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Brownian non-Gaussian diffusion of self-avoiding walks

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Abstract

Three-dimensional Monte Carlo simulations provide a striking confirmation to a recent theoretical prediction: the Brownian non-Gaussian diffusion of critical self-avoiding walks. Although the mean square displacement of the polymer center of mass grows linearly with time (Brownian behavior), the initial probability density function is strongly non-Gaussian and crosses over to Gaussianity only at large time. Full agreement between theory and simulations is achieved without the employment of fitting parameters. We discuss simulation techniques potentially capable of addressing the study of anomalous diffusion under complex conditions like adsorption- or Theta-transition.

Keywords: anomalous diffusion, polymers, critical phenomena

(Some figures may appear in colour only in the online journal)

1. Introduction

A wealth of recent experiments [1–18] and molecular dynamics simulations [19–21] highlighted the existence of diffusion processes in which the mean square displacement linearly grows in time (in agreement with Brownian behavior), but the probability density function (PDF) of the tracer displacement is non-Gaussian during long stages. This has triggered interest for the introduction of novel mesoscopic models relying on superposition of statistics [2, 17, 22, 23], diffusing diffusivities [24–30], continuous time random walk [31–33], and diffusion in disordered environments [34], as well as theoretical approaches based on a clear microscopic foundation [35–38]. A simple microscopic prototype displaying Brownian non-Gaussian diffusion is represented by a tracer undergoing conformational modifications which

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affect its diffusion coefficient. Consider for instance the centre of mass (CM) of a polymer in contact with a chemostat, so that its size N may fluctuate as it does in the grand canonical ensemble. Since the CM diffusion coefficient is a function of the size, size fluctuations naturally generate diffusivity fluctuations and this renders the initial PDF of the CM displacement non-Gaussian (see below). Importantly, the phenomenon becomes evident close to the polymer critical point separating the dilute from the dense phase [39–42], where size fluctuations diverge and the behaviour becomes universal, only depending on the spatial dimension d , on the polymer topology, and the solvent conditions. This state of affairs has been recently theorised in references [36, 37], where explicit predictions about the initial PDF shape have been made based on the critical exponents characterising the polymer universality class.

By exploiting the fact that a microscopic model is endowed with its own independent dynamics, here we consider grand canonical (i.e. N varying) simulations of the three-dimensional ($d = 3$) self-avoiding walks (SAW) on the cubic lattice as a model for a linear polymer fluctuating in size under good solvent conditions [41]. The use of Monte Carlo SAW schemes to simulate polymer dynamics dates back to the work of Baumgartner and Binder [43], where it was implemented the so-called ‘kink-jump dynamics’. Since then, many processes involving polymer dynamics have been simulated via the use of Monte Carlo methods (see for instance references [44, 45]). Although these schemes do not fully mimic Newtonian dynamics—e.g., they disregard inertial effects—in many cases they faithfully describe the diffusion properties of the system and allow for massive statistical sampling with respect to the molecular dynamics counterparts. The algorithm outlined in section 3, based on a variant of the ‘kink-jump’ suitable for SAWs on a cubic lattice, will be shown to correctly reproduce the polymer diffusion dynamics. After reviewing the universality class of three-dimensional SAWs, we address the CM displacements during the simulation dynamics and show a striking agreement between the corresponding PDF and the theoretical predictions reported in references [36, 37]. Importantly, this conformity is achieved without employing any fitting parameter and provides a standalone microscopic validation for the Brownian non-Gaussian character of the CM motion of polymers in proximity of their critical point.

The paper is organised as follows. In the next section we provide the theoretical background for three-dimensional SAWs and for the diffusion properties of their CM. We then supply details of the simulation methods and end the paper by a discussion of our results and future developments.

