Approach to equilibrium of particles diffusing on finite, curved surfaces with reflecting boundary conditions.

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Abstract

We present a simple analysis of the diffusion operator on any finite, curved surface given by the equation $\phi(\mathbf{r}) = 0$, with reflecting boundary conditions on the border of the domain $\mathbf{D} \subset \mathbf{R^3}$. The first non-vanishing eigenvalue of the diffusion operator is given by the long time decay rate of the effective diffusion coefficient for studied surface. This eigenvalue is the inverse of the mean time needed to reach uniform coverage of the surface by diffusing particles. We present here results for a variety of nodal surfaces like P, D, G, S, S1 and I-WP, where the domain $\mathbf{D}$ is the unit cell.

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1 Introduction

There is a growing interest in the study of diffusion on curved surfaces. This interest stems partially from experiments, where diffusional processes on curved membranes are studied by optical techniques with small gold particles [1, 2, 3, 4], fluorescent lipoproteins LDL [5], a photobleaching technique [6], or a photothermal self-diffracting technique [7].

The formal representation of the diffusion equation on any two dimensional surface [8, 9], or on a distorted lattice [10, 11] is known. The local frame of the diffusion equation on a Riemannian manifold has the Euclidean structure, where the diffusion can be described by an ordinary Langevin equation with a white noise, or by a random walk. The diffusion equation for a minimal periodic surface of a cubic symmetry has been also described using the finite element method [12] in terms of the stationary limit of the effective diffusion coefficient. Different mathematical methods to study isotropic transport processes on the Riemannian manifolds have been developed also in mathematics [13, 14, 15], but so far these methods have not been used in physics because of their mathematical complexity.

We have recently proposed a simple algorithm [16] for describing the diffusion on a curved surface. Here we apply it to study the process of approaching the equilibrium state by diffusing particles on a finite piece of surfaces, with reflecting boundary conditions on the edges. We can describe a surface not only by its topological or geometrical characteristics, but also by the first nonzero eigenvalue of the diffusion operator on it. This eigenvalue gives the inverse of the mean time needed to reach equilibrium state.

The physical realization of such a problem is given by the diffusion of heat [7, 17, 18]. The mean time needed to heat uniformly a finite two dimensional system using point heater attached to the fixed place on a surface (for example laser beam heating like in [7]), is given by the inverse of the first nonzero eigenvalue of the diffusion operator on it. This decay rate is also the mean time needed to reach stationary state of uniform coverage of a surface by a finite, large set of particles started form the fixed point of space.

In this paper we study the diffusion on surfaces given by the general equation:

$$\phi(\mathbf{r}) = 0. \tag{1}$$

This equation is valid on the domain $\mathbf{D} \subset \mathbf{R}^3$ defined as a cubic cell:

$$-\frac{L}{2} \leq \{x, y, z\} \leq \frac{L}{2}. \tag{2}$$

To describe a dynamical process on this surface we set reflecting boundary conditions on the border of the domain $\mathbf{D}$ (vanishing currents for $x = \pm\frac{L}{2}$, or $y = \pm\frac{L}{2}$, or $z = \pm\frac{L}{2}$).

The paper is organized as follows. In Sec. 2 we present the simulation algorithm. Sec. 3 contains the analytical results for the diffusion equation on a sphere and a finite cylinder. In Sec. 4 we propose the formula for describing the diffusion coefficient for any closed, curved surface for long times. We apply the formula for a variety of closed, nodal surfaces (P,D,G etc.). A summary is contained in Sec. 5.

