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Polymer translocation out of confined environments

Kaifu Luo,^{1,*} Ralf Metzler,¹ Tapio Ala-Nissila,^{2,3} and See-Chen Ying³

¹Physics Department, Technical University of Munich, D-85748 Garching, Germany

²Department of Applied Physics, Helsinki University of Technology, P.O. Box 1100, FIN-02015 TKK Espoo, Finland

³Department of Physics, Brown University, P.O. Box 1843, Providence, Rhode Island 02912-1843, USA

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We consider the dynamics of polymer translocation out of confined environments. Analytic scaling arguments lead to the prediction that the translocation time scales like $\tau \sim N^{\beta+\nu_{2D}} R^{1+(1-\nu_{2D})/\nu}$ for translocation out of a planar confinement between two walls with separation R into a three-dimensional (3D) environment, and $\tau \sim N^{\beta+1} R$ for translocation out of two strips with separation R into a two-dimensional (2D) environment. Here, N is the chain length, ν and ν_{2D} are the Flory exponents in 3D and 2D, and β is the scaling exponent of translocation velocity with N , whose value for the present choice of parameters is $\beta \approx 0.8$ based on Langevin dynamics simulations. These scaling exponents improve on earlier predictions.

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I. INTRODUCTION

The transport of a polymer through a nanopore has received increasing attention due to its importance in biological systems [1] such as gene swapping through bacterial pili, *m*-RNA transport through nuclear pore complexes, and injection of DNA from a virus head into the host cell, etc. Moreover, translocation processes have various potentially revolutionary technological applications [2,3], such as rapid DNA sequencing, gene therapy, and controlled drug delivery.

In addition to its biological relevance, the transport dynamics of polymers through nanopores is of fundamental interest in physics and chemistry. There exists a flurry of experimental [4–7] and theoretical [8–29] studies devoted to this subject. The passage of a flexible chain through a nanopore involves a large entropic barrier, thus polymer translocation needs driving forces, which can be provided by an external applied electric field in the pore [2,4–6,12,26–29], a pulling force exerted on the end of a polymer [17,25,29], binding particles (chaperones) [14,15], or geometrical confinement of the polymer [7,9,11,19–21].

Among these mechanisms, less attention has been paid to confinement-driven translocation. In particular its dynamics remains unclear, despite its importance to biological processes including viral ejection, drug delivery, controlled release from a nanochannel, etc. We here investigate the generic behavior of polymer release through a small pore from a confined environment (Fig. 1).

Based on Kantor and Kardar's scaling arguments for unhindered motion of the chain [17], Cacciuto and Luijten [19] suggested that $\tau \sim N^{1+\nu} R^{1/\nu}$ for planar confinement shown in Fig. 1, where ν is the Flory exponent [31–33] [$\nu=0.588$ in three-dimensional (3D)] and R the separation between the walls. Panja *et al.* [20] considered translocation out of planar confinement as two-dimensional version of translocation through a nanopore with an electric field applied in the pore, and suggested $\tau \sim N^{2\nu_{2D}}$ [$\nu_{2D}=3/4$ in two-dimensional (2D)],

in contrast to above scaling exponent $1+\nu$. To solve this contradiction, in this work we investigate the dynamics of polymer translocation out of planar confinement (3D) and out of two strips (2D) using both analytic scaling arguments and Langevin dynamics simulations. As in the above-mentioned theories, we also ignore the hydrodynamic interactions here. Our scaling arguments include geometric effects that have been left out in previous studies, leading to a set of scaling exponents of translocation time τ with regard to polymer length N as well as the size of the confinement space R . The theoretical findings for R dependence of τ are

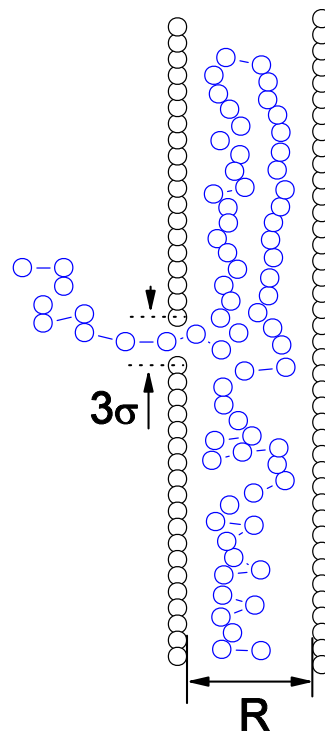


FIG. 1. (Color online) A schematic representation of the system in 2D. The simulations are carried out in both 2D and 3D. For a planar confinement (3D), two plates are separated by a distance R . One wall has a pore of length $L=\sigma$ and diameter $W=3\sigma$. For polymers confined between two strips (2D) the pore width is 3σ .

*Author to whom correspondence should be addressed. luokaifu@gmail.com

fully supported by numerical simulation results both in 2D and 3D. We consider geometry as shown in Fig. 1, where two walls with separation R are formed by stationary particles within a distance σ from each other.

II. SCALING ARGUMENT

A number of recent theories [8,10,16,17] have been developed for the dynamics of polymer translocation. Of them, Kantor, and Kardar [17] provided lower bounds for both the translocation time for pulling the polymer by the end and driving the polymer by a chemical potential difference applied across the membrane. Essentially, the lower bound is the time for the unimpeded polymer moving a distance of order of its size. For driven translocation with a chemical potential difference applied across the membrane, there is a force of $\Delta\mu/\sigma$ applied to the few monomers inside the pore. As a consequence, Kantor and Kardar [17] argued that its center of mass should move with a velocity $v \sim \Delta\mu/N$. Thus the lower bound for the translocation time of an unhindered polymer is the time to move through a distance R_g (radius of gyration of the polymer in an unconfined state), and scales as $\tau \sim R_g/v \sim N^{1+\nu}/(\Delta\mu)$.

Now, let us focus on the translocation out of confined environments. According to the blob picture [31], a chain confined between two parallel plates with separation $\sigma \ll R \ll R_g$ will form a 2D self-avoiding walk consisting of n_b blobs of size R . Each blob contains $g = (R/\sigma)^{1/\nu}$ monomers and the number of blobs is $n_b = N/g = N(R/\sigma)^{-1/\nu}$. The free-energy cost of the confined chain in units of $k_B T$, ΔF , is simply the number of the blobs. Thus the chemical potential difference that provides a driving force for the translocation process is given by

$$\Delta\mu = \Delta F/N \sim R^{-1/\nu}. \quad (1)$$

The remaining ingredients to complete the scaling argument are the length scale L_0 through which the polymer has to move during the entire translocation process and the average translocation velocity v . As a result, the translocation time can be estimated as $\tau \sim L_0/v$. Cacciuto *et al.* [19] chose the radius of gyration R_g in the unconfined state for L_0 and $v \sim \Delta\mu/N$ and obtained the scaling result $\tau \sim N^{1+\nu}R^{1/\nu} \sim N^{1.588}R^{1.70}$, while Panja *et al.* [20] criticized the scaling exponent $1+\nu$ for N dependence of τ and considered the translocation out of the planar confinement as the 2D version of translocation driven by a chemical potential difference applied across the membrane. As a result, they obtained the exponent $2\nu_{2D} = 1.50$ for the N dependence in a 3D system.

Here, we argue that the correct scaling results can only be obtained by properly identifying the length scale L_0 and the scaling of the translocation velocity v . Due to the highly nonequilibrium nature of the driven translocation problem, the scaling of the average translocation velocity v with respect to the chain length N is of the form $v \sim N^\beta$, where the exponent $\beta \leq 1$ [30]. For the present choice of parameters, we find that $\beta \approx 0.8$ as will be demonstrated below. For planar confinement (3D), the blob picture predicts the longitudinal size of the polymer to be [31–33]

$$R_{\parallel} \sim N^{\nu_{2D}} \sigma \left(\frac{\sigma}{R} \right)^{\nu_{2D}/\nu-1} \sim N^{3/4} \sigma \left(\frac{\sigma}{R} \right)^{0.28}, \quad (2)$$

with ν_{2D} being the Flory exponent in 2D. For successful translocation out of planar confinement, the polymer has to move a distance $L_0 \sim R_{\parallel}$. Therefore, the translocation time can be estimated as

$$\tau \sim \frac{R_{\parallel}}{v} \sim N^{\beta+\nu_{2D}} R^{1+(1-\nu_{2D})/\nu}. \quad (3)$$

