{r}) = \frac{1}{4} \sum_{\vec{r}} \delta(\sigma_{\vec{r}}, A) \delta(\sigma_{\vec{r} + \vec{r}}, B) \left( \frac{1}{4} \sum_{\vec{r}} \delta(\sigma_{\vec{r}}, A) \right) \left( \frac{1}{4} \sum_{\vec{r}} \delta(\sigma_{\vec{r}}, B) \right).$$

From this function we determine the spherically-averaged radial distribution function, defined as $RDF(r) = 1 - g(\vec{r})$. This function is 1 at $r = 0$, then decreases, and eventually approaches 0 for large $r$. After some time, the conserved dynamics gives rise to damped oscillations in the RDF. The frequency of these oscillations can be determined from the shortest distance $r_0$ at which the RDF equals zero. The typical domain size $d(t)$ is then obtained as twice this distance.
FIG. 4: Domain size of a phase-separating binary polymer mixture as function of the cubic root of the simulation time. The three symbols indicate three independent simulations. A good agreement with domain growth \( d(t) \sim t^{1/3} \) is found.

Figure 4 shows the domain size of the phase-separating mixture at different times. The domain size \( d(t) \), as defined above, is plotted as a function of the cubic root of the simulation time in Monte Carlo sweeps. The straight line shows that the domain size grows with \( d(t) \sim t^{1/3} \). This is to be expected in a system with conserved order dynamics and without hydrodynamics, as in “Model B” [26].

V. SUMMARY AND OUTLOOK

In summary, we have presented a lattice model to study the dynamics of polymeric systems. Compared to other lattice polymer models in current use, this model lends itself for highly efficient simulation: employing multispin coding, the computer time required per elementary move is reduced by more than three orders of magnitude, to a few nanoseconds.

To demonstrate the strength of our model, we simulated the phase separation of a binary mixture of polymers. We performed simulations of a system with about 50,000 polymers, each containing 100 monomers, located on a lattice with about 14 million sites. We verified that over the final two decades in time, the domain size \( d(t) \) grows according to \( d(t) \sim t^{1/3} \), as expected for a system with overdamped dynamics and a local conservation law. This simulation involves \( 3.4 \times 10^{13} \) elementary moves, but could nevertheless be carried out on a single-processor workstation in about 20 days.

Recently, we have used this lattice polymer model to study fractionation in quasi-binary (polydisperse) polymer mixtures [27]. Currently, we are using the model presented here to study the dynamics of absorption of a polydisperse polymer mixture on a surface, as well as the sieving process of polydisperse polymer mixtures through nanopores.

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