2 The Algorithm for Diffusion on a Curved Surface

The Brownian motion on a flat surface is the Wiener-Lévy process [19], which is described by the density distributions:

$$P_1(\mathbf{y}_2, t_2) = \int P_{1|1}(\mathbf{y}_2 - \mathbf{y}_1 | t_2 - t_1) P_1(\mathbf{y}_1, t_1) d\mathbf{y}_1,$$

$$P_{1|1}(\mathbf{y}_2 - \mathbf{y}_1 | t_2 - t_1) = \frac{1}{\sqrt{2\pi(t_2 - t_1)}} \exp(-\frac{(\mathbf{y}_2 - \mathbf{y}_1)^2}{2(t_2 - t_1)}), \tag{3}$$

with the initial condition $P_1(\mathbf{y}_1, 0) = \delta(\mathbf{y}_1)$. Here $\mathbf{y}_i$ denotes the position and t denotes time. This process, as a Markov one, satisfies the Chapman-Kolmogorov equation:

$$P_{1|1}(\mathbf{y}_3 - \mathbf{y}_1 | t_3 - t_1) = \int P_{1|1}(\mathbf{y}_3 - \mathbf{y}_2 | t_3 - t_2) P_{1|1}(\mathbf{y}_2 - \mathbf{y}_1 | t_2 - t_1) d\mathbf{y}_2. \tag{4}$$

The diffusion process can be discretized in time by introducing the Gaussian operator T_{τ_0} of the elementary, discrete step of duration τ_0. The elementary jump operator has the Gaussian form:

$$T_{\tau_0}(\mathbf{y}_2 - \mathbf{y}_1) = P_{1|1}(\mathbf{y}_2 - \mathbf{y}_1 | t_2 - t_1), \tag{5}$$

for $\tau_0 = t_2 - t_1$, and adding rule:

$$T^2_{\tau_0}(\mathbf{y}_3 - \mathbf{y}_1) = \int T_{\tau_0}(\mathbf{y}_3 - \mathbf{y}_2) T_{\tau_0}(\mathbf{y}_2 - \mathbf{y}_1) d\mathbf{y}_2,$$

or using the operator notation:

$$T^2_{\tau_0} = T_{\tau_0} T_{\tau_0} = T_{\tau_0 + \tau_0}.$$

After time t the operator takes the form:

$$T_{t=n\tau_0} = \underbrace{T_{\tau_0} T_{\tau_0} \ldots .}_{\text{n times}} = T_{\tau_0}^n. \tag{6}$$

The equation of the evolution can be rewritten as follows:

$$P(\mathbf{y}, t = n\tau_0) = \underbrace{T_{\tau_0} T_{\tau_0} \ldots .}_{\text{n times}} P(\mathbf{y}, 0) = T_{\tau_0}^n P(\mathbf{y}, 0), \tag{7}$$

where $P(\mathbf{y}, 0) = \delta(\mathbf{y})$ is the initial probability distribution. $P(\mathbf{y}, t)$ is the final distribution, which gives the probability of finding a particle after time t at point $\mathbf{y}$ on a flat surface.

This general method can be also used for a curved surface in the following way. At each point of the surface described by the Eq. 1 we define the plane tangent to the surface. At point $\mathbf{r}_0 = (x_0, y_0, z_0)$ the plane is described by the following equation:

$$\mathbf{n}(\mathbf{r}_0)(\mathbf{r} - \mathbf{r}_0) = 0, \tag{8}$$

where $\mathbf{n}(\mathbf{r}_0)$ is the vector normal to the surface. For a convenience we introduce on the tangent plane local polar coordinates given by the pair of variables: $\{J, \phi\}$. Then the Gaussian operator T_{τ_0} can be divided onto two operators: the angle rotation operator $T_{\tau_0}^a$ (to choose the direction of the movement - in the ϕ coordinate), and radial jump described by the $T_{\tau_0}^J$, with a proper distribution of jump length J.

