Diffusion through Permeable Interfaces: Fundamental Equations and their Application to First-Passage and Local Time Statistics

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The diffusion equation is the primary tool to study the movement dynamics of a free Brownian particle, but when spatial heterogeneities in the form of permeable interfaces are present, no fundamental equation has been derived. Here we obtain such an equation from a microscopic description using a lattice random walk model. The sought after Fokker-Planck description and the corresponding backward Kolmogorov equation are employed to investigate first-passage and local time statistics and gain new insights. Among them a surprising phenomenon, in the case of a semi-bounded domain, is the appearance of a regime of dependence and independence on the location of the permeable barrier in the mean first-passage time. The new formalism is completely general: it allows to study the dynamics in the presence of multiple permeable barriers as well as reactive heterogeneities in bounded or unbounded domains and under the influence of external forces.

Random movement is ubiquitous, appearing in many physical, biological and social systems, and is traditionally modelled by diffusion in a homogeneous environment. But, in realistic systems the homogeneity of the environment is often interspersed by spatial heterogeneities that interfere significantly with diffusive transport. In many instances these heterogeneities are due to the presence of permeable interfaces, often referred to as semi or partially permeable barriers. They appear at microscopic scales in different porous media such as biological tissue \cite{1,6}, but also at larger scales when whole organisms interact with chemical or physical cues \cite{7,9}.

Cell biology is replete with examples of permeable structures whose function is to regulate the flux of biochemical substances between different spatial regions \cite{10}. In magnetic imaging techniques the diffusion of water molecules through different cellular compartments is exploited to understand physiological and anatomical properties of the human body \cite{11,12}. The lateral movement of molecules within the bilayer plasma membrane of eukaryotic cells is inhibited by the formation of submicron compartments due to anchored-transmembrane proteins and other macromolecules bound to the underlying actin-based cytoskeleton network \cite{13}. Permeability is also of relevance to ecology where animal dispersal is affected by the heterogeneity of the landscape e.g. the type of habitat \cite{14,15} or the presence of roads and fences \cite{16}.

Various theoretical approaches to study diffusion through permeable interfaces have been proposed in the past: Green’s functions in discrete \cite{17,19} and continuous space \cite{20,23}, spectral decompositions \cite{22,24} and scattering techniques \cite{25}. These techniques, whilst valuable, have been limited in their scope as they either demand spatial symmetries, e.g. analytical Green’s functions, or employ a coarse-grained representation of the heterogeneities, e.g. effective medium approximations.

In addition, these various approaches have failed to construct a unified framework capable of representing the diffusive dynamics with both permeable and reactive heterogeneities and to derive important quantities, such as first-passage and local time (or other Brownian functionals) statistics (i.e. through a backward Fokker-Planck representation). Given the wide-spread occurrence of permeable membranes, the above limitations call for the development of a fundamental theory of diffusion through permeable interfaces.

In this letter we aim to provide such theory through a fully analytic treatment of the problem. Firstly we show how the permeable boundary condition arises from microscopic considerations in a simple unbiased lattice random walk model. Such model allows us to derive an inhomogeneous diffusion equation (DE), where the inhomogeneity accounts for the presence of a porous barrier. Extensions to the general case of finite domains and when an external force is present are also provided. As applications of our formalism we study explicitly first-passage and local time statistics of diffusion with a permeable barrier.

**Theoretical derivation:** We consider a nearest-neighbour unbiased random walker on an infinite 1D lattice. The jump rate of the random walk between neighbouring sites equals $F$ except between the lattice points $r$ and $r+1$ where the rate is $f$ with $F > f$. The Master equation that represents the dynamics of the occupation probability, $P_m(t)$, of the random walker at the $m$-th lattice point can be constructed as follows \cite{18},

$$
dP_m(t) = F[P_{m+1}(t) + P_{m-1}(t) - 2P_m(t)] - \Delta[P_{r+1}(t) - P_r(t)](\delta_{m,r} - \delta_{m,r+1}), \quad (1)
$$

where $\Delta = F - f$ accounts for a partially reflecting defect between the sites $r$ and $r+1$ and $\delta_{m,r}$ is the Kronecker
delta. With the help of the so-called defect technique \[20\] Eq. \[1\] is solved \[27\].

With the lattice spacing \( \alpha \to 0 \), we let \( m, r, f, F \) become infinitely large such that \( m\alpha \to x, r\alpha \to x_b, f\alpha \to \kappa, F\alpha^2 \to D \) and \( P_m(t)/\alpha \to P(x,t) \). That is \( P(x,t) \) is the probability density for a diffusing particle (with a diffusion coefficient \( D \)) to be at the spatial position \( x \) at time \( t \) with a barrier located at \( x_b \) whose permeability is \( \kappa \), with units of velocity. One can show \[27\] that \( P(x,t) \) in this case satisfies the (DE)

\[
\delta \left( x - x_b \right) \partial_x P(x,t) = \partial_t P(x,t), \tag{3}
\]

where \( \delta(x-x_b) \) is the derivative of the Dirac delta function. Let us introduce the free propagator of the DE, \( \mathcal{G}_0(x|t|x_0) = \exp\left\{ -(x-x_0)^2/4Dt \right\} / \sqrt{4\pi Dt} \). The solution of Eq. \[3\] with the localized initial condition \( P(x,0) = \delta(x-x_0) \), is given in the Laplace domain (for any function \( f(t) \), \( \tilde{f}(\epsilon) = \int_0^\infty f(t)e^{-\epsilon t}dt \) by \[27\]

\[
\tilde{P}(x,\epsilon|x_0) = \mathcal{G}_0(x,\epsilon|x_0) = \frac{1}{\sqrt{2\pi\epsilon}} e^{-\epsilon|x-x_0|^2/2}.
\]

In Eq. \[1\] we have used the notation \( P(x,t|x_0) \) to indicate the localized initial condition and \( J_0(x,t|x_0) = -D\partial_x \mathcal{G}_0(x,t|x_0) \) is defined as the free probability current \( \left( \partial_x \mathcal{J}_0 \right) = \frac{\partial}{\partial y} h(x) \mid_{y=x} \) for a generic function \( h(x) \). By inserting the correct propagator and its current into Eq. \[4\], one recovers the solution of the DE with the PBC \[2\].

It is instructive to look at the moments of \( P(x,t|x_0) \) i.e. \( \langle x^n(t) \rangle = \int_{-\infty}^{\infty} x^n P(x,t|x_0)dx \). Using Eq. \[3\] we find the following equations for the first and second moments. \[4\] \( \frac{d}{dt} \langle x(t) \rangle = -DJ(x_b,t)/\kappa \) and \[4\] \( \frac{d}{dt} \langle x^2(t) \rangle = 2D - 2x_b D J(x_b,t)/\kappa \) respectively. As \( J(x_b,t) \) is readily obtained from Eq. \[4\], these equations are solved by

\[
\langle x(t) \rangle = x_0 - \text{sgn}(x_b-x_0)\frac{D}{2\kappa} \beta(t) \tag{5}
\]

and

\[
\langle x^2(t) \rangle = 2Dt + x_0^2 - \text{sgn}(x_b-x_0)\frac{Dx_b}{\kappa} \beta(t), \tag{6}
\]

where \( \text{sgn}(z) \) is the sign function and

\[
\beta(t) = \text{erfc} \left\{ \frac{|x_0 - x_b|}{2\sqrt{Dt}} \right\} - \exp \left\{ \frac{2\kappa}{D} (|x_0 - x_b| + 2\kappa t) \right\} \text{erfc} \left\{ \frac{|x_0 - x_b| + 4\kappa t}{2\sqrt{Dt}} \right\}. \tag{7}
\]

Here \( \text{erfc}(z) = 1 - \text{erf}(z) \) with \( \text{erf}(z) \) the error function. In the limit of \( \kappa \to \infty \) and \( \kappa \to 0 \) Eqs. \[6\] and \[7\] tend to their counterparts for free diffusion and diffusion with a perfectly reflecting boundary, respectively. As \( \lim_{\kappa \to \infty} \beta(t) = 1 \), the mean reaches a stationary value, \( \lim_{\kappa \to \infty} \langle x(t) \rangle = x_0 - \text{sgn}(x_b-x_0)D/(2\kappa) \). In Fig. \[1\] we use Eqs. \[6\] and \[7\] to plot the mean square displacement (MSD) \( \nu(t) = \langle (x(t) - \langle x(t) \rangle)^2 \rangle \). The curves clearly show that the presence of the permeable barrier reduces the magnitude of the MSD for short times, yet at long times the \( 2D \) term is dominant and we have the standard diffusive linear increase.

We now rewrite Eq. \[3\] in the following form, \( \partial_t P(x,t) = L_x P(x,t) \), where \( L_x \) is a linear differential operator with respect to \( x \). To proceed, we exploit the property \( \delta'(x-x_b)J(x,t) = \delta(x-x_b)\partial_x J(x,t) + \delta'(x-x_b)J(x,t) \), and the definition of \( J(x,t) \), to write \( L_x = (D^2/\kappa)\partial_x \delta'(x-x_b) + \partial_x^2[D - (D^2/\kappa) \delta(x-x_b)] \). The operator, \( L_x \), corresponds to the one in the following Fokker-Planck equation (FPE) \[32\],

\[
\frac{\partial}{\partial t} P(x,t) = -\frac{\partial}{\partial x} \left[ A(x) P(x,t) \right] + \frac{\partial^2}{\partial x^2} \left[ B(x) P(x,t) \right], \tag{8}
\]

with \( A(x) = -(D^2/\kappa)\delta'(x-x_b) \) and \( B(x) = D - (D^2/\kappa) \delta(x-x_b) \). Through this description we see that the presence of a permeable barrier can be described by an infinitely large positive potential, \( (D^2/\kappa) \delta(x-x_b) \), that pushes away the Brownian particle from \( x_b \), and by a diffusion coefficient that is modified at the interface, becoming infinitely negative thereby trapping the particle instead of dispersing it.

Though standard techniques allow to relate the underlying Langevin equation corresponding to Eq. \[8\], the
appearance of the Dirac delta function and its derivative would render such exercise of little practical use. Instead, we use the FPE to find the corresponding backward (Kolmogorov) FPE. In terms of $L$, the backward FPE is $-\partial_{\epsilon}P(x_0,t_0) = L^\dagger x_0 P(x_0,t_0)$, where $L^\dagger$ is the formal adjoint of $L$, i.e. $L^\dagger x_0 = A(x_0)\partial x_0 + B(x_0)\partial^2 x_0$. Note that this equation is now in terms of $x_0$ and $t_0$, where $t_0 < t$. The adjoint is then $L^\dagger x_0 = -((D^2/\kappa)\delta(x_0 - x_b)\partial x_0 + [D - (D^2/\kappa)]\delta(x_0 - x_b)]\partial^2 x_0$, meaning $L$ is self-adjoint.

First-passage processes: Using the backward FPE we study the process in the presence of a perfectly absorbing point at $x_a$ to the left or right of both $x_0$ and $x_b$. Note, if this absorbing boundary is placed at the same point as the permeable barrier, $x_a = x_b$, a radiation boundary \cite{33,34} is recovered \cite{27}. Defining the survival probability as $S(t|x_0) = \int_{-\infty}^{\infty} P(x,t|x_0)dx$ or $S(t|x_0) = \int_{-\infty}^{x_0} P(x,t|x_0)dx$, respectively, for $x_0 < x_b < x_c$ or $x_a < x_b < x_0$. Taking $t_0 = 0$, exploiting the time homogeneity of the process and utilising the self-adjoint nature of $L$, we find that for $S(t|x_0)$,

$$\frac{\partial S(t|x_0)}{\partial t} = D \frac{\partial^2 S(t|x_0)}{\partial x_0^2} - \frac{D^2}{\kappa} \delta(x_0 - x_b) \frac{\partial S(t|x_0)}{\partial x_0} \bigg|_{x_0=x_c},$$

Eq. \ref{eq:9} is supplemented by the initial condition, $S(0|x_0) = 1$ and the Dirichlet boundary conditions (BC), $S(t|x_c) = 0$ and $\lim_{x_0 \to \pm \infty} S(t|x_0) = 1$. Using the free propagator, we satisfy the Dirichlet BC via $G(x,t|x_0) = G_0(x,t|x_0) - G_0(x,t|2x_c-x_0)$ \cite{36} and write the solution to Eq. \ref{eq:9} as

$$\tilde{S}(\epsilon|x_0) = \tilde{S}_0(\epsilon|x_0) + \theta_{x_0} \tilde{S}_0(\epsilon|x_b) - \frac{\partial_x \tilde{G}(x_b,\epsilon|x_0)}{\partial t} - \frac{\partial_x \tilde{G}(x_b,\epsilon|x_b)}{\partial t}.$$  

(10)

where the free survival probability (i.e. for $\kappa = \infty$) is $\tilde{S}_0(t|x_0) = \text{erf} (|x_c-x_0|/\sqrt{4Dt})$ \cite{36}. From $\tilde{F}(x_c,\epsilon|x_0) = 1 - \epsilon \tilde{S}(\epsilon|x_0)$, we obtain the Laplace transform of the first-passage probability (FPP) distribution (see Ref. \cite{27} for the expression for when $x_0$ is between $x_b$ and $x_c$),

$$\tilde{F}(x_c,\epsilon|x_0) = \frac{2\kappa e^{-|x_c-x_0|\sqrt{t}}}{\sqrt{4Dt}}. $$

(11)

Through Tauberian theorems \cite{37} we find the long time dependence of the FPP distribution as

$$\mathcal{F}(x_c,t|x_0) \approx \frac{|x_c-x_0| + D/\kappa}{\sqrt{4\pi Dt^3}}.$$  

(12)

Eq. \ref{eq:12} shows that the FPP distribution possesses the same $t^{-3/2}$ asymptotic dependence as free diffusion but the coefficient includes the additional term $D/\kappa$. In Fig. \ref{fig:2} we draw Eq. \ref{eq:11} to show the full time dependence, while the inset shows the non-linear dependence of the magnitude of the mode of the distribution, $M$, as a function of the barrier position relative to $x_c$.

![Figure 1](image1.png)

**FIG. 1.** (Color online) The MSD, $\nu(t)$, as a function of time for a Brownian particle initially placed at the origin, in the presence of a permeable barrier, with permeability $\kappa$, placed at $x_b$, for different values of the scaled permeability parameter $x_c\kappa/D$. An infinite permeability indicates the absence of a barrier and a zero permeability indicates a fully reflecting barrier. Inset: corresponding long time behaviour of the MSD.

![Figure 2](image2.png)

**FIG. 2.** (Color online) The FPP distribution of a Brownian particle, $\mathcal{F}(x_c,t|x_0)$, is computed via numerical inversion \cite{38} of Eq. \ref{eq:11} for different values of the scaled permeability parameter, $x_c\kappa/D$. The particles starting location is $x_0$ with $x_0/x_c = -1$ and a permeable barrier is placed in between $x_0$ and $x_c$ at the origin. Inset: magnitude of the modal peak of the FPP distribution, $M$, plotted against different scaled barrier positions, $x_b/x_c$, with $x_0\kappa/D = -1$. To gain further understanding of the impact a permeable barrier has on the dynamics of a Brownian particle, we study the mean first-passage time (MFPT) to $x_c$, $\tau(x_0) = \int_0^\infty t\mathcal{F}(x_c,t|x_0)dt$. Since, the MFPT
of a Brownian particle is infinite in a semi-infinite domain, we add a perfectly reflecting boundary at \(x_r\), such that the permeable barrier lies between \(x_r\) and \(x_c\). As 
\[
\tau(x_0) = \int_0^\infty S(t|x_0)dt,
\]
from Eq. 9 we have 
\[
-1 = D\tau''(x_0) - \frac{D^2}{\kappa} \delta'(x_0 - x_b)\tau'(x_b), \tag{13}
\]
where \(\tau'(x_0) = \frac{d}{dx_0}\tau(x_0)\). Eq. 13 is then supplemented by the Dirichlet and Neumann BC, \(\tau(x_r) = 0\) and \(\tau'(x_c) = 0\), respectively. Eq. 13 is solved to give
\[
\tau(x_0) = \left\{ \begin{array}{ll}
\frac{x_0^2 - x_b^2 + 2x_b(x_0 - x_b)}{2D} + \frac{|x_0 - x_c|}{\kappa}, & x_0 \in [x_r, x_b), \\
\frac{x_0^2 - x_b^2 + 2x_b(x_0 - x_b)}{2D}, & x_0 \in (x_b, x_c].
\end{array} \right.
\tag{14}
\]
Eq. 14 shows the interesting feature that when the barrier is not placed between \(x_0\) and \(x_c\), the MFPT is identical to the barrier free case. Yet when the barrier is placed between \(x_0\) and \(x_c\), the impact of the MFPT is merely the addition of a term dependent on the position of the barrier that is scaled by the strength of its permeability. To clarify this aspect we may split the contributions to \(\tau(x_0)\) between those trajectories that travel to \(x_c\) without returning to \(x_0\) and those that do return. The permeable interface clearly has no effect on the former trajectories as \(x_c\) does not lie between \(x_0\) and \(x_c\). For the latter trajectories, the particle may return to \(x_0\) multiple times before directly travelling to \(x_c\) from \(x_b\). Since the mean return time for an unbiased Brownian particle to any point is only dependent on the overall domain size \[39\], the presence of a permeable interface will have no impact on these trajectories either.

Local time: Returning to the backward FPE, we can study the probability distribution of various functionals of Brownian motion. One of interest is the so-called local time of Brownian motion, defined as \(\ell_t = \int_0^t \delta(x(t') - a)dt'\), which characterises the amount of time a Brownian particle spends at a given point \(a\) \[40\]. We seek the probability density describing the random variable, \(\ell_t\), namely the local time distribution (LTD), \(\rho(\ell, t|x_0)\), of a Brownian particle in the presence of a permeable barrier. To do so we take the Laplace transform of the LTD with respect to \(\ell\), i.e. \(\rho(p, t|x_0) = \int_0^\infty e^{-\ell p}\rho(\ell, t|x_0)\,d\ell\). Such quantity may be written in terms of a conditional expectation \[41\],
\[
\rho(p, t|x_0) = \langle \exp\left\{-p \int_0^t \delta(x(t') - a)\,dt'\right\} x(0) = x_0 \rangle,
\]
where the expectation is over all trajectories of the particle starting at \(x(0) = x_0\) up to time \(t\). Through the Feynman-Kac formula \[42\] \[43\], \(\rho(p, t|x_0)\) satisfies the following 
\[
\frac{\partial \rho}{\partial t} = A(x_0) \frac{\partial \rho}{\partial x_0} + B(x_0) \frac{\partial^2 \rho}{\partial x_0^2} - p\delta(x_0 - a)\rho, \tag{15}
\]
where \(A\) and \(B\) are defined after Eq. 8. Eq. 15 is supplemented by the initial condition, \(\rho(p, 0|x_0) = 1\), and the BC, \(\rho(p, t|x_0 \to \pm \infty) = 1\) \[44\]. By treating the last term on the RHS of Eq. 15 as an inhomogeneity, it is straightforward to construct the general solution via the solution of the homogeneous equation (i.e. for \(p = 0\)). For a localized initial condition, the solution of the homogeneous part is equivalent to the solution of Eq. 8 through Eq. 1. The Laplace transform of the solution of Eq. 15 is thus 
\[
\bar{\rho}(p, \epsilon|x_0) = \frac{1}{\epsilon} \left[ 1 - \frac{\bar{P}(a, \epsilon|x_0)}{\bar{P}(a, \epsilon|a)} \right]. \tag{16}
\]
Considering that we have a permeable barrier in an unbounded domain, we exploit the translational invariance of the problem and set \(x_b = 0\) and calculate the LTD at the barrier, that is \(a = x_b\). Recalling the PBC \[2\], we need to distinguish whether we are looking at \(x_b^+\) or \(x_b^-\). Furthermore, let us consider the case \(x_b = 0^+\) and \(x_0 = 0^+\); using Eq. 16 and after inverse Laplace transforming with respect to \(p\), we find the barrier LTD to be 
\[
\bar{\rho}(\ell, \epsilon|0^+) = \frac{2\sqrt{D\pi\ell+D\kappa}}{\epsilon} \exp\left\{-\frac{(2\kappa\sqrt{D\pi\ell+D\kappa})^2}{\epsilon}\right\}. \tag{17}
\]
The limit \(\lim_{\epsilon \to 0} \bar{\rho}(\ell, \epsilon|0^+) = 0\) shows that Eq. 17 has no steady state distribution at long times, indicative of the unbounded nature of the dynamics. In the limit \(\kappa \to \infty\) we recover the barrier free distribution, \(\rho(\ell, t|0) = 2\sqrt{D/\pi} e^{-\ell^2/4D}\) and for \(\kappa \to 0\) we obtain the perfectly reflecting distribution, \(\rho(\ell, t|0) = \sqrt{D/\pi} e^{-\ell^2/4D}\) \[45\]. From Eq. 17 we can also find the mean, 
\[
\langle \ell_t \rangle = \frac{1}{4\kappa} \left[ 1 - e^{-\frac{2\kappa}{\sqrt{\pi\ell}}} \text{erfc} \left\{ \frac{2\kappa \sqrt{\frac{\tau}{\ell}}} {\sqrt{\pi}} \right\} \right] \tag{18}
\]
At long times the mean local time at the barrier is dominated by the final term on the RHS of Eq. 18, i.e. \(\langle \ell_t \rangle \sim t^{1/2}\), as in the barrier free case. A comparison of the temporal dependence of the mean local time, \(\langle \ell_t \rangle\), for different values of permeability, is displayed in the inset of Fig. 3. The unbounded nature of its long time dependence can also be evinced from the main plot of Fig. 3, which shows the flattening of the LTD as time increases.