2. Theoretical background

SAWs are a fundamental mathematical model employed to reproduce the properties of linear polymers in good solvent [41]. Assuming for simplicity the same energy for all SAWs, their equilibrium statistical behaviour in the grand canonical ensemble is described by the partition (or generating) function

$$Z_{\text{gc}}(z) = \sum_n c_n z^n, = \sum_{\omega} z^{|\omega|}, \quad (1)$$

where c_n is the number of distinct n -step SAWs, and z the step fugacity; the second sum is over all SAWs ω , of arbitrary size $|\omega|$. In the grand canonical ensemble, the fugacity $z = e^{\mu/k_B T}$ (where μ is the chemical potential, T temperature, and k_B the Boltzmann constant) corresponds to an intensive thermodynamic variable controlling the interchanges of particles between the system and the chemostat; for SAWs, added and removed particles amount to added and removed steps. It is well known that a model-dependent critical fugacity z_c separates the dilute

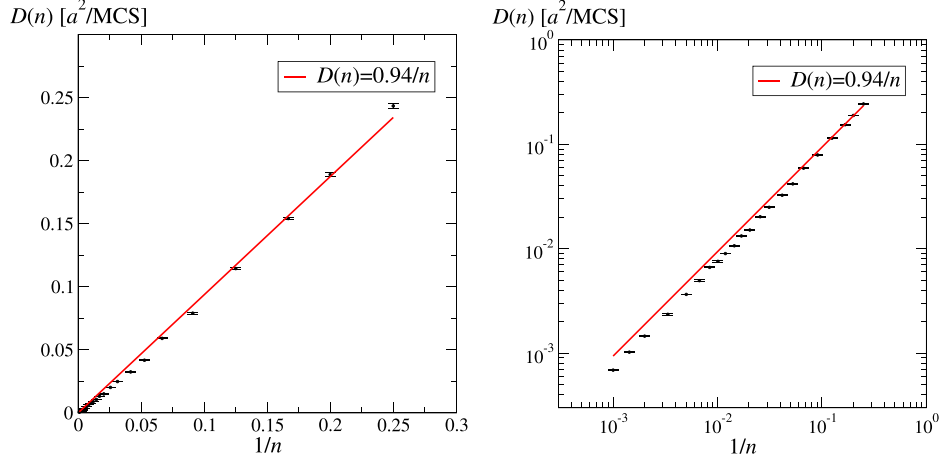


Figure 1. CM diffusion coefficient for fixed size $n \geq 4$ SAWs in a cubic lattice of constant a , both in linear and log–log scale. Symbols: $D(n)$ is measured in units of a^2 over Monte Carlo steps (MCS)—see section 3.1 for the description of n -preserving Monte Carlo moves. Line: null-intercept linear regression yielding $D_m = 0.94 a^2/\text{MCS}$ in equation (5), with regression coefficient $r = 0.9991$.

polymer phase in which the average size $\mathbb{E}[N]$ is finite from a dense one characterized by a divergent $\mathbb{E}[N]$; with a three-dimensional cubic lattice critical fugacity, $z_c \simeq 0.213\,490$ [46]. As $z \rightarrow z_c^-$ the partition function takes the universal form

$$Z_{gc}(z) = \sum_{n=n_{\min}}^{\infty} (z/z_c)^n n^{\gamma-1} = \text{Li}_{1-\gamma}(z/z_c) - \sum_{n=1}^{n_{\min}} (z/z_c)^n n^{\gamma-1} \sim (1 - z/z_c)^{-\gamma}, \quad (2)$$

where γ is the critical entropic exponent whose estimate in $d = 3$ is $\gamma = 1.156\,953\,00(95)$ [47], and $\text{Li}_s(z) \equiv \sum_{n=1}^{\infty} z^n / n^s$ the polylogarithm function. In equation (2) we have assumed a minimal number of steps n_{\min} , which will be discussed below.

If we now consider SAWs of fixed size $N = n \geq n_{\min}$, from one hand equation (2) tells that their equilibrium occurrence is given by the probability distribution

$$P_N^*(n) = \frac{(z/z_c)^n n^{\gamma-1}}{Z_{gc}(z)} \sim (1 - z/z_c)^{\gamma} (z/z_c)^n n^{\gamma-1}. \quad (3)$$