Our numerical method [16] consists of a sequence of elementary steps. A particle at point $\mathbf{r}_0$ jumps along randomly chosen direction ($T_{\tau_0}^a(\phi) = \frac{1}{2\pi}$) in the plane tangent to the surface. The length of the jump J is drawn from the distribution (see also Eqs. 3, 5):

$$T^J_{\tau_0}(J) = \frac{J}{2D_0\tau_0}\exp\left(-\frac{J^2}{4D_0\tau_0}\right), \tag{9}$$

where D_0 is the planar diffusion coefficient, τ_0 is the duration of the elementary time step. After the jump the particle is at point $\mathbf{r}_1$ on the tangent plane ($\mathbf{n}(\mathbf{r}_0)(\mathbf{r}_1 - \mathbf{r}_0) = 0$). Then we project the point $\mathbf{r}_1$ back to the curved surface along the direction given by $\nabla\phi(\mathbf{r}_1)$ (approximately normal to the surface). The final location is given by the formula:

$$\mathbf{r}_2 = \mathbf{r}_1 - \frac{\phi(\mathbf{r}_1)\nabla\phi(\mathbf{r}_1)}{|\nabla\phi(\mathbf{r}_1)|^2}. \tag{10}$$

The elementary step is next repeated from the point $\mathbf{r}_2$.

In the case of a finite piece of a surface we introduce reflecting boundary conditions to ensure zero current flow of particles through borders of the domain. The boundary condition is given by the equation:

$$\partial_n P(\mathbf{r}, t) = 0,$$

where ∂_n is the derivative normal to the boundary for $\mathbf{r}$ on the border of the domain $\mathbf{D}$.

The algorithm satisfies the detailed balance condition and is stable [16], providing that steps are much smaller than the typical radius of curvature R_1 averaged over whole surface. One cannot see any roughness of the surface less then a typical jump length.

We determine in numerical simulations the effective diffusion coefficient $D_{eff}(t)$ defined here by the formula:

$$D_{eff}(t) = \frac{< \Delta\mathbf{R}(t)^2 >}{4t}, \tag{11}$$

where t is the time of measurement, and

$$\Delta \mathbf{R}^2(t) = |\mathbf{r}(t) - \mathbf{r}(0)|^2, \tag{12}$$

is the mean square displacement of the probe particle during time t. The average, $< ... >$ is taken with the probability distribution $P(\mathbf{r}, t)$ of finding a particle at point $\mathbf{r}(t)$ at time t, and over all initial positions $\mathbf{r_0} : P(\mathbf{r}, t = 0) = \delta(\mathbf{r} - \mathbf{r_0})$.

We use one random walker, and take averages over all its trajectories on the surface. Because of the ergodicity and Markov nature of the random walk [16] the precise location of the starting point is unimportant. A typical simulation consists of $K = 3 * 10^3$ trajectories (we take last point of each trajectory as a starting point $\mathbf{r}_0$ for the next one), each of $M = 10^6$ steps. A typical size of the elementary jump is equal to $J_0 = \sqrt{4D_0\tau_0}$ in comparison to a typical linear size of the cubic cell $L \approx 100J_0$. The averaged value of $D_{eff}(t)$ at time $t = N\tau_0$ is taken over $m = (M - N) * K$ steps, and given by the formula:

$$D_{eff}(t)/D_0 = \sum_{j=1}^{K} \sum_{i=1}^{M-N} \frac{|\mathbf{r}_j(t_i + t) - \mathbf{r}_j(t_i)|^2}{K * (M - N)} \frac{1}{4t}. \tag{13}$$

Single trajectory is described by j index, and each step of this trajectory by the i index.

In the short time limit the effective diffusion coefficient $D_{eff}(t)$ is equal to the planar diffusion constant D_0. In the long time limit $D_{eff}(t)$ for all closed surfaces goes to zero due to the fact that the region available for the diffusing particle is bounded. The effective diffusion coefficient aproaches zero after time roughly proportional to the surface area of a closed surface. So in order to compare the time evolution of $D_{eff}(t)$ for various surfaces, one needs to rescale the size L of the domain $\mathbf{D}$, to have the same surface area for all surfaces.

