Short-Path Statistics and the Diffusion Approximation

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In the field of first return time statistics in bounded domains, short paths may be defined as those paths for which the diffusion approximation is inappropriate. This is at the origin of numerous open questions concerning the characterization of residence time distributions. We show here how general integral constraints can be derived that make it possible to address short-path statistics indirectly by application of the diffusion approximation to long paths. Application to the moments of the distribution at the low-Knudsen limit leads to simple practical results and novel physical pictures.

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We consider here a geometric system Ω of volume V with a boundary $\partial\Omega$ of surface S. Particles enter Ω uniformly and isotropically through $\partial\Omega$ and move across Ω along a trajectory of length l until their first exit through $\partial\Omega$. The random variable corresponding to the statistics of l is denoted L. It was shown in Ref. [1] that, for a three dimension diffusion random walk, the mean value of L is

$$\langle L\rangle = \frac{4V}{S} \tag{1}$$

independent of the random walk characteristics. $\langle L\rangle$ is therefore a purely geometric quantity: It is not modified when changing the fields of the local mean free path [$\lambda(\mathbf{x})$ at location $\mathbf{x}$ for the average of the exponentially distributed paths between successive scattering events] and the single scattering phase function [the probability density function $p(\mathbf{u_s}, \mathbf{u_i}, \mathbf{x})$ of the scattering direction $\mathbf{u_s}$ for an incident direction $\mathbf{u_i}$ at $\mathbf{x}$]. Practical uses of this invariance property are reported in fields such as biology [2–4], colloid physics [5], radiative transfer [1,6,7], and neutronics [8]. It was also theoretically reexamined in the field of integral geometry as an extension of Cauchy's formula [9,10]. Various further extensions were then proposed in Ref. [11]: General diffusion processes were considered, including Hamiltonian forces, and the property was generalized to mean residence times inside subdomains with reflecting boundary conditions. (The problem may indifferently be considered in terms of average trajectory lengths or average residence times using the phase space average of the velocity module.) Attempts were also made to use such invariance properties for further characterization of the trajectory length distribution by the establishment of quantitative relationships with the statistics of particles starting uniformly inside the volume [11,12].

These theoretical derivations are directly related to the field of first return time statistics (here the time of first return to the system boundary). Such statistics can be simply addressed in only two particular cases: when the system geometry is simple (numerous analytical solutions are available for various types of one dimension random walks) or when the mean free path is large compared to the system extension (even for complex geometries, the statistics can be simplified by considering a limited number of diffusion events). But for complex geometries and small mean free paths, no systematic practical approach is available. The main reason is that the diffusion approximation is inaccurate for first return time problems: It can be used for long paths (particles exiting the system after a large number of diffusion events), but it is meaningless for an always significant number of particles that are reflected out of the system after a few diffusion events (short paths). This specific feature of first return time statistics is at the origin of an extensive literature in which attempts are made to identify other physical approximations for use in complex geometries or to find indirect ways of accurately using the diffusion approximation in spite of short-path statistics [13–17].

In the present Letter, all formal derivations are limited to a strict diffusion random walk, and generalization to other types of statistical processes (including nonexponentially distributed free paths) is discussed at the end. Our starting point is the following simple conjecture: When changing the characteristics of the random walk, the corresponding changes of long-path statistics are strictly compensated by those of short-path statistics in order to ensure the invariance property identified in Ref. [1]; this compensation can therefore be used to get quantitative information on short paths by application of the diffusion approximation on long paths.

Methodology.—The whole question is to find practical ways to separate short paths and long paths in order to apply the diffusion approximation on long paths only. We believe that a useful solution is provided by the statistical relation between the above defined path lengths l of first exit trajectories starting uniformly and isotropically at the boundary [the random variable is denoted L and the probability density function is $p_L(l)$] and path lengths r of first exit trajectories starting uniformly and isotropically within the system [the random variable is denoted R and the probability density function is $p_R(r)$]. It was shown in Ref. [12] that p_L and p_R verify

$$p_L = -\langle L\rangle p_R', \tag{2}$$

where $\langle L\rangle$ is given by Eq. (1). This relation is a direct consequence of the fact that each trajectory of length l from the boundary back to the boundary can be used to define a continuous set of trajectories of length $r \in [0, l]$ from inside the system to the boundary (see Fig. 1) and that the starting points of such R trajectories are distributed uniformly within phase space. Obviously, low values of r correspond to low and high values of l, but high values of r correspond only to high values of l. Addressing long-path statistics can therefore be performed in terms of R. But with R's statistics, although still a first passage time problem, we are not dealing with a first return time problem any more and the diffusion approximation can be used with confidence: When reducing the mean free path λ compared to the system size, the probability that a R trajectory occurs with a small number of diffusion events tends to zero (which is not true for L trajectories).