On the other hand, random conformational changes of these SAWs induce a diffusive motion for the position $\mathbf{R}_{CM}(t) = (X_{CM}(t), Y_{CM}(t), Z_{CM}(t))$ of their CM according to

$$d\mathbf{R}_{CM}(t) = \sqrt{2D(n)}d\mathbf{B}(dt), \quad (4)$$

with $\mathbf{B}(dt)$ being a Wiener process (Brownian motion) and $D(n)$ the corresponding diffusion coefficient. As explained in the next section, the stochastic motion of the SAWs is implemented via a kinetic Monte Carlo with local deformations on the lattice. As a consequence, hydrodynamic effects are not taken into account by our dynamics and the size-dependency of the diffusion coefficient is expected to be Rouse-like [48], namely

$$D(n) \simeq \frac{D_m}{n}, \quad (5)$$

for sufficiently large n 's and D_m being an effective single-monomer diffusion coefficient. Figure 1 confirms this expectation.

Coming back to the grand canonical ensemble, size fluctuations may be described in terms of a birth-death process $N(t)$ [36, 37]:

$$\begin{aligned}\partial_t P_N(n, t|n_0) &= \mu P_N(n+1, t|n_0) + \lambda(n-1)P_N(n-1, t|n_0) \\ &\quad - (\mu + \lambda(n))P_N(n, t|n_0) \quad (n > n_{\min}) \\ \partial_t P_N(n_{\min}, t|n_0) &= \mu P_N(n_{\min}+1, t|n_0) - \lambda(n_{\min})P_N(n_{\min}, t|n_0),\end{aligned}\tag{6}$$

where $\lambda(n)$ and μ are respectively the association and dissociation rates. Here, $P_N(n, t|n_0)$ is the probability for $N(t) = n$, given $N(0) = n_0$. Calling τ the autocorrelation time of $N(t)$, the expected asymptotic behaviours for $P_N(n, t|n_0)$ are $P_N(n, t|n_0) \underset{t \ll \tau}{\sim} \delta_{n, n_0}$ and $P_N(n, t|n_0) \underset{t \gg \tau}{\sim} P_N^*(n)$. Note that we have assumed the dissociation rate to be independent of the size n , since it typically represents a decay process occurring at the extremities of the chain. Conversely, the detail balance condition applied to the equilibrium distribution straightforwardly yields [37]

$$\lambda(n) = \mu \frac{P_N^*(n+1)}{P_N^*(n)} = \mu \frac{z}{z_c} \left(\frac{n+1}{n} \right)^{\gamma-1}.\tag{7}$$

In what follows μ , with the dimension of an inverse time, can be regarded as a free parameter which may be tuned to match the model with the autocorrelation τ of real physical conditions [37].

A fluctuating $N(t)$ adds a second source of randomness to equation (4); in the literature, $N(t)$ is called the *subordinator* and $\mathbf{B}(t)$ the *subordinated* process [49, 50]. It is convenient to reparametrize the diffusion path in terms of the coordinate $s \geq 0$, $ds = 2D(n(t))dt$, corresponding to the realization of the stochastic process

$$S(t) \equiv 2 \int_0^t dt' D(N(t')) = 2D_m \int_0^t dt' N^{-1}(t'),\tag{8}$$

with PDF $p_S(s, t|n_0)$. The conditional PDF for the CM displacement $\mathbf{R}_{\text{CM}} = \mathbf{r}$ at time t given the initial conditions $N = n_0$, $\mathbf{R}_{\text{CM}} = \mathbf{0}$ at time 0, $p_{\mathbf{R}_{\text{CM}}}(\mathbf{r}, t|n_0; \mathbf{0})$, is then obtained through the subordination formula [25, 36, 37]

$$p_{\mathbf{R}_{\text{CM}}}(\mathbf{r}, t|n_0; \mathbf{0}) = \int_0^\infty ds \frac{e^{-\frac{\mathbf{r}^2}{2s}}}{(2\pi s)^{3/2}} p_S(s, t|n_0).\tag{9}$$

Taking an equilibrium distribution $P_N^*(n_0)$ for the initial sizes of the SAW, from equation (9) one gets the Brownian character of the CM diffusion:

$$\mathbb{E}[\mathbf{R}_{\text{CM}}^2(t)] = 3\mathbb{E}[S(t)] = 6D_{\text{av}}t,\tag{10}$$

with $D_{\text{av}} \equiv \sum_{n=n_{\min}}^\infty \frac{D_m}{n} P_N^*(n)$. The shape of the initial non-Gaussian PDF for the polymer CM is instead conveniently studied by switching to the unit-variance dimensionless variable $\bar{X}_{\text{CM}}(t) \equiv X_{\text{CM}}(t)/\sqrt{\mathbb{E}[X_{\text{CM}}^2(t)]}$. From equation (9) and the asymptotic limits of the birth-death process, as $t \rightarrow 0^+$ we have

$$p_{\bar{X}_{\text{CM}}}(x, 0^+) \simeq \sum_{n=n_{\min}}^\infty P_N^*(n) \frac{e^{-\frac{\mathbb{E}[N^{-1}]}{2} x^2}}{\sqrt{2\pi \frac{n^{-1}}{\mathbb{E}[N^{-1}]}}}.\tag{11}$$

At large $|x|$ the PDF is asymptotic to the Gaussian cutoff $\sim e^{-\mathbb{E}[N^{-1}]x^2/(2n_{\min}^{-1})}$, and as $z/z_c \rightarrow 1$ this cutoff is pushed towards $|x| \rightarrow \infty$, since $\mathbb{E}[N^{-1}] \rightarrow 0$. Conversely, in the long time limit the ordinary Gaussian behavior is restored, $p_{\bar{X}_{\text{CM}}}(x, \infty) \sim e^{-x^2/2}$. The autocorrelation τ is the time scale separating the two behaviours; as $z \rightarrow z_c^-$ this autocorrelation time diverges [37] (critical slowing down).

3. Simulation methods

On-lattice models are a schematic representation of actual polymers, allowing for massive data sampling. In the present simplification in which all configurations share the same energy, they are particularly effective in reproducing the equilibrium statistics under detail balance conditions. Since we want to sample SAWs in the grand canonical ensemble, we apply a Monte Carlo method based on a variation of the Berretti-Sokal algorithm [51], perhaps the simplest variable-length dynamic Monte Carlo method whose state space is the set of all SAWs (i.e. SAWs on any lengths) in a cubic lattice. The basic idea of the original implementation is that, at each iteration, one of the two ends of the chain is randomly chosen; then, with probability $P_N(n-1|n)$ the last step of the walk is deleted (deletion or -1 move), while with probability $P_N(n+1|n) = 1 - P_N(n-1|n)$ an attempt is made to increase the SAWs length by appending a new edge with equal probability in each of the $2d = 6$ possible directions ($+1$ move). In the latter case, if the new configuration is not self-avoiding the proposed move is rejected and the old configuration is counted again in the sampling (null transition). A null transition is also made if a deletion move is attempted on a walk with $n = n_{\min}$ steps. To assure that the invariant probability distribution of a walk is correctly given by $P_{\Omega}^*(\omega) = z^{|\omega|}/Z_{\text{gc}}(z)$, the transition probabilities for the ± 1 moves are taken according to [51]

$$P_N(n+1|n) = \frac{2dz}{1+2dz}, \quad P_N(n-1|n) = \frac{1}{1+2dz}. \quad (12)$$

The implementation of this algorithm recovers the estimates of z_c and γ given above; in particular, figure 2 reports a very good agreement between the equilibrium size distribution sampled with the Monte Carlo strategy and equation (3). Grand canonical Monte Carlo simulations reported in figure 2 and in the following ones are performed at $z/z_c = 0.997677$; evaluation of the autocorrelation time for this fugacity returns $\tau \simeq \times 10^6$ MCS.