3 The effective diffusion coefficient for sphere and cylinder

We solve the diffusion equation on a surface in three dimensional cartesian space $\mathbf{R^3}$:

$$\frac{\partial P(x,y,z;t)}{\partial t} = D_0 \Delta P(x,y,z;t), \tag{14}$$

where (x,y,z) denotes a point at the surface. For the sphere of radius R this equation takes the form:

$$\frac{\partial P(\phi,\theta;t)}{\partial t} = D_0 \left(\frac{1}{R^2 \tan\theta} \partial_\theta + \frac{1}{R^2} \partial_\theta^2 + \frac{1}{R^2 \sin^2\theta} \partial_\phi^2 \right) P(\phi,\theta;t), \tag{15}$$

where θ and ϕ are spherical angles, which define the position of a probe particle on a sphere. The solution of this equation can be written [16] as an infinite sum of exponential terms, describing different modes coupled to different eigenvalues indexed by n :

$$P(\beta;t) = \frac{1}{2} \sum_{l=0}^{\infty} (2l+1) P_l(\cos\beta) \exp\left(-t \frac{l(l+1)D_0}{R^2} \right) \sin\beta, \tag{16}$$

where P_l are the Legendre polynomials, and β is the angle between start and the final position on the sphere, given by the Pitagoras lemma:

$$\beta = 2 \arcsin \left(\sqrt{\sin^2\left(\frac{\theta-\theta_0}{2}\right) + \sin^2\left(\frac{\phi-\phi_0}{2}\right)} \right).$$

The effective diffusion coefficient is given by only one exponential term coupled to the first eigenvalue of the full solution:

$$D_{eff}(t) = \frac{R_\infty^2}{4t}\left(1 - \exp\left(-\frac{\alpha t}{R_\infty^2}\right)\right), \tag{17}$$

where

$$R_\infty^2 = \lim_{t\to\infty} < \Delta\mathbf{R}(t)^2 >= 2R^2,$$

and

$$\alpha = 4D_0.$$

As the second step of our simple analysis we solve the diffusion equation for a finite cylinder of radius R and length L. First we introduce new coordinates: $\{\phi, z\}$, which describe the position of the random walker on the cylinder. The diffusion equation than takes the form:

$$\frac{\partial P(\phi, z; t)}{\partial t} = D_0\left(\frac{1}{R^2}\partial_\phi^2 + \partial_z^2\right) P(\phi, z; t). \tag{18}$$

The solution is given by infinite series of exponential modes indexed by two integer numbers $\{n, m\}$:

$$\begin{gathered}P(\phi, z; t) = \tfrac{1}{2\pi L}\{1+\\ 2\textstyle\sum_{n=1}^{\infty} \cos\left(\frac{n\pi z_0}{L}\right)\cos\left(\frac{n\pi z}{L}\right)\exp\left(-D_0\frac{\pi^2 n^2}{L^2}t\right) +\\ 2\textstyle\sum_{m=1}^{\infty} \cos(m\phi)\cos(m\phi_0)\exp\left(-D_0\frac{m^2}{R^2}t\right) +\\ 4\textstyle\sum_{m=1,n=1}^{\infty} \cos\left(\frac{n\pi z_0}{L}\right)\cos\left(\frac{n\pi z}{L}\right)\cos(m\phi_0)\cos(m\phi)\exp\left(-D_0\left(\frac{\pi^2 n^2}{L^2} + \frac{m^2}{R^2}\right)t\right)\}.\end{gathered}$$

The effective diffusion coefficient can be calculated using the equation:

$$D_{eff}(t) = \frac{< \Delta\mathbf{R}(t)^2 >}{4t} = \frac{1}{4t} < (z - z_0)^2 + 4R^2\sin^2\left(\frac{\phi - \phi_0}{2}\right) >, \tag{19}$$

where the average is taken over all possible trajectories (each trajectory starts at the point marked by the pair of variables: $\{\phi_0, z_0\}$). The result of this averaging is given by the formula:

$$
\begin{aligned}
D_{eff}(t) = \left(\tfrac{R^2}{2t} + \tfrac{L^2}{12t}\right) - \tfrac{R^2}{2t} \exp\left(-\tfrac{D_0 t}{R^2}\right) - \\
\tfrac{4L^2}{\pi^4 t} \textstyle\sum_{k=0}^{\infty} \exp\left(-D_0 \tfrac{\pi^2 (2k+1)^2}{L^2} t\right),
\end{aligned}
\tag{20}
$$

with $n = 2k + 1$. In the long time limit only terms with lowest exponents are important:

$$
\begin{aligned}
D_{eff}(t \to \infty) = \left(\tfrac{R^2}{2t} + \tfrac{L^2}{12t}\right) - \tfrac{R^2}{2t} \exp\left(-\tfrac{D_0 t}{R^2}\right) - \\
\tfrac{4L^2}{\pi^4 t} \exp\left(-D_0 \tfrac{\pi^2}{L^2} t\right).
\end{aligned}
\tag{21}
$$