As a consequence of Eq. (2), the average value of any function $f(l)$ may be written

$$\langle f(L)\rangle = \int_0^{+\infty} p_L(l)f(l)dl = \int_0^{+\infty} -\langle L\rangle p_R'(l)f(l)dl, \tag{3}$$

and, when the limits $(p_R f)_0 = \lim_{x\to 0} p_R(x)f(x)$ and $(p_R f)_{+\infty} = \lim_{x\to+\infty} p_R(x)f(x)$ are defined,

$$\langle f(L)\rangle = \langle L\rangle\left[\int_0^{+\infty} p_R(r)f'(r)dr + (p_R f)_0 - (p_R f)_{+\infty}\right]. \tag{4}$$

Applying this relation to the particular case of $f = 1$ leads to $p_R(0) = \frac{1}{\langle L\rangle}$. If R^{diff} is the random variable corresponding to the diffusion approximation applied to R, we get the following approximation for $\langle f(L)\rangle$:

$$\langle f(L)\rangle \approx \langle L\rangle\langle f'(R^{\text{diff}})\rangle + f_0 - \langle L\rangle(p_R f)_{+\infty}. \tag{5}$$

Therefore, for all functions that ensure $(p_R f)_{+\infty} = 0$, as $\langle L\rangle$ is known, $\langle f(L)\rangle$ can be estimated by application of the diffusion approximation on R even for cases where short L trajectories play an essential part. This means that for small mean free paths, in any confined geometry, the statistics of L can be characterized by application of standard techniques of statistical physics on R, despite all of the difficulties associated with short paths in first return time statistics.

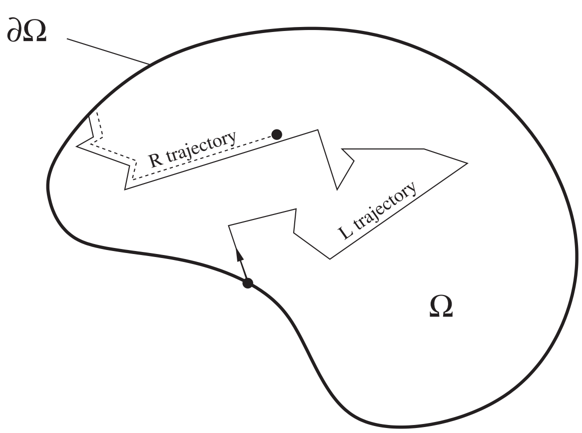

FIG. 1. L trajectories (from the boundary to the boundary) and R trajectories (from inside the system to the boundary).

Application.—We show hereafter that a first simple application of this methodology leads to the conclusion that the nth moment of L scales as the $(n-1)$th exponent of $k = 1/\lambda^*$, where λ^* is the transport mean free path (λ^* reduces to the mean free path λ in the particular case of isotropic diffusion). If λ^* is homogeneous, this simply means that

$$\langle L^n\rangle \approx \alpha_n k^{n-1}, \tag{6}$$

where α_n is a purely geometric quantity: It does not depend on the random walk characteristics. The case $n = 1$ is only a repetition of Eq. (1). $\langle L\rangle$ is constant and $\alpha_1 = \frac{4V}{S}$ in three dimensions. But $\langle L^2\rangle$ now appears as proportional to the inverse of the transport mean free path, $\langle L^3\rangle$ is quadratic, etc. A first comment on this property is that it highlights the contribution of short paths. If only long L trajectories were to contribute to $\langle L^n\rangle$, the diffusion approximation could be directly applied on L and $\langle L^n\rangle$ would scale as k^n instead of k^{n-1}. By application of the diffusion approximation on R, we manage to quantitatively include the compensation of short and long L trajectories leading to a reduction of the exponent of k by 1.

Equation (6) and the expression of α_n are simply derived from Eq. (5) with $f(x) = x^n$ [ensuring $f_0 = 0$ and $(p_R f)_{+\infty} = 0$]:

$$\langle L^n\rangle \approx \langle L\rangle n\langle (R^{\text{diff}})^{n-1}\rangle. \tag{7}$$

[Note that our theoretical developments started with Eq. (16) in Ref. [12], but for the particular application to the moments of L we could have directly started from Eq. (13) in the same reference.] A strong property of diffusion random walks is that traveled distances scale as the inverse of the transport mean free path defined as $\lambda^*(\mathbf{x}) = \frac{\lambda(\mathbf{x})}{1-g(\mathbf{x})}$, where $g(\mathbf{x}) = \int_{4\pi} p(\mathbf{u_s}, \mathbf{u_i}, \mathbf{x})\mathbf{u_s}\cdot\mathbf{u_i}d\mathbf{u_s}$ is the asymmetry parameter of the scattering phase function ($g = 0$ for isotropic scattering). Quantitatively speaking, this property allows that $\langle (R^{\text{diff}})^{n-1}\rangle$ and therefore $\langle L^n\rangle$ be simply related to the solution of the diffusion equation with an initial density uniform within Ω and a density fixed to zero on $\partial\Omega$ as a boundary condition, the boundary flux being directly related to the temporal version of the R^{diff} distribution. In the particular case where λ^* is uniform,

$$\langle L^n\rangle \approx \alpha_n\left(\frac{1}{\lambda^*}\right)^{n-1}, \tag{8}$$

$$\alpha_n = \langle L\rangle n q^{n-1}\int_0^{+\infty} p^{\text{diff}}(\tau)\tau^{n-1}d\tau, \tag{9}$$

$$p^{\mathrm{diff}}(\tau) = \int_{\partial\Omega} \mathbf{u}.\nabla\rho|_{\mathbf{y},\tau} d\mathbf{y}, \tag{10}$$

where q is the problem dimension, $\mathbf{u}$ is the inward normal unit vector at the boundary, and $\rho(\mathbf{x},\tau)$ is the solution of $\frac{\partial\rho}{\partial\tau} = -\nabla^2\rho$, with $\rho(\mathbf{x},0) = \frac{1}{V}$ and $\rho(\mathbf{y},\tau) = 0$ for $y \in \partial\Omega$. Equation (8) leads to Eq. (6) with $k = 1/\lambda^*$, and Eqs. (9) and (10) demonstrate that α_n is a purely geometric quantity. Table I displays Monte Carlo simulations compared to analytical solutions of α_1 and α_2 for a slab geometry (three-dimensional diffusion random walk in a one-dimensional plane parallel geometry). Note that in the one-dimensional case (diffusion random walk in the unit interval) an analytical solution is available for $\langle L^n\rangle$ that is a polynomial function of k of order $n-1$ whatever the value of mean free path [19].

Corresponding physical pictures.—The fact that short paths impose that the nth moment of L scales as k^{n-1} (instead of k^n as a direct application of the diffusion approximation would indicate) is a consequence of Eq. (5), where $\langle f(L)\rangle$ is related to the average of the derivative of f applied to R^{diff}. Although mathematically quite simple, this relation is not physically intuitive. However, a simple physical picture can be built in terms of the diffusive nature of long paths, on one hand, and the probability that a L trajectory enters enough within Ω for the diffusion approximation to be meaningful, on the other hand. Let us define P_{int} as the probability that a particle starting at the boundary visits $\mathcal{I}_\delta$ before its return to the boundary, where $\mathcal{I}_\delta$ is the interior part of Ω located at a distance greater than δ from the boundary (see Fig. 2). $P_{\mathrm{ext}} = 1 - P_{\mathrm{int}}$ is the probability that the particle remains in the exterior part $\Omega - \mathcal{I}_\delta$. This exterior part may be locally regarded as a one-dimensional slab if δ is small compared to the characteristic size of Ω. The nth moment of L may then be seen as a weighted sum of the moments of the random variables L_{int} and L_{ext} corresponding, respectively, to the lengths of trajectories that have visited $\mathcal{I}_\delta$ and of those trajectories that remained in $\Omega - \mathcal{I}_\delta$:

$$\langle L^n\rangle = P_{\mathrm{int}}\langle L^n_{\mathrm{int}}\rangle + P_{\mathrm{ext}}\langle L^n_{\mathrm{ext}}\rangle. \tag{11}$$

The lengths of all L_{int} trajectories are greater than δ. In the limit $\lambda \ll \delta$, the diffusion approximation tells us that L_{int} scales as $1/\lambda^*$ and therefore $\langle L^n_{\mathrm{int}}\rangle$ is proportional to $(1/\lambda^*)^n$. It is the probability to cross the slab of thickness δ that reduces the exponent of $k = 1/\lambda^*$ by 1: Simple physical pictures such as the gambler's ruin tell us indeed that P_{int} is proportional to λ^*/δ. The content of Eq. (6) then becomes quite intuitive provided that we can further argue that $(P_{\mathrm{ext}}\langle L^n_{\mathrm{ext}}\rangle) \ll (P_{\mathrm{int}}\langle L^n_{\mathrm{int}}\rangle)$:

$$\langle L^n\rangle \approx P_{\mathrm{int}}\langle L^n_{\mathrm{int}}\rangle \sim \frac{\lambda^*}{\delta}\left(\frac{1}{\lambda^*}\right)^n \sim k^{n-1}. \tag{12}$$

But the fact that $P_{\mathrm{ext}}\langle L^n_{\mathrm{ext}}\rangle$ may be neglected is not obvious: Rapid references to physical pictures such as those of Brownian motion could lead us to an opposite conclusion. Indeed, when reducing λ^* compared to δ, the probability to cross the boundary without exiting the slab of thickness δ tends to unity ($P_{\mathrm{ext}} \approx 1$) and the random variables L and L_{ext} are very similar. In particular, for whatever length scale γ and whatever $\epsilon > 0$, the transport mean free path may always be reduced so that $|C_L(L) - C_{L_{\mathrm{ext}}}(l)| < \epsilon$, for $l \in [0,\gamma]$, where C_L and $C_{L_{\mathrm{ext}}}$ are the cumulative distribution functions of L and L_{ext}, respectively. This means that most trajectories exit the system without encountering the interior $\mathcal{I}_\delta$, and there is therefore no distinction between L and L_{ext} up to $l = \gamma$ that can be taken as large as required.

TABLE I. Monte Carlo estimations of $\langle L\rangle$ and $\partial_k\langle L^2\rangle$ for a one-dimensional slab of thickness δ. The derivative $\partial_k\langle L^2\rangle$ is directly computed using the Monte Carlo sensitivity estimation procedure introduced in Ref. [18]. The mean free path $\lambda = \frac{1}{k}$ is uniform, and scattering is isotropic. The number of sampled trajectories is 10^9. $\sigma\langle L\rangle$ and $\sigma_{\partial_k\langle L^2\rangle}$ are the corresponding statistical uncertainties. Equation (6) leads to $\partial_k^{n-1}\langle L^n\rangle \approx (n-1)!\,\alpha_n$, where α_n is provided by Eq. (9) with an analytical resolution of the diffusion equation ($\alpha_1 = 2\delta$ and $\alpha_2 = \delta^3$). Monte Carlo results are presented for increasing values of $k\delta$. $\langle L\rangle = \alpha_1$ is confirmed whatever the value of $k\delta$, which corresponds to the fact that Eq. (1) is exact. $\partial_k\langle L^2\rangle \approx \alpha_2$ is true only for large values of $k\delta$, which corresponds to the fact that Eq. (6) is valid only when λ is small compared to the size of the system. $\partial_k^{n-1}\langle L^n\rangle$ for $n > 2$ cannot be accurately estimated with Monte Carlo simulations, but with the proposed methodology it can be simply addressed by analytical resolution of the diffusion equation leading to $\alpha_3 = \frac{9}{10}\delta^5$ and $\alpha_4 = \frac{153}{140}\delta^7$.

$k\delta$	$\langle L\rangle/\alpha_1$	$\sigma_{\langle L\rangle}/\alpha_1$	$\partial_k\langle L^2\rangle/\alpha_2$	$\sigma_{\partial_k\langle L^2\rangle}/\alpha_2$
1	1.000 04	2.44×10^{-5}	0.3334	1.40×10^{-3}
2	0.999 98	2.22×10^{-5}	0.7522	1.07×10^{-3}
4	1.000 003	3.42×10^{-5}	0.9135	1.21×10^{-3}
6	0.999 97	4.05×10^{-5}	0.9578	1.52×10^{-3}
10	1.000 04	5.10×10^{-5}	0.9833	2.32×10^{-3}
15	0.999 92	5.12×10^{-5}	0.9884	3.55×10^{-3}

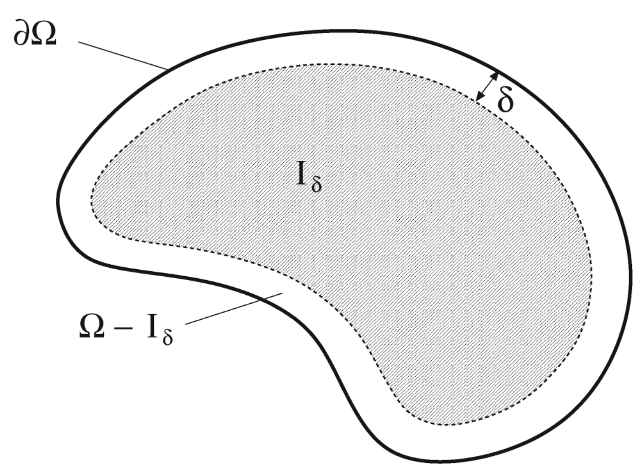

FIG. 2. Interior part $\mathcal{I}_\delta$ of Ω.

This could lead us to believe that $\langle L^n \rangle \approx P_{\rm ext}\langle L^n_{\rm ext}\rangle \simeq \langle L^n_{\rm ext}\rangle$. But even if trajectory lengths greater than γ are extremely unlikely, the distinction between L and $L_{\rm ext}$ on $[\gamma, +\infty[$ is always quantitatively essential to the present argument. The invariance property of Eq. (1) tells us indeed that $\langle L \rangle = \frac{4V}{S}$, whereas $(P_{\rm ext}\langle L_{\rm ext}\rangle) < 2\delta$ may be easily obtained by application of the same invariance property on the slab of thickness δ. Therefore, $P_{\rm ext}\langle L_{\rm ext}\rangle$ can be reduced by reducing δ compared to $\frac{4V}{S}$ and Eq. (11) leads to $\langle L \rangle \approx P_{\rm int}\langle L_{\rm int}\rangle$. Starting from Eq. (6), a simple dimensional analysis can be performed to conclude similarly that $(P_{\rm ext}\langle L^n_{\rm ext}\rangle) < \beta\delta^{2n-1}$, where β is independent of δ, and $P_{\rm ext}\langle L^n_{\rm ext}\rangle$ may be neglected to get Eq. (12). Altogether, the physical picture associated to $\langle L^n \rangle \approx \alpha_n k^{n-1}$ may be summarized as follows: (i) The diffusion approximation tells us that long paths scale as $1/\lambda^*$; (ii) the probability that a particle enters enough within Ω for the diffusion approximation to be valid scales as λ^*; (iii) particles that enter deep within Ω always play a dominant part in $\langle L^n \rangle$, even when $\lambda^* \to 0$, ensuring, in particular, that $\langle L \rangle = \frac{4V}{S}$ whatever λ^*. The same argument applies to other functions than $f(x) = x^n$ with the constraint that f has a finite limit f_0 in $x = 0$:

$$\langle f(L)\rangle \approx f_0 + P_{\rm int}\langle g(L_{\rm int})\rangle, \tag{13}$$

where $g(x) = f(x) - f_0$ ensures that g tends to zero in $x = 0$. As above, $P_{\rm int}$ scales as λ^* and $L_{\rm int}$ scales as $1/\lambda^*$, which means that $\langle f(L)\rangle$ may be interpreted as

$$\langle f(L)\rangle \approx f_0 + \xi\lambda^*\left\langle g\left(\frac{U}{\lambda^*}\right)\right\rangle, \tag{14}$$

where ξ is a constant and U is a random variable that depends only on the geometry of Ω and not on the characteristics of the random walk.

Generalization.—In this Letter, the discussion was restricted to diffusion random walks, that is to say, exponentially distributed straight paths between successive scattering events. Extension to any random walk compatible with the diffusion approximation is quite straightforward. The only subtlety is that Eq. (1) is valid only if a specific distribution is introduced for the travelled distance from the boundary to the first scattering event, so that the whole process is compatible with equilibrium. Introduction of reflective boundary conditions as well as Hamiltonian forces (inducing path curvature) can also be performed following the same approach as in Ref. [11]: Incident particles are distributed according to the corresponding equilibrium distribution, and the diffusion equation is replaced by equations accounting for the macroscopic effects of forces in the low-Knudsen limit.

But more significant is the fact that the physical pictures summarized in Eqs. (12) and (14) are independent of the incident distribution at the boundary. This allows for the methodology to be extended to a much wider class of physical applications. In particular, it can be extended to the case where particles are incident on a restricted part of the boundary, including point source particle imaging problems in any complex geometry. However, Eqs. (12) and (14) lead only to the scaling properties; they are not fully quantitative. Reasoning with I_δ does not provide any way to determine α_n, or ξ and U, from the resolution of the diffusion equation. This means that Monte Carlo simulations are required, and we have seen in Table I that strong convergence difficulties may be rapidly encountered. If we want to preserve the main feature of the proposed methodology (the fact that L can be accurately characterized on the basis of a simple resolution of the diffusion equation), then it is required that the extension start with Eqs. (2) and (5). We have checked that Eq. (2) can be rigorously extended to any incident distribution, and starting from Eq. (5) it was possible to derive a generalization of Eq. (9) (both extensions will be published elsewhere). But up to now there remains one practical limitation for fast application to complex geometries: As soon as the incident distribution is nonuniform and/or anisotropic, $\langle L \rangle$ is no longer given by the ratio $\frac{4V}{S}$ and one Monte Carlo run is still required for its quantification (but this is much less computationally extensive than the evaluation of α_n for large values of n).

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