3.1. Motion of the CM

In order to reproduce the diffusion dynamics of a polymer, the algorithm is enriched by adding three types of n -preserving local moves as shown in figure 3. These are: (i) the one-vertex flip or kink move; (ii) the 90° rotation of end steps; (iii) the 90° crankshaft move [52]. In all cases, if the move leads to a violation of self-avoidance, the attempted move is rejected. For a SAW of size n , each MCS consists of one attempted Berretti-Sokal move followed by a sequence of n attempts of moves of the kind (i)–(iii) with a random choice of the location and of the specific n -preserving move. Since moves of kind (iii) attempt to change the position of two vertices in place of that of a single one, they are proposed with half the probability of moves (i), (ii). Note that for the whole set of three local moves to be potentially applicable, a minimal number of three steps is required; in our simulations we then set $n_{\min} = 4$ to have ensured proper mobility at all sizes. This minimal size is still sufficiently small to guarantee the ergodicity of the algorithm [51]. Ergodicity is in fact granted once it is assured the possibility of passing from any n -step SAW to any other n' -step SAW within a finite number of moves ($n_{\min} \leq n, n' < \infty$). In view of the reversibility of the moves, this is equivalent to the capability

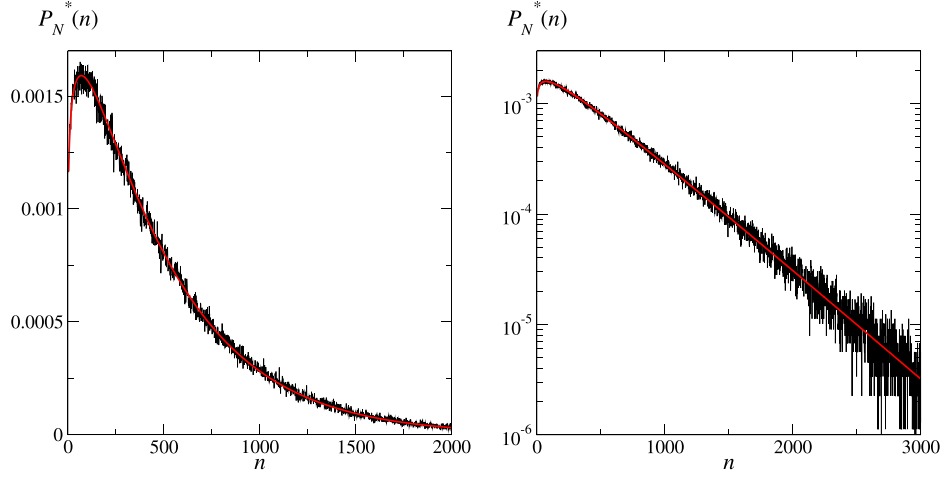


Figure 2. Comparison between the Monte Carlo estimation for $P_N^*(n)$ (black line) and equation (3) (red line), both in linear and log-linear plot.

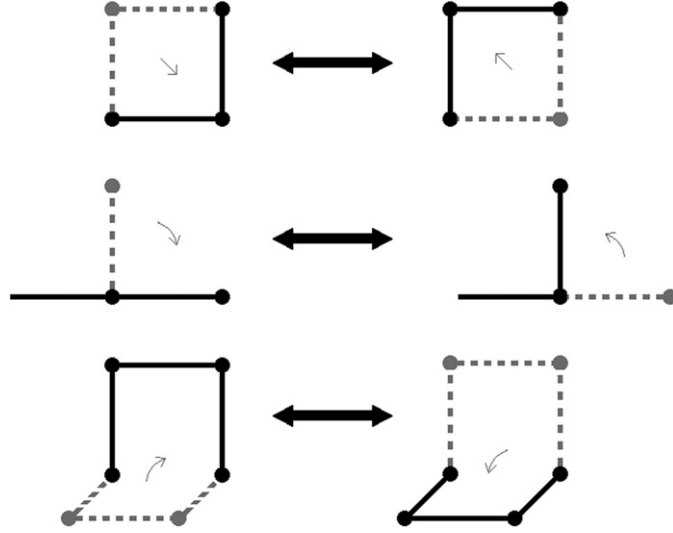


Figure 3. n -preserving Monte Carlo moves. For each of the three kind of moves, two reciprocal realizations are plotted.

of reaching a specific target (say, e.g., a four-step SAW completely straighten along the x direction) from any n -step SAW. It is easy to figure out a possible finite sequence of moves performing the last task as follows: first, the application of $n - 4$ moves of the kind -1 reaches one of the 726 [53, 54] distinct four-step SAWs occurring on the cubic lattice; then, within a finite sequence of n -preserving moves the target is certainly reached. In all our simulations for the motion of CM we start from a grand canonical equilibrium configuration.

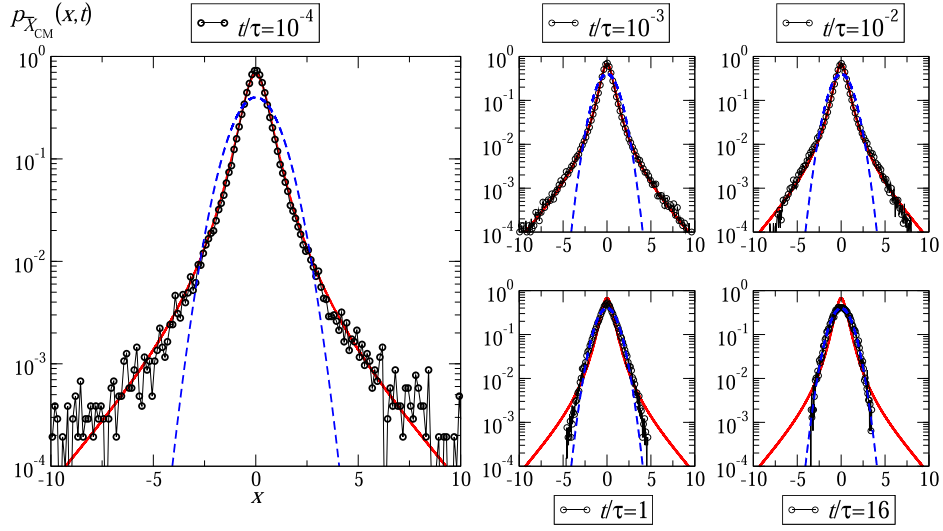


Figure 4. Non-Gaussian diffusion of SAWs. Exact theoretical predictions for the PDF of the unit-variance dimensionless variable $\bar{X}_{\text{CM}}(t) \equiv X_{\text{CM}}(t)/\sqrt{\mathbb{E}[X_{\text{CM}}^2(t)]}$ (curves) are compared with normalized displacement histograms obtained from Monte Carlo simulations (circles). With $t/\tau \leq 10^{-2}$, histograms are built with 4.9×10^5 realizations; otherwise with 6.6×10^4 realizations. Full red lines are given by equation (11); dashed blue lines reproduce a zero-mean unit-variance Gaussian.

Eventually, it is interesting to point out that the correct diffusive properties for our polymer model are reproduced even neglecting thermal fluctuations induced by the interaction with the solvent, which might excite translational and rotational degrees of freedom of the polymer. Such a simplification, enabling a larger statistical sampling, is in line with kinetic Monte Carlo standards [43–45], and is ultimately allowed by the fact that the overdamped motion of the CM is in fact an attractor for a wide class of dynamical models.

4. Discussion

According to equation (11) the shape of the initial PDF $p_{\bar{X}_{\text{CM}}}(x, 0^+)$ is universal close to criticality, being identified by the value of the fugacity, by γ , and by the exponent of the D vs n relation (-1 in the present case). The only remaining detail is n_{min} , fixed equal to 4 in our simulations. Hence, no fitting parameter are left free. Figure 4 highlights a striking agreement between the universal predictions and the normalized histograms for $p_{\bar{X}_{\text{CM}}}(x, t)$ obtained from the Monte Carlo simulations. Initially, $p_{\bar{X}_{\text{CM}}}$ sticks close to the PDF of the anomalous fixed point $z = z_c$ (characterized in the present universality class by an infinite kurtosis [37]), and only with $t \simeq 10^{-2}\tau$ some deviation develops at probability values of the order of 10^{-4} . As $t \simeq \tau$, $p_{\bar{X}_{\text{CM}}}$ crosses over the (trivial) Gaussian fixed-point. We remind the reader that, as $z \rightarrow z_c^-$, τ becomes infinite. Although we concentrate here on the short- and long-time limit in which analytical predictions are handy, using the Gillespie algorithm [55] for the process $N(t)$ in combination with an ordinary Wiener process it is also possible to simulate equation (4) and obtain thus the theoretical full time evolution of $p_{\bar{X}_{\text{CM}}}(x, t)$.