In order to describe the long time limit of evolution of $D_{eff}(t)$ we analyze the function of time $F(t)$ given by the equation:

$$
F(t) = \ln\left[R_\infty^2 - < \Delta\mathbf{R}^2(t) >\right].
\tag{22}
$$

Encouraged by the analytic result for a sphere we state the following hypothesis:

For all nodal surfaces for long times function the mean square displacement of the probe particle is a single exponential function of time with the form:

$$
< \Delta\mathbf{R}^2(t) > = R_\infty^2 \left[1 - \exp\left(-\frac{\alpha}{R_\infty^2} t\right)\right]
\tag{23}
$$

Then we have a linear behaviour of the function $F(t)$:

$$
F(t) = -\frac{\alpha}{R_\infty^2} t + \ln\left[R_\infty^2\right].
\tag{24}
$$

The time dependence of this function in the case of a sphere, for long times, is given by the exponential term of the $D_{eff}(t)$ function. For the sphere with radius R ($R^2_\infty = 2R^2$) the late decay coefficient is given by:

$$\alpha^{sphere} = 4D_0. \tag{25}$$

For the cylinder (see Eq. 21) generally one cannot use such a single parametrized function $F(t)$. For long times and radius R much larger than the length L of the cylinder (then $R^2_\infty = 2R^2$) the $F(t)$ function is linear also for this structure, and the coefficient is given by the formula:

$$\alpha = 2D_0. \tag{26}$$

4 Long time exponential decay of the effective diffusion coefficient for finite, nodal surfaces

In this section we present values of the first non-zero eigenvalue of the diffusion operator (the coefficient α rescaled by the R^2_∞ in the function $F(t)$) for a variety of nodal structures.

All structures presented here are defined in a cubic box (domain **D**) with edges of length L. For each surface we rescale the size L of the domain **D** to ensure the proper comparison of the time evolution of D_{eff} for various surfaces. We investigate a variety of closed, nodal surfaces, and a sphere of radius R.

We have applied the algorithm to study the diffusion on the P closed, nodal surface [25] given by Eq. 1 with

$$\phi(\mathbf{r}) = \cos X + \cos Y + \cos Z, \tag{27}$$

where $X = 2\pi x/L$, $Y = 2\pi y/L$, $Z = 2\pi z/L$, and the size of the cubic cell $L = 100\sqrt{4D_0\tau_0}$. On Fig 1 there is also D closed nodal surface [16, 25] given by the equation:

$$\cos X \cos Y \cos Z - \sin X \sin Y \sin Z = 0. \tag{28}$$

On Fig. 2 we present G closed nodal surface [16, 25]:

$$\sin X \cos Z + \sin Y \cos X + \sin Z \cos Y = 0, \tag{29}$$

and S nodal surface [23]:

$$\begin{gathered}\cos Y \cos 2Z \sin X + \cos 2X \cos Z \sin Y + \\ \cos X \cos 2Y \sin Z = 0.\end{gathered} \tag{30}$$

On Fig. 3 we see the I-WP nodal surface given by Eq. 1 with [23]:

$$\begin{gathered}\phi(\mathbf{r}) = 2(\cos X \cos Y + \cos Y \cos Z + \cos Z \cos X) \\ -(\cos 2X + \cos 2Y + \cos 2Z),\end{gathered} \tag{31}$$

and S1 nodal surface by [24]:

$$\begin{gathered}\phi(\mathbf{r}) = \cos X \sin Y \sin 2Z + \cos Y \sin Z \sin 2X + \\ \cos Z \sin X \sin 2Y + 2A_2(cos2X \cos 2Y + \\ \cos 2Y \cos 2Z + \cos 2Z \cos 2X),\end{gathered} \tag{32}$$

where $X = 2\pi x/L$, $Y = 2\pi y/L$, $Z = 2\pi z/L$, and A_2 is a parameter ($A_2 = 0.1$).

