Diffusion in the two-dimensional necklace model for reptation

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An extension of a recently introduced one-dimensional model, the necklace model, is used to study the reptation of a chain of N particles in a two-dimensional square lattice. The mobilities of end and middle particles of a chain are governed by three free parameters. This new model mimics the behavior of a long linear and flexible polymer in a gel. Noninteracting and self-avoiding chains are considered. For both cases, analytical approximations for the diffusion coefficient of the center of mass of the chain, for all values of N, are proposed. The validity of these approximations for different values of the free parameters is verified by means of Monte Carlo simulations. Extensions to higher dimensions are also discussed.

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I. INTRODUCTION

The study of diffusion and transport properties of polymers using reptation models has attracted a great of interest in recent years (see, e.g. [1–4]). The reptation mechanism, originally introduced by de Gennes [5], describes the dynamics of entangled polymer melts and also the motion of DNA molecules in gel electrophoresis. In both cases a long linear and flexible polymer moves in a medium of dense obstacles that confine its motion to a one-dimensional diffusion along a tube [1–3]. Thus, a chain can progress by leaving part of the initial tube and creating a new part as it moves.

The first one-dimensional discretized model to analyze the chain dynamics under reptation was introduced by Rubinstein (the repton model) [6]. This model consists of N random walkers (the reptons, the basic units of the chain) in one dimension. The reptons move in such a way as to not break the connectivity of the cluster. A site in the middle of the chain cannot be vacated and the original order of the reptons is preserved.

The repton model was adapted by Duke to study electrophoresis of DNA chains in a gel [3,7,8]. The gel was pictured by Duke as a square lattice of cells and the DNA molecule was considered to be a flexible repton chain that moves from cell to cell. Reptons can hop to an adjacent cell without violating the connectivity and the number of reptons that each cell can accommodate is unlimited. On the other hand, self-avoidance is not considered.

Later, a slightly modified repton model in one dimension was introduced [9]. We will refer this model as the one-dimensional *necklace model*. Although at first glance the rules of this model look different than those of the repton model, their diffusion dynamics are identical. Recently, the exact analytical expressions for the diffusion coefficient and the drift velocity for the one-dimensional necklace model (and then for the original repton model) were obtained [10–13]. At odds with the repton model, the necklace model includes hardcore interactions between the reptons. Also, the necklace model is more flexible regarding the jumping probabilities of reptons at the ends of the chains relative to those for the central ones, allowing the study of cases that cannot be addressed with the original repton model.

In the present work, we study the reptation of a chain in a square lattice using an extension of the recently introduced one-dimensional necklace model. As for one-dimension, the necklace model in two dimensions differs with Duke's model in that reptons cannot occupy the same cell. With the necklace model we can introduce two alternatives, namely the noninteracting and the self-avoiding cases [14]. Also, jumping probabilities of reptons at the end of the chains are allowed to adopt values that can be larger or smaller than those of the central ones. We propose analytical approximations for the chain diffusion constant for both cases, which are in agreement with Monte Carlo results. We also discuss the extension of these approximations to higher dimensions.

II. MODELS

As it was mentioned, we will consider two different cases with noninteracting and with self-avoiding chains. We begin defining the two-dimensional noninteracting necklace model. The gel fibers (or other molten polymers), which play the role of obstacles, are represented by crosses on a square lattice of a lattice constant a [see Fig. 1(a)]. The diffusing chain, represented by a string of beads (or particles) and holes (or vacancies), is placed among the obstacles in a second square lattice shifted by a distance $a/2$, in the x and y directions, from the first lattice. In what follows we will only consider the second square lattice. The distance between two consecutive particles can be either a, $2^{1/2}a$, or $2a$ (in the last two cases there is a hole between the particles).

Only loops of the string in which at least one obstacle is surrounded by the chain are allowed. Although consecutive beads or holes are not allowed to occupy the same site, when loops form a given lattice site can be occupied by more than one particle or hole. This takes into account the noninteracting character of the chain. For more details, see Fig. 2.

The number of particles in the chain is denoted by N. The chain has two end particles and $N-2$ middle particles (particles which are not located at the end of the chain). The

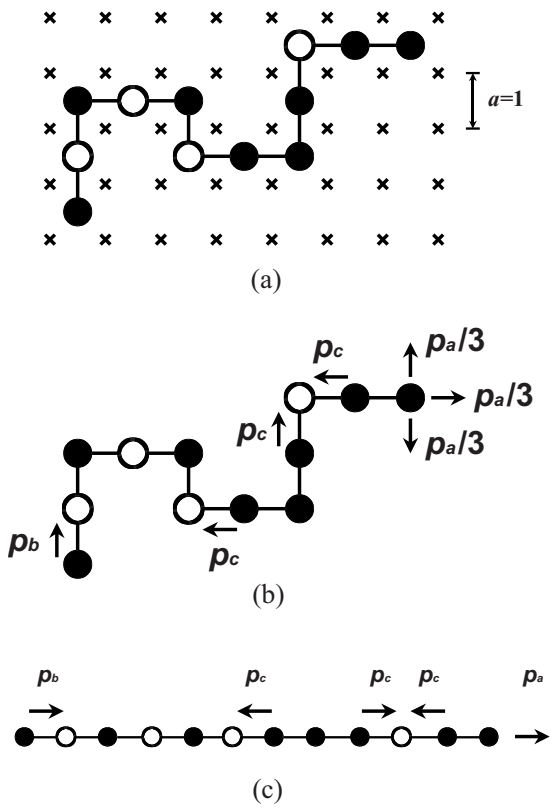

FIG. 1. Scheme representing the two-dimensional necklace model. (a) Chain in a melt of other polymers chains or in a gel. Filled circles represent particles of the chain, open circles represent holes of the chain, and crosses represent other polymers or gel fibers. (b) Jumping probabilities per unit time for end and middle particles. Particles without arrows cannot jump. (c) One-dimensional projection of the configuration in (b).

number of holes can vary from 0 to $N-1$. Each end particle has a corresponding pre-end particle, that is, the consecutive particle of this end particle along the chain.

The jumping rules of the model are as follows.

(i) An end particle with a nearest site occupied by its corresponding pre-end particle jumps with a probability per unit time $p_a/3$ to each of the three nearest sites that are not occupied by the pre-end particle; see Fig. 1(b). Then, the total jumping probability per unit time is p_a. If the jump takes place, a hole is created.

(ii) An end particle not having a nearest site occupied by its corresponding pre-end particle (i.e., there is a hole between these two particles) jumps towards the hole with a probability per unit time p_b. If the jump takes place, a hole is annihilated.

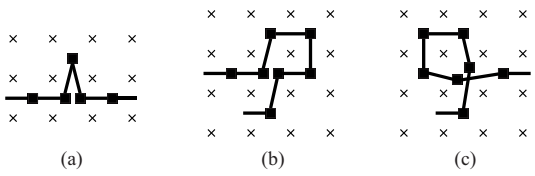

FIG. 2. Chain configurations. Square represent particles or holes of the chain. For the noninteracting and self-avoiding cases the configuration (a) is forbidden. Configurations (b) and (c) are allowed for the noninteracting case and they are forbidden for the self-avoiding case.

(iii) A middle particle with one of its nearest site along the chain occupied and the other one empty jumps to the hole with a probability per unit time p_c.

(iv) A middle particle with both nearest sites along the chain occupied, or both nearest sites along the chain empty (i.e., a middle particle between two holes), does not jump and remains at its original position.

Hence, p_a, p_b, and p_c are the free parameters of the model. In the following we will use that the distance a between adjacent sites of the square lattice and the unit time are both equal to 1.

A hole is created or annihilated every time an end particle jumps moving away from the chain or towards the chain, respectively. An end particle jumping attempt that creates a

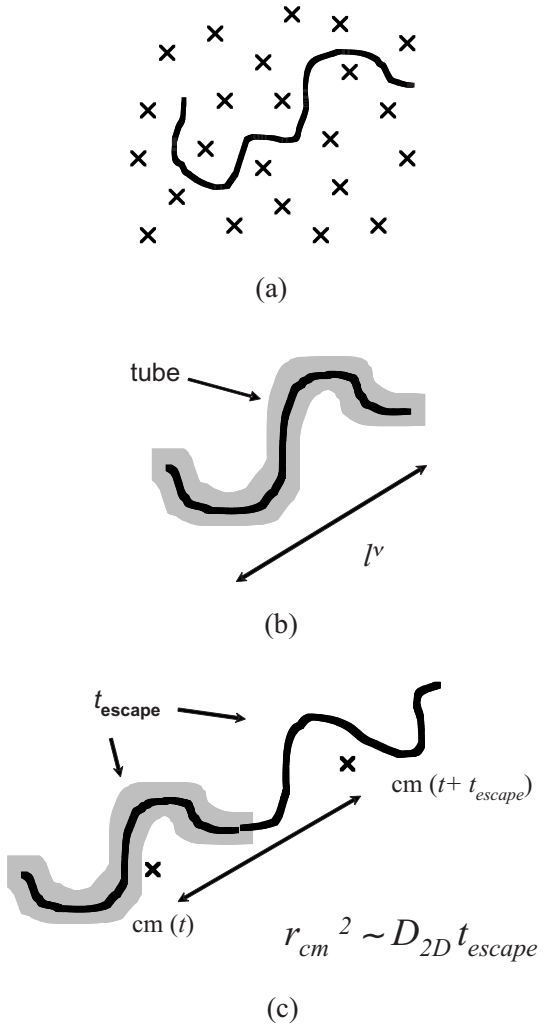

FIG. 3. Reptation mechanism. (a) The chain in a melt of other polymer chains or in a gel. (b) Scheme for the polymer chain within its tube at one instant, l^v is the end-to-end distance of the chain. (c) Polymer chain at one instant and at a later time when it has diffused out of its original tube. Crosses represent the position of the center of mass. (Strictly, the tube is continuously modified when the chain moves leaving part of the intitial tube and creating a new part as it diffuses.)

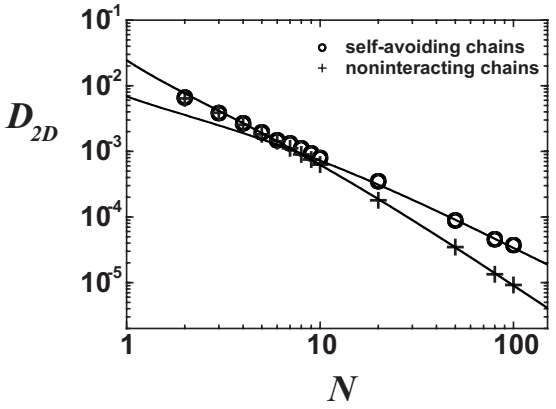

FIG. 4. Diffusion coefficient of the center of mass for chains consisting of N beads. The parameters of the model are $p_a=0.2$, $p_b=0.05$, and $p_c=1$. Crosses correspond to noninteracting chains and circles to self-avoiding chains. Numerical results were obtained using Monte Carlo simulations. Lines correspond to the analytical approximation of Eqs. (8) and (11) with $A=1.06$ and $B=0.30$.

hole is successful with frequency $p_a(1-P_h)$, where P_h is the hole probability. Similarly, an end particle jumping attempt that annihilates a hole succeeds with frequency p_bP_h. In equilibrium we expect the same frequency for creation and annihilation. Thus, P_h can be expressed as

$$P_h = \frac{p_a}{p_a + p_b}. \tag{1}$$

Then, the average length of noninteracting chains, l_{ni}, is given by

$$l_{\mathrm{ni}} = N + P_h(N-1). \tag{2}$$

Note that if $p_a > p_b$, then $P_h > 1/2$ and we are dealing with "long chains." Conversely, if $p_a < p_b$ we are dealing with "short chains." The validity of Eq. (2) has been verified by means of Monte Carlo simulations.

Figure 1(c) shows the equivalent one-dimensional configuration of the two-dimensional configuration (b). The jumping probabilities in the equivalent one-dimensional configuration are exactly the same as the jumping probabilities of the one-dimensional necklace model introduced in Refs. [9]. This equivalence holds for all configurations of the two-dimensional noninteracting necklace model. Therefore, we conclude that the one-dimensional diffusion coefficient of the center of mass of the two-dimensional noninteracting chain in the tube, D_{1D}, is equal to the diffusion constant D of the one-dimensional necklace model. The exact expression of D for $N \geqslant 2$ was found in Refs. [10,12] (see also [13] where related results are obtained) and it is given by

$$D = \frac{p_a p_b p_c}{(p_a + p_b)[(N-2)(p_a + p_b) + 2p_c]}. \tag{3}$$

We now consider the two-dimensional self-avoiding necklace model. The definition of the model is similar to the noninteracting case, but now loops are forbidden and only

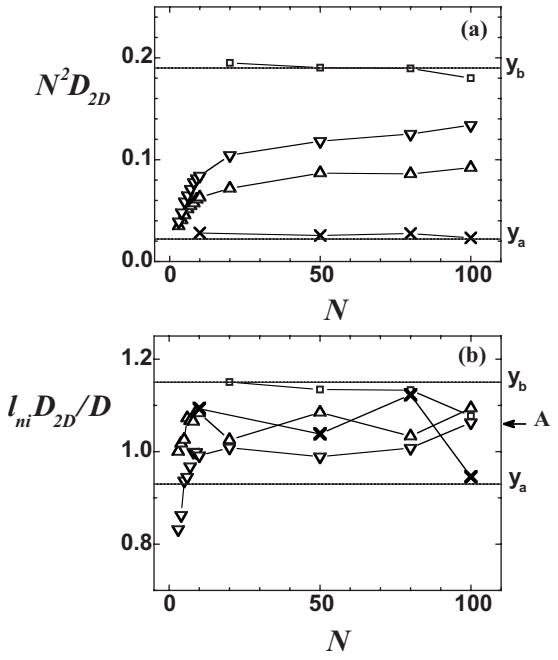

FIG. 5. Testing the validity of the analytical approximation for the noninteracting case. (a) N^2D_{2D} vs. N, where D_{2D} is the diffusion coefficient in two dimensions and N is the number of particles of the chain, see Eq. (9). (b) $l_{\mathrm{ni}}D_{2D}/D$ vs. N where l_{ni} is the average length of the chain and D is the diffusion coefficient in one dimension, see Eq. (7). The Monte Carlo results were obtained using $p_a=p_b=0.5$ and $p_c=1$ (squares), $p_a=0.2$, $p_b=0.05$ and $p_c=1$ (up triangles), $p_a=0.05$, $p_b=0.2$, and $p_c=1$ (down triangles), and $p_a=0.95$, $p_b=0.05$, and $p_c=1$ (crosses). The lines between symbols are drawn as a guide to the eye. For larger values of N the numerical results lie between the two horizontal straight lines of ordinates y_a and y_b, with $y_b/y_a=8.6$ in (a) and $y_b/y_a=1.2$ in (b). For $N \geqslant 50$ the error bar for each point is about 7%. Taking into account these results, and using standard statistical analysis, one obtains $A=1.06\pm0.02$.

one particle or one hole is allowed at each lattice site (see Fig. 2). The above jumping rules (ii)–(iv) are the same, and the rule (i) is replaced by the following one.

(i′) An end particle with a nearest site occupied by the corresponding pre-end particle jumps with a probability per unit time $p_a/3$ to each of the nearest sites which are not occupied by a particle or a hole of the chain. In other words, an end particle cannot jump to a site that belongs to the chain avoiding the formation of loops (i.e., hardcore particle-particle, particle-hole, and hole-hole interactions are taken into account).

Due to the rule (i′) for the self-avoiding case, and depending on the configuration, the total jumping probability per unit time of the end particle can be either p_a or less than p_a. Therefore, the hole probability must be smaller than the corresponding one for the noninteracting case, and the average chain length, l_{sa}, smaller than l_{ni} given by Eq. (2). However, for various sets of parameters (p_a,p_b,p_c), we verified by means of Monte Carlo simulations that, for large enough values of N (typically for $N \geqslant 30$) the ratio $l_{\mathrm{sa}}/l_{\mathrm{ni}}$ is close to one and does not depends on N. For example, for $(p_a=0.2$, $p_b=0.05$, *and* $p_c=1)$, $l_{\mathrm{sa}}/l_{\mathrm{ni}}=0.9930\pm0.0004$, and for

(p_a=0.05, p_b=0.2, *and* p_c=1), l_{sa}/l_{ni}=0.9886±0.0005 (these values were obtained in the range of N from 50 to 100 particles).

Let r be the mean value of the end-to-end distance of a chain of N particles. For large enough value of N one expects that

$$r^2 \sim l^{2v}, \tag{4}$$

where v is the end-to-end distance exponent, and l is the average chain length (l is l_{ni} or l_{sa} for the noninteracting and the self-avoiding chains, respectively). From the Monte Carlo results we found for large enough values of N that $v=1/2$ for the noninteracting case and $v=3/4$ for the self-avoiding case. These results are in agreement with the well-known values of the v exponent for the end-to-end distance of the trajectories in random and self-avoiding random walks.

III. ANALYTICAL APPROXIMATIONS

Using scaling arguments [8] we will obtain analytical approximations for the diffusion constant, D_{2D}, of the center of mass of the chains in two dimensions for the noninteracting and self-avoiding cases. Let us consider the general case of a chain that moves in a medium with obstacles (see Fig. 3). These obstacles restrict the lateral motion of the chain. As it was mentioned above, this situation can be approached with a chain confined in a tube [1–3,5]. Consider then that initially the chain is confined in a tube. After a time, t_{escape}, the chain escapes from its original tube and adopts a new configuration that is not correlated with the initial one. In other words, at the scale of time t_{escape} the center of mass of the chain performs and ordinary (uncorrelated) random walk. Then, one can write that $r_{c.m.}{}^2 \sim D_{2D}t$, where $r_{c.m.}{}^2$ is the mean square displacement of the center of mass and D_{2D} is the diffusion coefficient in two dimensions.

Considering now the one-dimensional motion of the chain along the tube, one has $l^2 \sim D_{1D}\, t_{escape}$, where l is the average length of the chains and D_{1D} is the diffusion constant in its tube (i.e., D_{1D} is the one-dimensional diffusion coefficient). The mean square displacement, $r_{c.m.}{}^2$, behaves as the square of the mean value of the end-to-end distance of a chain, r^2 [see Eq. (4)]. Finally, by combining all these expressions, one obtains that, in the asymptotic limit ($N\to\infty$), the diffusion constant of the center of mass of the chains behaves as

$$D_{2D} \sim l^{2v-2} D_{1D}. \tag{5}$$

Although Eq. (5) holds only in the asymptotic limit ($N\to\infty$), we propose the following approximation for all values of $N \geqslant 2$:

$$D_{2D} = \text{const}\; l^{2v-2} D_{1D}. \tag{6}$$

For noninteracting chains, $D_{1D}=D$ [see Eq. (3)], $l=l_{ni}$, and $v=1/2$, then

$$D_{2D} = A l_{ni}^{-1} D, \tag{7}$$

where A is a constant. From Eqs. (1)–(3), one finally obtains

$$D_{2D} = A \frac{p_a p_b p_c}{[(N-2)(p_a+p_b)+2p_c][N(p_a+p_b)+(N-1)p_a]}. \tag{8}$$

and for $N \gg 1$,

$$D_{2D} = A \frac{p_a p_b p_c}{N^2(p_a+p_b)(2p_a+p_b)} \sim \frac{1}{N^2}. \tag{9}$$

In order to obtain an analytical expression for D_{2D} in the self-avoiding case, we approximate D_{1D} with the exact diffusion coefficient D (which correspond to the noninteracting case), and l by l_{ni} [see Eq. (2)]. Then, from Eq. (6) one obtains (using $v=3/4$)

$$D_{2D} = B l_{ni}^{-1/2} D, \tag{10}$$

where B is a new constant. Now from Eqs. (1)–(3) one has

$$D_{2D} = B \frac{p_a p_b p_c}{[(N-2)(p_a+p_b)+2p_c][N(p_a+p_b)^2+(N-1)p_a(p_a+p_b)]^{1/2}}, \tag{11}$$

and for $N \gg 1$

$$D_{2D} = B \frac{p_a p_b p_c}{N^{3/2}(p_a+p_b)^{3/2}(2p_a+p_b)^{1/2}} \sim \frac{1}{N^{3/2}}. \tag{12}$$

IV. RESULTS

Using Monte Carlo simulations the diffusion constant D_{2D} was obtained through $D_{2D}=\langle[R_{c.m.}(t)-R_{c.m.}(0)]^2\rangle/4t$, where $R_{c.m.}(t)$ is the position of the center of mass of a chain at time t, and the brackets denote the ensemble average. We performed simulations for chains lengths from $N=2$ to 100. The simulations starts with an arbitrary chain configuration and, following the rules of the model, many jumps were performed to reach an equilibrium configuration. At this point the origin of the timescale, $t=0$, is defined. Simulations were performed up to $t=2.10^7$ and average values were obtained using more than 10^3 samples.

Figure 4 shows the Monte Carlo results of the two dimensional diffusion constant D_{2D} as a function of N for the noninteracting and self-avoiding cases using the same values of parameters p_a, p_b, and p_c, and the analytical approximations (8) and (11) (where $A=1.06$ and $B=0.30$; see below). Note

that for small values of N the self-avoiding effect is null or negligible (the chain cannot cross itself or the probability of loops formation is very small). Therefore, the numerical results for both cases coincide for $N \leqslant 6$. One can clearly see that for large values of N the analytical approximations are in agreement with the corresponding Monte Carlo results.

In order to verify the asymptotic behavior $1/N^2$ given by Eq. (9), in Fig. 5(a) we plotted the Monte Carlo results of $N^2 D_{2D}$ against N for the noninteracting case and for different sets of p_a, p_b, and p_c. For large values of N, Eq. (9) holds but $N^2 D_{2D}$ strongly depends on the values of the free parameters of the model. All these results are in a strip of a relative width of $y_b/y_a = 8.6$, where y_b refers to the maximum and y_a to the minimum values of the strip ordinates. In Fig. 5(b) we plot $l_{ni} D_{2D}/D$ as a function of N for the same sets of the free parameters than those used in (a). Note that now $y_b/y_a = 1.2$ and if we take into account the fluctuations of the numerical results (i.e., the error bars) one can conclude that the data are in agreement with Eqs. (7) and (8) with $A = 1.06 \pm 0.02$. Let us stress that, even from relatively small values of $N (N \cong 10)$, the analytical approximation of Eq. (8) works reasonably well.

In Fig. 6 we show the Monte Carlo data of $N^{3/2} D_{2D}$ and $l_{ni}^{1/2} D_{2D}/D$ versus N for the self-avoiding case with different sets of p_a, p_b, and p_c [see Eqs. (10)–(12)]. In this case one can conclude that Eq. (11), with $B = 0.30 \pm 0.02$, is a reasonably good approximation of the two-dimensional diffusion coefficient for $N \geqslant 50$. We cannot prove that A and B are exactly the same for all sets of the free parameters. However, we can state that the dependence of A and B on p_a, p_b, and p_c, if it exists, is very weak.

The necklace model was defined on a square lattice, but other types of two-dimensional lattices can be used (e.g., triangular, hexagonal, etc.). We expect that the same analytical approximations (8) and (11) hold, with the constants A and B depending on the specific type of the lattice used.

V. SUMMARY AND FINAL REMARKS

In this work, we have introduced and analyzed a discretized model of reptation of a chain consisting of N particles in a square lattice (the two-dimensional necklace model). Two cases were considered. In the first one, the hard-core interaction is not taken into account and the chain can cross itself (the noninteracting case). In the second one, the formation of chain loops is forbidden (the self-avoiding case). For both cases, and using scaling argument in the asymptotic limit $(N \to \infty)$, analytical approximation for the diffusion constant of the center of mass, for all values of N, were proposed [see Eqs. (8) and (11)]. Even for small values of N the Monte Carlo results are in good agreement with these approximations.

The necklace model and the analytical approximations for the diffusion coefficient can be easily extended to higher d-dimensions. We expect that Eq. (6) holds for all d (i.e., $D_{nD} = \text{const}\, l^{2\upsilon-2} D_{1D}$, where $n = 2, 3, 4, \ldots$) with the corresponding value of the υ exponent. That is, the well-known values of the υ exponent for random and self-avoiding random walks should be used. It is known that (see e.g., [15])

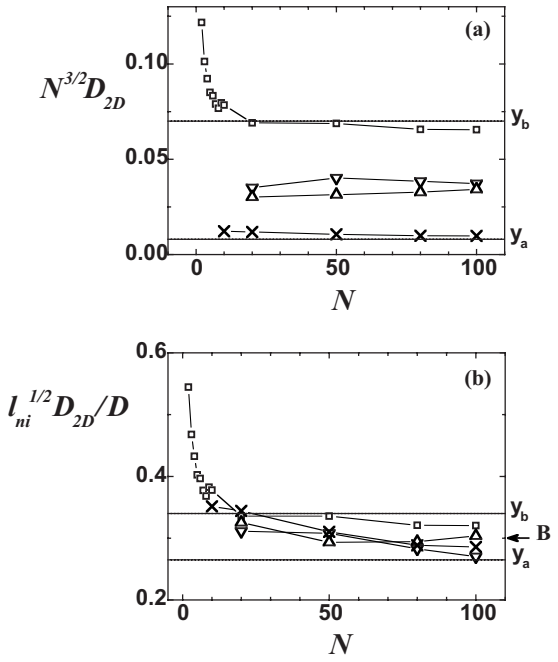

FIG. 6. Testing the validity of the analytical approximation for the self-avoiding case. (a) $N^{3/2} D_{2D}$ vs. N where D_{2D} is the diffusion coefficient in two dimensions and N is the number of particles of the chain, see Eq. (12). (b) $l_{ni}^{0.5} D_{2D}/D$ vs. N where l_{ni} is the average length of the chain for the noninteracting case and D is the diffusion coefficient in one dimension, see Eq. (10). The Monte Carlo results were obtained using $p_a = p_b = 0.5$ and $p_c = 1$ (squares), $p_a = 0.2$, $p_b = 0.05$, and $p_c = 1$ (up triangles), $p_a = 0.05$, $p_b = 0.2$, and $p_c = 1$ (down triangles), and $p_a = 0.95$, $p_b = 0.05$, and $p_c = 1$ (crosses). For large values of N the numerical results lie between the two horizontal straight lines of ordinates y_a and y_b, with $y_b/y_a = 8.75$ in (a) and with $y_b/y_a = 1.28$ in (b). For $N \geqslant 50$ the error bar for each point is about 7%. Taking into account these results, and using standard statistical analysis, one obtains $B = 0.30 \pm 0.02$.

$\upsilon = 1/2$ for $d \geqslant 2$ for the noninteracting case; and $\upsilon = 3/4$ for $d = 2$, $\upsilon = 0.588 \pm 0.001$ for $d = 3$, and $\upsilon = 1/2$ for $d \geqslant 4$ for the self-avoiding case. For the noninteracting case $D_{1D} = D$, and $l = l_{ni}$ [see Eqs. (2) and (3)]. For the self-avoiding case we approximate D_{1D} by D, and l by l_{ni}. Note that for $d \geqslant 4$ it is expected that the self-avoiding character does not play a relevant role, and then the diffusion coefficient in both cases would be exactly the same. In the noninteracting case $\upsilon = 1/2$ for $d \geqslant 2$, then the same analytical approximation (8) holds for $d \geqslant 2$, where the constant A depends on the dimensionality and type of the lattice used. Then, for large values of $N, l \sim N$ and thus diffusion constants behave as N^{-2} for $d \geqslant 2$. This result seems to be universal, i.e., it is not sensitive to the details of the model. Indeed, a variety of models presents the same scaling exponent such as Duke's model [7] or de Gennes' model as numerically simulated by Barkema and Krenzlin [16]. Moreover, Willmann and Schütz show that the scaling of the diffusion constant in the limit of long chains is unaffected by a kinematic disorder [17].

The scaling behavior of Eq. (5) as a function of N can be extended to other reptation models. In general $l \sim N$, and it is expected that $D_{1D} \sim 1/N$, for $N \gg 1$. Therefore, one obtains $D_{2D} \sim N^{2\upsilon-3}$. This asymptotic behavior holds for $d \geqslant 2$ (i.e.,

$D_{\mathrm{nD}} \sim N^{2v-3}$, where $n=2,3,4,\ldots$) with the corresponding value of the v exponent. For $d=2,3$ this is in agreement with the power-law scaling obtained for the self-avoiding fast extron model introduced in Ref. [14].

The Duke model [7] is one of the most well-known discretized model for reptation. It is hard to obtain its exact diffusion constant for all values of N, which up to now it is not known. Exact solutions are only known for small values of N and the asymptotic scaling behavior was obtained (see, e.g., [8,18,19]). Conversely, the analytical approximations for the necklace model, we have presented here, work reasonably well even for relatively small values of N, and they improve as N increases.

Since the Duke model does not take into account hardcore interactions, one is prone to compare this model with the noninteracting necklace model. For the Duke model and large N it was found $N^2 D_{\mathrm{Duke}} = C_1[1 + C_2 N^{-\gamma}]$. The exact value of γ is an unresolved issue. Barkema *et al.* [19], from numerical simulations, determined that $\gamma = 2/3$; Prahofer and Spohn [20] found analytically that $1/2 < \gamma < 2/3$ and also argue for $1/2$; and Carlon *et al.* [21] also proposed $\gamma = 1/2$. From Eq. (8) one can obtain the first two leading terms for $N^2 D_{2\mathrm{D}}$ to be

$$N^2 D_{2\mathrm{D}} = C(1+f), \tag{13}$$

where the constant C and the function f are given by

$$C = A \frac{p_a p_b p_c}{(p_a + p_b)(2p_a + p_b)}, \tag{14}$$

$$f(N, p_a, p_b, p_c) = \frac{1}{N}\left[2\frac{p_a + p_b - p_c}{p_a + p_b} + \frac{p_a}{2p_a + p_b}\right]. \tag{15}$$

Equation (8) is based on several assumptions and then we cannot be sure of the exact dependence on N of the subleading term. Due to the precision of our calculations, we cannot either determine the exact dependence from our Monte Carlo results. However, Eq. (15) predicts that f can be positive, null, or negative and this is something that we can check. Indeed, Monte Carlo simulations clearly show that the subleading term can be positive or negative. In particular, the results of Fig. 5(a) obtained using $p_a=0.2$, $p_b=0.05$, $p_c=1$, and $p_a=0.05$, $p_b=0.2$, $p_c=1$ clearly show negative values of f. This is consistent with the expected negative values that can be obtained with Eq. (15). Since the Duke model presents a positive second leading term, we conclude that, in this respect, the necklace model can behave differently.

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