Numerically, Eq. (3) yields a scaling result of $\tau \sim R^{1.43}$ for confinement-driven translocation in 3D, which is different from the prediction $\tau \sim R^{1.70}$ obtained by Cacciuto *et al.* [19]. In fact, their numerical results based on Monte Carlo simulations show $\tau \sim R^{1.54 \pm 0.10}$, which is close to our scaling prediction in Eq. (3).

Next, we also consider translocation out of confinement into a 2D environment which has not been addressed previously [9,11,19,20]. For a polymer confined between two strips embedded in 2D, the blob picture predicts the longitudinal size of the chain to be [31,33]

$$R_{\parallel} \sim N \sigma \left(\frac{\sigma}{R} \right)^{-1+1/\nu_{2D}} \sim NR^{-1/3}. \quad (4)$$

In this case the free-energy excess in Eq. (1) is valid if ν is replaced by ν_{2D} . Thus, the translocation time scales as

$$\tau \sim \frac{R_{\parallel}}{v} \sim \frac{NR^{1-1/\nu_{2D}}}{N^{-\beta}R^{-1/\nu_{2D}}} \sim N^{\beta+1}R, \quad (5)$$

showing a linear dependence on R . Both predictions in Eqs. (3) and (5) for R dependence are confirmed by simulations below.

III. MODEL AND METHODS

In our numerical simulations, the polymer chains are modeled as bead-spring chains of Lennard-Jones (LJ) particles with the finite extension nonlinear elastic (FENE) potential. Excluded volume interaction between monomers is modeled by a short range repulsive LJ potential: $U_{LJ}(r) = 4\epsilon[(\frac{\sigma}{r})^{12} - (\frac{\sigma}{r})^6] + \epsilon$ for $r \leq 2^{1/6}\sigma$ and 0 for $r > 2^{1/6}\sigma$. Here, σ is the diameter of a monomer, and ϵ is the potential depth. The connectivity between neighboring monomers is modeled as a FENE spring with $U_{FENE}(r) = -\frac{1}{2}kR_0^2 \ln(1-r^2/R_0^2)$, where r is the distance between consecutive monomers, k the spring constant, and R_0 the maximum allowed separation between connected monomers. Between all monomer-wall particle pairs, there exists the same short-range repulsive LJ interaction as described above.

In the Langevin dynamics simulation, each monomer is subjected to conservative, frictional, and random forces, respectively, with [34] $m\dot{\mathbf{r}}_i = -\nabla(U_{LJ} + U_{FENE}) - \xi\dot{\mathbf{r}}_i + \mathbf{F}_i^R$, where m is the monomer's mass, ξ is the friction coefficient, and \mathbf{F}_i^R is the random force which satisfies the fluctuation-dissipation theorem. In the present work, we use the LJ parameters ϵ and σ and the monomer mass m to fix the energy, length, and mass scales, respectively. Time scale is then given by $t_{LJ} = (m\sigma^2/\epsilon)^{1/2}$. The dimensionless parameters in our simula-

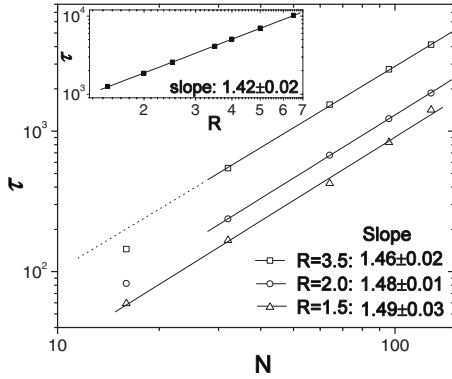


FIG. 2. Translocation time τ as a function of the chain length N for different R in 3D. The insert shows τ as a function of R for chain length $N=128$ in 3D.

tions are $R_0=2$, $k=7$, $\xi=0.7$, and $k_B T=1.2$. The Langevin equation is integrated in time by a method described by Ermak and Buckholz [35] in both 3D and 2D. To create the initial configuration, the first monomer of the chain is placed in the entrance of the pore, while the remaining monomers are initially squeezed into the space between two plates (3D) and the space between two strips (2D) under thermal collisions described by the Langevin thermostat to obtain an equilibrium configuration. Typically, we average our data over 1000 independent runs.

According to the definition of the translocation time, at the completion of the translocation process, the chain has moved a distance of $R_{||}$ along the direction perpendicular to the axis of the pore. We now define the translocation velocity as $v = \langle R_{||} \rangle / \tau$. Using the definition $v = \langle R_{||i} / \tau_i \rangle$ for the translocation velocity, we observed similar results. Here $R_{||i}$ and τ_i denote the values of $R_{||}$ and τ for each successful run.

IV. NUMERICAL RESULTS

For translocation out of a planar confinement, the results are shown in Fig. 2. The insert of Fig. 2 shows the R dependence of the translocation time. The scaling exponent is 1.42 ± 0.02 , which is in good agreement with the exponent 1.43 from our scaling prediction in Eq. (3). For τ with N for $R=3.5, 2.0$ and 1.5 , we get the scaling exponents of $1.46 \pm 0.02, 1.48 \pm 0.01$, and 1.49 ± 0.03 , respectively. With decreasing R , the exponent slightly increases. As to translocation velocity, such as for $R=1.5$ we get $\beta=0.77 \pm 0.01$. According to Eq. (3), $\beta + \nu_{2D}=1.52$, which is very close to 1.49. These results demonstrate that the scaling arguments for unimpeded translocation provide an accurate estimate for the translocation time. Although the scaling exponent for N dependence is quite close to the value 1.55 obtained by Caciuto *et al.* [19], the physical origin is quite different.

The dynamics of a single segment passing through the pore during translocation is an important quantity considerably affected by different driving mechanisms. The nonequilibrium nature of translocation has a significant effect on it. We have numerically calculated the waiting times for all monomers in a chain of length N . We define the waiting time of monomer s as the average time between the events that

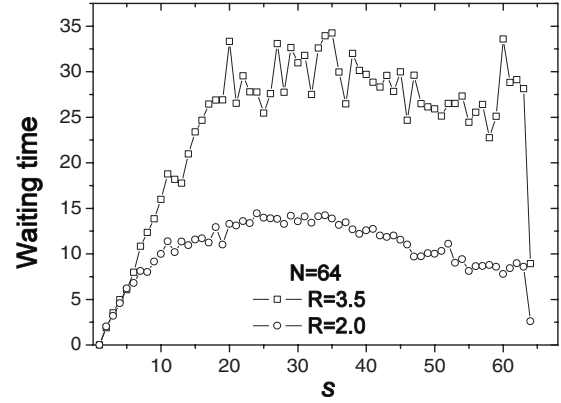


FIG. 3. Waiting time distribution for 3D simulations.

monomer s and monomer $s+1$ exit the pore. In our previous work [25,26] studying translocation driven by a voltage across the pore without confinement on either side, we found that for short polymers, such as $N=100$, the monomers in the middle of the polymer need the longest time to translocate and the distribution is close to symmetric with respect to the middle monomer [26]. Figure 3 shows the waiting time distributions for translocation out of planar confinements with $R=2.0$ and 3.5 for $N=64$. Compared with the unconfined potential driven case, the waiting time distribution is quite different. The waiting times increase until $s \sim 20$ and then almost saturate. This can be qualitatively understood as follows. The initial rise of the waiting time has the same origin as the potential driven case, with $\Delta\mu$ given by Eq. (1) playing the role of the applied voltage. Here, the driving force has two components, one from the cost of the free energy due to blobs and the second due to entropic wall repulsion for the translocated beads. These are balanced by frictional force in the pore. The reduction in the overall driving causes the initial rise of the waiting time distribution, which is eventually balanced by decreasing frictional force leading to a plateau in the waiting time distribution.

For translocation out of two strips, the results are shown in Fig. 4. The translocation time increases linearly with increasing R with the scaling exponent 1.04 ± 0.01 , see the insert of Fig. 4, which is in excellent agreement with the prediction in Eq. (5). Moreover, for τ as a function of N , we get scaling exponent of 1.82 ± 0.01 for $R=4.5$. For translocation velocity, we get $\beta=0.79 \pm 0.02$. According to Eq. (5), $\beta+1=1.79$, which is very close to the scaling exponent 1.82.

Figure 5 shows the waiting time distributions for translocation out of two strips with $R=4.5$ for $N=16, 32, 64$, and 96 . For $N \geq 64$, the waiting time increases with s until the maximum at $s \sim 20$, and then almost linearly decreases with s . For all chain lengths, the monomers at the end of the chain from $s=N-4$ to $N-1$ still need a little longer time to translocate due to very weak confinement. The observed behavior is due to the interplay of decreasing $\Delta\mu$ and increasing pulling entropic force. The balance of these two factors is differs from 2D to 3D.

V. CONCLUSIONS

In this work, we have considered the dynamics of polymer translocation out of confined environments based on

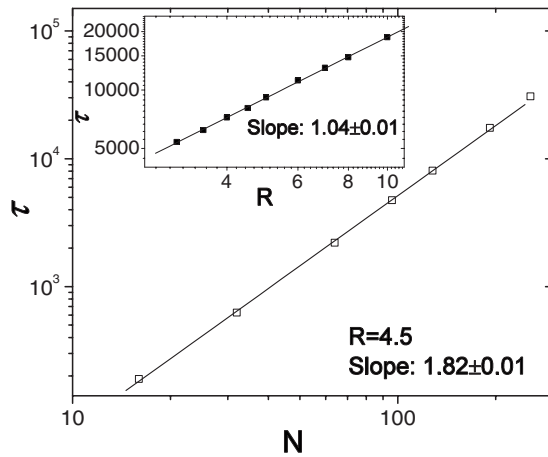


FIG. 4. Translocation time τ as a function of the chain length N for $R=4.5$ in 2D. The insert shows τ as a function of R for chain length $N=128$ in 2D.

both scaling arguments and Langevin dynamics simulations. Analytic scaling arguments lead to the prediction that the translocation time scales such as $\tau \sim N^{\beta+\nu_{2D}}R^{1+(1-\nu_{2D})/\nu}$ for translocation out of a planar confinement between two walls with separation R into a 3D environment, and $\tau \sim N^{\beta+1}R$ for translocation out of two strips with separation R into a 2D environment. Here, N is the chain length, ν and ν_{2D} are the Flory exponents in 3D and 2D, and β is the scaling exponent

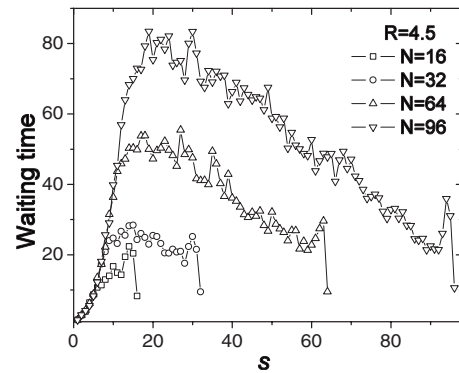


FIG. 5. Waiting time distribution for 2D simulations.

of translocation velocity with N , whose value for the present choice of parameters is $\beta \approx 0.8$ based on Langevin dynamics simulations. These scaling exponents improve on the previously reported results [19,20].

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