In order to compare results for these surfaces we take different sizes of cubic cells for different surfaces in the units of the jump length $J_0 = \sqrt{4D_0\tau_0}$: $d_P = 100J_0$, $d_G = 87J_0$, $d_D = 78J_0$, $d_S = 65J_0$, $d_{I-WP} = 80J_0$, and $d_{S1,A_2=0.1} = 60J_0$, so that their surface areas of cells are the same.

Values of α are given in Table **I** for closed, nodal surfaces and a sphere. The typical behaviour of $F(t)$ is presented on Fig. 4 for P closed, nodal surface. The estimated error of simulations for α coefficient is equal to 1%.

5 Summary

We presented here a formal, mathematical description (in terms of stochastic processes) of a simple numerical method for studying the long time behaviour of the mean square displacement of the probe particle, averaged over the whole surface. The time evolution of the effective diffusion coefficient $D_{eff}(t)$ for nodal surfaces for long times is described by a single exponential term. Exponent value for the surface is its fingerprint and depends on its geometry, shape and topology. It is given by the α coefficient presented in Table **I** rescaled by the R^2_∞ (see Eq. 23). The first nonzero eigenvalue of the diffusion operator on such a surface gives the inverse of the mean time needed to reach equilibrium.

The diffusion process is a fundamental phenomenon, so our method should find applications in various fields of physics (modeling of the diffusion in two dimensional systems, heat conductance problems), mathematics (theory of diffusion operator on Riemannian manifolds), chemistry (theory of chemical reactions - an enhancement of the reaction constant by lowering dimensionality of the system) and biophysics (gene expression, or biochemical reactions on membranes in organic cells).

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7 Tables

structure	α	R_∞
S	1.54	45.8
P	1.5	68.5
I-WP	1.46	56.1
G	1.42	61.4
S1($A_2 = 0.1$)	1.36	45.9
D	1.14	52.5

Table **I**.

The α coefficient and the value of R_∞ (in units of the average jump length J_0) in the function $F(t)$. The inverse of this coefficient is the late stage decay time of $< R^2(t) >$, and the first non-zero eigenvalue of the diffusion operator for a given surface rescaled by R_∞^2 (see Eq. 22, 23, 24). We investigate following surfaces: a sphere (for a sphere with radius R we have $\alpha^{sphere} = 2$, and $R_\infty = \sqrt{2}R = 60.8J_0$), closed, nodal surfaces: P, D (see Fig. 1 and Eq. 27, 28), G, S (see Fig. 2 and Eq. 29, 30), and I-WP, S1 (see Fig. 3 and Eq. 31, 32). The average error for the α coefficient and R_∞ is equal to 1%.

8 Figure Captions

- Fig. 1 P (top), D (bottom picture) nodal surfaces given by equations 27, 28 in a unit cell (domain **D**).

- Fig. 2 G (top, Eq. 29), S (bottom picture, Eq. 30) nodal surfaces in a unit cell (domain **D**).

- Fig. 3 I-WP (top), S1 (for $A_2 = 0.2$ - bottom picture) surfaces described by equations 31, 32 in a unit cell (domain **D**).

- Fig. 4 The $F(t)$ function for P closed, nodal surface (see Eq. 27). For this surface $\alpha = 1.5$. Squares are results of simulations, and the dashed line is the linear fit to the $F(t)$ function. One can see the linear behaviour in agreement with our hypothesis of the decay described by only one exponential term. The exponent gives the first nonzero eigenvalue of the diffusion operator on the P nodal surface.

9 Figures

Figure 1:

Figure 2:

Figure 3:

Figure 4: