First-passage times in conical varying-width channels biased by a transverse gravitational force: Comparison of analytical and numerical results

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We study the crossing time statistic of diffusing point particles between the two ends of expanding and narrowing two-dimensional conical channels under a transverse external gravitational field. The theoretical expression for the mean first-passage time for such a system is derived under the assumption that the axial diffusion in a two-dimensional channel of smoothly varying geometry can be approximately described as a one-dimensional diffusion in an entropic potential with position-dependent effective diffusivity in terms of the modified Fick-Jacobs equation. We analyze the channel crossing dynamics in terms of the mean first-passage time, combining our analytical results with extensive two-dimensional Brownian dynamics simulations, allowing us to find the range of applicability of the one-dimensional approximation. We find that the effective particle diffusivity decreases with increasing amplitude of the external potential. Remarkably, the mean first-passage time for crossing the channel is shown to assume a minimum at finite values of the potential amplitude.

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I. INTRODUCTION

The Brownian motion of molecules, particles, or even living microorganisms in confined geometries such as pores and channels, plays a key role on various scales in both nature and technology [1–25]. Transport in confined geometries within quasi-one-dimensional systems exhibits a very rich and striking phenomenology and has been studied in depth in many contexts, examples including diffusion in human metabolism, breathing, or medical drug delivery [26], as well as in living cells [27], the motion of viruses and bacteria [28,29], solid-state and protein nanobiosensors for the detection and structural analysis of individual molecules [30–32], transport in zeolites [33], synthetic nanopores [34–37], microfluidic devices [38], channels in biological systems [39], and artificial pores in thin solid films [40].

A universal description of an unbiased Brownian particle is given by the free diffusion coefficient $D_0$ in homogeneous systems. In heterogeneous environments with finite characteristic length scales of the disorder, the particle motion becomes Brownian at times sufficiently exceeding the correlation time of the system [41], albeit the crossover time may be significantly delayed [42]. When the diffusion takes place in systems decorated with excluded-volume obstacles, the diffusion may be locally free and characterized by the diffusivity $D_0$, while at long times the particle motion is again Brownian but with an effective diffusion coefficient $D_{\text{eff}}$ [43–46]. Typically the mean squared displacement (MSD) in such systems monotonically crosses over from the short time behavior $\langle \Delta x^2(t) \rangle \approx D_0 t$ to $\approx D_{\text{eff}} t$, where $D_0 > D_{\text{eff}}$. Another relevant case is that of confinement by boundaries, in channels or porous media, in which a significant slowdown of the MSD is effected [47–49]. Spatial confinement modifies the equilibrium of the system and its dynamical properties, increasing the hydrodynamic drag on such components and limiting the configuration space accessible to its diffusing parts [50]. In this sense, asymmetry plays a major role in the transport of a Brownian particle through a channel [51,52], Brownian pumps [53,54], and Brownian ratchets [14,55].

In simpler systems transitions across entropic or energetic barriers effect single-exponential kinetics of processes such as channel-facilitated transport of solutes to isomerization reactions. Recent experiments with single biological nanopores, pulling proteins and nucleic acids, as well as single-molecule fluorescence spectroscopy have raised a number of questions that stimulated the theoretical and computational investigation of barrier-crossing dynamics [56–73]. The quantity of interest in such studies is the time required for the system to pass over the barrier region. This time, called direct-transit time or transition path, is a random variable, characterized by the associated probability density and mean value.

While the transition path quantifies exclusively successful crossing events, the first-passage time counts the entire time elapsed until the first accomplished crossing event. First-passage problems arise in an extensive range of stochastic processes of practical interest [12]. Indeed, examples for first encounter-controlled events [74] include chemical and biochemical reactions [75–78], distance-effects on rapid search of signaling molecules [79,80], trafficking receptors on biological membranes [81], animal foraging [82–84], and the spreading of sexually transmitted diseases in a human social network or of viruses across the World Wide Web [85]. It is
FIG. 1. Schematic representation of a two-dimensional asymmetric conical channel formed by straight walls in the presence of a constant transverse (“gravitational”) force $G$ (shown as a green downwards arrow). The lower boundary is given by $h_1(x) = \lambda_1 x - b$ (shown as the blue solid line), while the upper boundary is given by $h_2(x) = \lambda_2 x + b$ (shown as the blue solid line). The channel’s variable width is given by $w(x) = h_2(x) - h_1(x)$, and its straight midline by $y_0(x) = [h_1(x) + h_2(x)]/2$ (shown as the dotted orange line). Panel (a) shows an expanding channel, i.e., the transition of the particle occurs from the narrow to the wide end ($n \rightarrow w$). In such a case, the Brownian particle starts from the reflecting boundary located at $x = 0$ and is then removed by the absorbing boundary at $x = L$ (shown as the red vertical dashed line). Panel (b) shows a narrowing channel, i.e., the transition of the particle is from the wide to the narrow end ($w \rightarrow n$). In this case, the Brownian particle starts from the reflecting boundary located at $x = L$ and is removed by the absorbing boundary at $x = 0$ (shown as the red vertical dashed line).

It is well known that confinement in higher dimensions may give rise to an effective entropic potential in reduced dimensions. In fact, Eq. (1) is formally equivalent to the Smoluchowski equation

$$\frac{\partial c(x,t)}{\partial t} = \frac{\partial}{\partial x} \left\{ D(x) w(x) \frac{\partial}{\partial x} \left[ c(x,t) \right] \right\}, \quad (1)$$

where $w(x)$ is the channel width and $D(x)$ is the position-dependent diffusion coefficient. Equation (1) with a position-independent diffusion coefficient, $D(x) = D_0$, is known as the ordinary FJ equation [1,88]. The effective 1D probability density $c(x,t)$ is related to the 2D probability density $\rho(x,y,t)$ by the projection

$$c(x,t) = \int_{w(x)} w \rho(x,y,t) dy. \quad (2)$$

The expression for the position-dependent effective diffusion coefficient for a narrow 2D channel of varying width that has a straight midline suggested by Reguera and Rubi (RR) based on heuristic arguments reads

$$D(x) \approx D_{\text{RR}}(x) = \frac{D_0}{\left[ 1 + \frac{D_0}{4 w^2(x)} \right]^{\gamma}}, \quad (4)$$

where $w'(x) = dw(x)/dx$. This last equation is a generalization of Zwanzig’s expression [2]. Alternative derivations of this equation were given by Kalinay and Percus [16], Martens et al. [8] and García-Chung and co-workers [9].

Reguera and Rubi [6], Kalinay [21], and later Pompa-García and Dagdug [20], studied how Eq. (1) is modified when a gravitational and entropic potential coexist. Pompa-García and Dagdug showed that for an overdamped Brownian particle diffusing in a 2D asymmetric channel of varying...
cross-section in the presence of a constant force in the transverse direction, Eq. (3) takes on the modified form [89]

$$\frac{\partial c(x, t)}{\partial t} = \frac{\partial}{\partial x} \left( D(x) A(x) \frac{\partial}{\partial x} \left[ \frac{c(x, t)}{A(x)} \right] \right), \quad (5)$$

where

$$A(x) = \int_{h_1(x)}^{h_2(x)} e^{-\eta y} dy = \frac{1}{\xi} \left[ e^{-\eta h_1(x)} - e^{-\eta h_2(x)} \right]. \quad (6)$$

Equation (5) is obtained when a 2D asymmetric channel is bounded by perfectly reflecting walls given by smooth functions $h_1(x)$ and $h_2(x)$, and $\eta = G/k_BT$, where $G$ stands for the constant transverse-direction force, and $-\beta U(x) = \ln [A(x)/A(x_0)]$. In Eq. (6) the coupling between entropy and energy barriers is reflected by the presence of the term $\exp[gh_i(x)] (i = 1, 2)$. For a symmetrical channel, when $h_1(x) = -h_2(x)$, $A(x)$ reduces to $2/(2g) \sinh[gh_2(x)]$ [21].

Given the bulk diffusion constants $D_x$ and $D_y$ in the longitudinal and transverse directions, respectively, we can introduce the ratio $\epsilon = D_x/D_y$ of the diffusion constants in the longitudinal and transverse directions. This quantity is used as an expansion parameter of a perturbative series when the projection method is applied. Then, the resultant second- and higher-order derivatives of $h(x)$ are neglected. Inspired by RR’s and Kalinay’s work, Pompa-García and Dagdug found the effective diffusivity with the same structure as Eq. (4),

$$n_\eta(gw, y_0') = \frac{1}{\sinh^2 \left( \frac{1}{2} gw \right)} \times \left\{ 1 + \cosh^2 \left( \frac{1}{2} gw \right) - gw \coth \left( \frac{1}{2} gw \right) \right\} + \frac{4}{w^2} \left( y_0' - w' \coth \left( \frac{1}{2} gw \right) \right) + \tau \frac{1}{\sinh \left( \frac{1}{2} gw \right)} \coth^2 \left( \frac{1}{2} gw \right), \quad (7)$$

where $y_0'(x)$ is the midline of the channel. One of the most important features of Eq. (4) when $\eta$ is given by Eq. (7) is the breaking of symmetry for a strong field. Thus while the predicted diffusivity is $D_0/[1 + h^2(x)]$ when $G$ tends to infinity, for $G$ going to minus infinity, the predicted diffusivity is $D_0/[1 + h^2(x)]$. In both cases, this constrains the Brownian dynamics to one dimension over the boundaries $h_1$ or $h_2$, respectively. In Eq. (7), when $y_0' = 0$, $\eta(gh(x))$ goes from $1/3$ to $1$, from negligible $G$ to the strong field case, respectively.

It is worthwhile mentioning that when $G$ goes to zero, Eq. (7) is equal to $1/3 + 4y_0'^2/\epsilon w'$, and Eq. (4) for an asymmetric 2D channel reduces to

$$D(x) = \frac{D_0}{\left[ 1 + \frac{1}{2} w^2(x) \right]^{1/3 + 4y_0'^2/\epsilon w'^2}}. \quad (8)$$

When $y_0'$ goes to zero the diffusivity for a 2D symmetric channel, as proposed by RR, is recovered [6], and it differs less than 1% from the expression obtained by Dagdug and Pine [18,20].

Along with the problem of deriving the modified FJ equation, there are also questions of the range of applicability of this approximate one-dimensional description and the accuracy of the expressions for the effective position-dependent diffusivity. To establish the range of applicability of the effective diffusivity proposed by Pompa-García and Dagdug, the present paper focuses on the wide-to-narrow and narrow-to-wide transitions between the two ends of a conical channel under a transverse gravitational external field, as shown in Fig. 1. We study the MFPT for various parameters and observe a remarkable minimum of the MFPT at intermediate strengths of the external potential.

### III. RESULTS AND DISCUSSION

We now consider a particle diffusing in a 2D asymmetric conical tube of length $L$ and variable width $w(x)$ in the presence of the transverse gravitational external field $G$, as shown in Fig. 1. Note that here we use the arrow notation $x_a \to x_b$, which represents particles moving from $x_a$ to $x_b$. Then, $\tau(x_0 \to L)$ denotes the particle MFPT from the initial position at $x_0$ to the wide end of the channel located at $x = L$, in the presence of a reflecting boundary at the narrow channel end located at $x = 0$, see the setup in Fig. 1, panel (a). The MFPT, considered as a function of $x_0$, and assuming that the reduction to the effective 1D description is applicable by means of Eq. (5), satisfies [90]

$$\frac{1}{A(x_0)} \frac{d}{dx_0} \left[ D(x_0) A(x_0) \frac{d\tau(x_0)}{dx_0} \right] = -1, \quad (9)$$

subject to the boundary conditions

$$\tau \bigg|_{x_0=L} = \frac{d\tau(x_0)}{dx_0} \bigg|_{x_0=0} = 0. \quad (10)$$

The solution for $\tau(x_0 \to L)$ is given by

$$\tau(x_0 \to L) = \int_{x_0}^{L} \frac{dx}{D(x)A(x)} \int_{x_0}^{x} A(y)dy. \quad (11)$$

The MFPT $\tau_{n\to w}$ is the MFPT encoded by Eq. (11) with initial condition $x_0 = 0$, $\tau_{n\to w} = \tau(0 \to L)$.

Now, let $\tau(x_0 \to 0)$ be the MFPT from the initial position $x_0$ to the narrow end of the channel at $x = 0$, in the presence of the reflecting boundary at the wider channel end at $x = L$, see Fig. 1, panel (b). This MFPT satisfies Eq. (9) as well, with the boundary

$$\tau \bigg|_{x_0=0} = \frac{d\tau(x_0)}{dx_0} \bigg|_{x_0=L} = 0. \quad (12)$$

Integrating Eq. (9) with the boundary conditions (12) we obtain

$$\tau(x_0 \to 0) = \int_{x_0}^{L} \frac{dx}{D(x)A(x)} \int_{x_0}^{x} A(y)dy. \quad (13)$$

The MFPT $\tau_{w\to n}$ is the MFPT in Eq. (13) with $x_0 = L$, $\tau_{w\to n} = \tau(L \to 0)$. To compute the integrals in Eqs. (11) and (13), $A(x)$ given by Eq. (6), and the interpolation formula for $D(x)$ given by Eqs. (4) and (7) has to be replaced.

In Fig. 2 the effective diffusion coefficient for a conical 2D channel corresponding to Eqs. (4) and (7) are shown for intermediate $g$ values as well as the limiting cases, when $g \to 0$ and $g \to \infty$ as functions of the position $x$ and the channel boundary slope $\lambda$. One of the main characteristics of
In our simulations the interaction of particles with the reflecting boundaries are treated entirely as elastic collisions, such that the bouncing off from a boundary corresponds to a geometrical reflection. Thus if the particle’s next position would lie outside a channel boundary, the calculated trajectory corresponds to a reflection of the trajectory’s portion outside of the boundary. Simulations are run with the time step $\Delta t = 10^{-8}$, and the bulk diffusivity is set to $D_0 = 1$, so that $\sqrt{2D_0\Delta t} \ll 1$. Such a small time step is needed due to the magnitude of the involved forces. Taking a larger $\Delta t$ would lead to unmanageable bouncing effects with the boundaries, caused by the missing information of the steps in the discretized positions of the Brownian particle.

Finally, we set thermal energy to unity, $k_B T = 1$. Stochastic averages were obtained as ensemble averages over $5.0 \times 10^4$ independent trajectories.

The results for the MFPT $\tau_{w\rightarrow n}$ are shown in Fig. 3, demonstrating very good agreement of the theoretical expressions with the Brownian dynamics simulations. However, for growing values of the channel wall slope $\lambda$ we see that the theoretical results overestimates the MFPT somewhat, especially for the force strength $g = 10$ (left panel of Fig. 3). In contrast, the MFPT is somewhat underestimated for intermediate $\lambda$ and larger $g$ values (right panel of Fig. 3). In this channel setup, the external force directs the particles towards the boundary with a positive slope, causing an effective drift away from the narrow channel end. As can be seen in Fig. 3 the MFPT drastically increases with growing external force strength. As function of the channel slope $\lambda$ all curves for different $g$ values converge to a unique value for $\tau_{w\rightarrow n}$ in the limit of fully horizontal boundaries, $\lambda = 0$. In this case, the channel passage is not affected by the force, as can also be seen for the effective diffusivity in Fig. 2. As a function of the force strength $g$, the MFPT $\tau_{w\rightarrow n}$ has different values for different channel slopes $\lambda$ in the limit of vanishing $g$. This behavior is the purely geometric effect of a narrowing channel. We finally note that both panels show a monotonic increase of the MFPT $\tau_{w\rightarrow n}$ for growing parameter $g$ as function of the channel slope $\lambda$ as well as for growing slope $\lambda$ as function of force strength $g$.

The results for the MFPT $\tau_{n\rightarrow w}$ for channel passage from the narrow to the wide end in Fig. 4 also show a very good agreement between the theoretical predictions and the Brownian dynamics simulations. Generally, the absolute values for the MFPT are considerably lower than for the opposite case $\tau_{w\rightarrow n}$. Moreover, the discrepancies are significantly less for $\tau_{n\rightarrow w}$ for all values of channel slope $\lambda$ and force strength $g$. Finally, we observe that the value of $\tau_{n\rightarrow w}$ has the opposite trend as function of $\lambda$ and $g$ as compared to $\tau_{w\rightarrow n}$: here, growing slope as well as increasing force strength lead to an effective drift towards the channel exit at the wide end, and thus to a reduction of $\tau_{n\rightarrow w}$. In this narrow-to-wide configuration we observe an interesting result. As can be seen in the left panel of Fig. 4 there occurs an optimum for the channel passage at higher values of the channel boundary slope $\lambda$. Namely, the theoretical result for the MFPT for very high force strength ($g \rightarrow \infty$) exceeds the MFPT values for $g = 20$ (the curves cross at around $\lambda = 0.5$) and $g = 50$ (crossing at around $\lambda = 0.2$).

This crossover behavior with an optimal MFPT warrants some closer inspection, however. On the one hand it is

![FIG. 2. Top: Effective diffusion coefficient as function of $x$ and $\lambda$ as predicted by Eqs. (4) and (7) for a 2D symmetrical cone-shaped channel formed by straight boundaries $h_1 = -\lambda x - 0.1$ and $h_2 = \lambda x + 0.1$. The channel width variation is given by $w(x) = h_2(x) - h_1(x)$ and $w'(x) = 2\lambda$. The surface graphs from top to bottom correspond to $g = 0, 5, 10, 20, \infty$ (almost coinciding with the result for $g = 20$, respectively). Bottom: Plot of the effective diffusion coefficient for fixed position $x = 1$ as function of $\lambda$. For $g = 10, 20, \infty$ the curves are almost indistinguishable.](image)
physically reasonable to argue that such a minimal MFPT at intermediate $g$ values is the result of the two opposing effects relevant for higher $g$. Namely, while moderate values of $g$ effect a resulting drift towards the wide channel exit, when $g$ gets too high it prevents the particle from exploring the channel in the perpendicular y direction. As the MFPT depends on the exact form of the diffusivity, we can use this quantity to gain some insight for the theoretically counterintuitive behavior when $g \to \infty$. In this limiting case $D(x) = D_0/[1 + h^2(x)]$ [20], where any information relating to the external force is missing. Comparison with the simulations therefore points out a limitation of the theory in this extreme limit. This is one of the objectives of this study. The particle therefore cannot profit from the entropic force pushing it towards the wider channel end. From this observation we can appreciate the importance of the conspirative interplay between the transverse external field and the entropic potential. We note that this interplay is hardly noticed in the right panel of Fig. 4 in which the MFPT is depicted as a function of $g$, for which only moderate $g$ values are shown. On the other hand, it remains unclear whether the approximations used here to obtain the effective one-dimensional description with effective entropic forcing remains valid in the limit $g \to \infty$. Concurrently, we cannot use our computer simulations to explore the true $g \to \infty$ limit, as the necessary time steps become prohibitively short with increasing $g$.

From a physical vantage the stark difference in behavior between the plotted case $g = 10^5$ and $g \to \infty$ opens up the possibility of a discontinuous transition of the MFPT dynamics at $g \to \infty$. Moreover, from a practical point of view this interesting crossover behavior represents new possibilities for controlling the transport of Brownian particles in narrow confined structures for a range of potential applications, including particle separation, fluid mixing, gating, and catalysis, among others.
IV. SUMMARY AND CONCLUSIONS

We studied the crossing dynamics of diffusing point particles of expanding and narrowing 2D conical channels under the action of a transverse external gravitational field by means of the MFPT. We derived the theoretical expression for the MFPT under the assumption that the axial diffusion in the 2D channel with its smoothly varying geometry can be approximately described as a one-dimensional diffusion in an entropic potential with position-dependent effective diffusivity in framework of the modified Fick-Jacob equation. To this end we use the theoretical expression by Pompa-García and Dagdug [20] for the interpolation of the effective diffusivity, $D_0/[1+(1/4)\eta w^2(x)]^{-G/(w+y)}$, where spatial confinement, asymmetry, and the presence of a constant transverse force can be encoded in $\eta$, as a function of the channel width $w$, channel midline $y_0$, and transverse force $G/(g/k_BT)$. This expression explicitly shows the coupling between the entropic and energetic effects.

We found very good agreement between the approximate theoretical result for the MFPT in the two possible configurations: channel passage from the narrow to the wide end and vice versa. While some deviations are observed for the wide-to-narrow case at intermediate channel boundary slopes and larger $g$ values, almost perfect agreement is observed for the narrow-to-wide case. Despite these deviations the general predictions of the approximate 1D description in terms of the modified Fick-Jacob equation is validated for this setting.

A remarkable effect is observed in the narrow-to-wide configuration, where the theoretical result for the MFPT is not monotonically decreasing with growing $g$, and thus not bounded by the limiting case $g \to \infty$. Instead, the MFPT assumes a minimum at intermediate $g$ values for larger values of the boundary slope $\lambda$. We interpret this result as an optimum in the interplay between the effective drift exerted by the entropic potential of the channel walls (widening towards the channel exit) and a pinning down of the particle to the channel wall by very high external forcing. In this case the effect of the entropic force vanishes, and the resulting MFPT increases. The fact that we can control the exit time of a Brownian particle from a 2D channel in the presence of an external transverse force may allow the development of practical applications including particle separation, gathering, controlling effective fluid mixing, and catalysis, among others. However, even though the theoretical expressions can be used to predict this crossover behavior of the MFPT values under the influence of large external forces, their full range of applicability remains somewhat tricky to establish. The theoretical affirmation is based on the fact that we are trying to predict the behavior of a two-dimensional system while using an effective diffusivity obtained by means of a dimensional reduction, that removes the degree of freedom, which coincides with the direction of the applied force. Moreover, the effective diffusivity model, and as a consequence, the expressions for the MFPT, does not contain any information about the direct interaction between the boundary walls and the particle further than its role as a boundary condition. Concurrently, while for moderate to large $g$ values the simulations tend to be close to numerical results, simulations of confined particles under a very high potential is subtle to implement because the conditions of the system require a small time step and an appropriate particle-wall interaction, involving a high computational cost. Thus, while this effect is physically interesting and, to our knowledge, reported for the first time, further research is needed to exactly establish the precise quantitative behavior.

Here we considered a single-particle picture for diffusion in the channel. This is an appropriate choice for low concentrations of tracer particles. At higher concentrations, particle particle will become relevant. Adding such effects will offer new perspectives to the concepts developed here, which will be the focus of future work. It will also be interesting to verify the effect of an external force perpendicular to the symmetry axis of the channel in 3D settings as well as for channels filled with complex liquids, e.g., when the particle exhibits viscoelastic subdiffusion.

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

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