External forces: We have shown so far the applications of our formalism to situations where no external forces are present. However the formalism is completely general and may include the dynamics in the presence of a potential, \(U(x)\), in some domain \(x \in \Omega\). In this case the ‘homogeneous’ system is described by the Smoluchowski equation (SE) \[46\],
\[
\partial_t P(x, t) = \partial_x \left[ U'(x) P(x, t) \right] + D \partial^2_x P(x, t). \tag{19}
\]
where the probability current is now \( J(x,t) = -U'(x)P(x,t) - D\partial_x P(x,t) \). Let us call the propagator of the SE, \( G_0(x,t|x_0) \), which exists over \( \Omega \), with \( J_0(x,t|x_0) \) the barrier free counterpart of \( J(x,t) \). The solution of Eq. (15), with localized initial conditions, may be written as in Eq. (4). We are again able to transform Eq. (15) into the FPE (8), with \( A(x) = -U'(x)[1 - (D/\kappa)\delta'(x-x_b)] - (D^2/\kappa)\delta'(x-x_b) \) and \( B(x) = D - (D^2/\kappa)\delta(x-x_b) \), and then construct the analogous of Eqs. (9), (13) and (15) in the presence of a potential.

In summary, we have derived an inhomogeneous form of the DE and SE to account for the presence of a permeable barrier. We have used the former to investigate first-passage and local time statistics of a Brownian particle through the construction of a backward FPE. Explicit analytic dependence of the LTD and FPP distribution and their respective means have also been presented. Due to the linearity of the problem, our methods readily extend to the case of multiple permeable interfaces by appending the inhomogeneity for each interface position to Eq. (15). Reactive heterogeneities can be accounted for in Eq. (15) via the standard defect technique (26). Future directions include the extension of these methodologies to higher dimensions and the application of our formalism to anomalous diffusion (47).

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FIG. 3. (Color online) The barrier local time distribution for \( \kappa = 0.1 \) and \( D = 1 \) (in arbitrary units), computed via a numerical inverse Laplace transform of Eq. (17) and plotted against \( \ell \) at different times, \( t = 1, 5, 10, 15 \), respectively. Inset: the mean barrier local time, Eq. (18) plotted over a time window, for varying permeability values, \( \kappa = \infty, 1, 0.1, 0.01, 0 \) (in arbitrary units), represented by the markers: circular, cross, square, diamond and no marker, respectively. For \( \kappa \to \infty \), we have the barrier free mean local time, \( \sqrt{t/\pi D} \) and for \( \kappa \to 0 \), we have the perfectly reflecting barrier mean local time, \( 2\sqrt{t/\pi D} \).
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