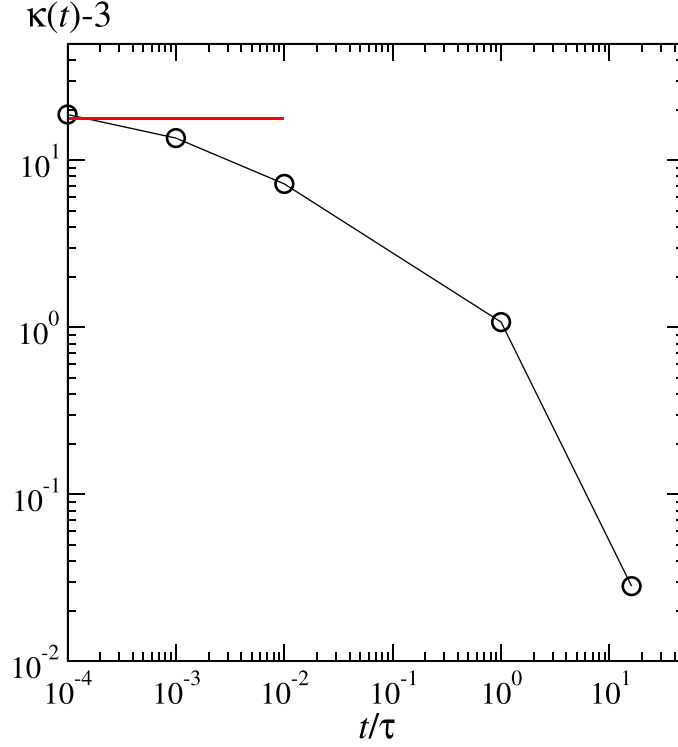


Figure 5. Time evolution for the excess kurtosis in Monte Carlo simulations (circles). The horizontal line represents the theoretical asymptotic behaviour, according to equation (11).

The crossover behaviour is made evident in figure 5, where it is reported the time evolution of the excess kurtosis,

$$\kappa(t) - 3 \equiv \frac{\mathbb{E}[(\bar{X}_{\text{CM}} - \mathbb{E}[\bar{X}_{\text{CM}}])^4]}{(\mathbb{E}[(\bar{X}_{\text{CM}} - \mathbb{E}[\bar{X}_{\text{CM}}])^2])^2} - 3 = \frac{\mathbb{E}[\bar{X}_{\text{CM}}^4]}{(\mathbb{E}[\bar{X}_{\text{CM}}^2])^2} - 3 \quad (13)$$

according to the Monte Carlo simulations. Despite the large sensitivity of the fourth moment of the PDF to fluctuations in the tails, figure 5 confirms a substantial agreement between simulations and theoretical predictions as $t \rightarrow 0^-$, and highlights that the excess kurtosis tends to zero (Gaussian PDF) above the autocorrelation time scale.

Stochastic models are typically constructed assuming specific features for noise terms in the equations. Here we followed a different path, in which the model begets from a genuine microscopic instance endowed with an autonomous dynamics. We have thus been able to provide the first evidence in which the Brownian non-Gaussian diffusion compliant with a stochastic model is independently and exactly produced by a microscopic dynamics. Noting that the anomalous diffusion of the polymer CM is ruled by its equilibrium critical behaviour, and ultimately by the value of the entropic critical exponent γ , it would be worthwhile to extend future theoretical and numerical considerations to models of polymers either characterized by different, fixed, topologies [56] (i.e. linear vs star or branched polymer) or to polymeric systems

still in diluted condition but undergoing conformational phase transitions such as the Theta- or the adsorption-transition [39, 41]. For instance, one should compare the theoretical predictions of the CM diffusion of linear polymers undergoing a collapse transition from a swollen to a compact phase with gran canonical (i.e. N -varying ensemble) Monte Carlo simulations of interacting SAW on the cubic lattice [41]. Similarly, one might look at the CM diffusion of three-dimensional SAWs once adsorbed on an attractive impenetrable wall [57] or localised in proximity of an interface separating two bulk regions characterized by different solvent conditions [58, 59].

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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