diffusion-fundamentals.org

The Open-Access Journal for the Basic Principles of Diffusion Theory, Experiment and Application

Projection of two-dimensional diffusion in a curved midline and narrow varying width channel embedded on a curved surface

Guillermo Chacón-Acosta[1⋆], Inti Pineda[2*], Leonardo Dagdug[2†]

[1]Department of Applied Mathematics and Systems, Universidad Autónoma Metropolitana-Cuajimalpa, Artificios 40, México D. F. 01120, México
[2]Physics Department, Universidad Autónoma Metropolitana-Iztapalapa, San Rafael Atlixco 186, México D. F. 09340, México
⋆gchacon@correo.cua.uam.mx, *inti@xanum.uam.mx, †dll@xanum.uam.mx

Abstract

This study focuses on the derivation of a general effective diffusion coefficient to describe the two-dimensional (2D) diffusion in a narrow and smoothly asymmetric channel of varying width that lies on a curved surface, in the simple diffusional motion of noninteracting point-like particles under no external field. To this end we extend the generalization of the Kalinay-Percus' projection method [J. Chem. Phys. 122, 204701 (2005); Phys. Rev. E 74, 041203 (2006)] for the asymmetric channels introduced in [J. Chem. Phys. 137, 024107 (2012)], to project the anisotropic 2D diffusion equation on a smooth curved manifold into an effective one-dimensional generalized Fick-Jacobs equation which is modified due to the curvature of the surface. The lowest order in the perturbation parameter, corresponding to the Fick-Jacobs equation, contains an extra term that accounts for the curvature of the surface. We found explicitly the first order correction for the invariant effective concentration, which is defined as the correct marginal concentration in one variable, and we obtain the first approximation to the effective diffusion coefficient analogous to Bradley's coefficient [Phys. Rev. E 80, 061142 (2009)] as a function of metric elements of the surface. Straightforwardly we study the perturbation series up to the n-th order, and we derive the full effective diffusion coefficient for 2D diffusion in a narrow asymmetric channel, which have modifications due to the curved metric. Finally, as an example we show how to use our formula to calculate the effective diffusion coefficient considering the case of an asymmetric conical channel embedded on a torus.

1. Introduction

Diffusion theory played a major role in science and technology. Effective control of transport in several micro- and nanostructures requires a deep understanding of the diffusive mechanisms. The diffusion of particles or, more generally, small objects, spatially confined within quasi-one-dimensional systems (pores, channels, fibers or carbon nanotubes) has gained increasing attention over the last decade because such systems are ubiquitous in both nature and technology [1]. However, the analysis of transport through them traditionally has been addressed using usual diffusion equation on flat spaces.

It is well known that in biophysics and nanotechnology the diffusion can occur on a given two-dimensional (2D) curved surfaces, for instance, the lateral motion of integral proteins or receptors embedded on lipid bilayers. The biological membranes are primarily composed of hundreds of different lipids and huge diversity of proteins spatially and temporally organized as a requirement for its biological function [2, 3]. Lateral mobility of components of cell membranes can be hindered by the presence of impermeable heterogeneities, lipid microdomains, patches and rafts, with the effective diffusion constant thus being reduced [4]. Furthermore, the apparent anisotropy in the effective diffusion on cell membranes is due to real biological membranes containing tubular networks, holes, and large curvature variations. The boom of new techniques, such as the single-particle tracking, fluorescence recovery after photobleaching and fluorescence correlation spectroscopy has allowed for unprecedented ways to study the motion of proteins, molecular receptors, and lipids on the cell surfaces; and has greatly furthered our understanding of its crucial role in the cellular functioning [5, 6, 7].

In material science, surface processes are of high importance. The applications range from self-organized growth of nanostructures over crystal growth, shape transitions in alloys, and adsorption and desorption phenomena to surface reactions among others [8, 9, 10]. Analysis of the experimental data have reveled that in some cases, diffusion on curved surfaces proceeds at a different rate from the conventional diffusion on the plane.

Aside from the development of experimental procedures, the problem of particle transport through confined structures on flat spaces has led to recent theoretical efforts to study diffusion dynamics appearing in those geometries [11]. Earlier studies by Jacobs and Zwanzig triggered renewed research on this subject [12, 13]. The so-called Fick-Jacobs (FJ) approach consists in eliminating transverse stochastic degrees of freedom by assuming fast equilibration in such directions.

For wide quasi-one-dimensional structures, one can map particle motion onto an effective one-dimensional description in terms of diffusion along the midline of the channel $y_0(x)$, in the presence of entropy potential $U_{ent}(x)$ given by $\beta U_{ent}(x) = \ln(1/w(x))$, where x is the particle coordinate measured along the x axis, $w(x)$ is the channel width as a function of x, $\beta = 1/(k_B T)$, k_B is the Boltzmann constant, and T is the absolute temperature.

Using the entropy potential, one can write the one-dimensional Smoluchowski equation for the probability density $p(x,t)$ in the channel, namely,

$$\frac{\partial}{\partial t}p(x,t) = \frac{\partial}{\partial x}\left[D(x)e^{-\beta U(x)}\frac{\partial}{\partial x}e^{\beta U(x)}p(x,t)\right], \tag{1}$$

where $D(x)$ is a position-dependent diffusion coefficient. Equation (1) is equivalent to the generalized FJ equation,

$$\frac{\partial}{\partial t}p(x,t) = \frac{\partial}{\partial x}\left[D(x)w(x)\frac{\partial}{\partial x}\frac{p(x,t)}{w(x)}\right]. \tag{2}$$

This equation with a position-independent diffusion coefficient, $D(x) = D_0$, is known as the FJ equation [13].

Recently, using the projection method proposed by Kalinay and Percus (KP)[14], a more general effective diffusion coefficient in two dimensions was obtained [15] (DP),

$$D(x) = \frac{D_0}{w'(x)}\left\{\arctan\left[y_0'(x) + \frac{w'(x)}{2}\right] - \arctan\left[y_0'(x) - \frac{w'(x)}{2}\right]\right\}. \tag{3}$$

Equation (3) generalizes all known effective diffusion coefficients theoretically derived so far for 2D narrow channels, and was validated recently by brownian dynamicas simulations [16, 17, 18]. Setting $y_0'(x) = 0$ in Eq. (3), KP's results for symmetric channels is recovered,

$$D(x) \approx D_{\mathrm{KP}}(x) = \frac{\arctan[\frac{1}{2}w'(x)]}{\frac{1}{2}w'(x)}D_0. \tag{4}$$

If $w'(x) = 0$, the case of a serpentine channel previously studied by Yariv and co-workers [19] is obtained,

$$D(x) \approx D_{\mathrm{YBK}}(x) = \frac{D_0}{1 + y_0'(x)^2}. \tag{5}$$

Furthermore, when Taylor expansion of Eq. (3) is kept up to the first order in $w'(x)$ and $y_0'(x)$, the diffusion coefficient proposed by Bradley [20] (Br) is recovered,

$$D(x) \approx D_{\mathrm{Br}}(x) = D_0\left(1 - y_0'(x)^2 - \frac{1}{12}w'(x)^2\right), \tag{6}$$

which is essentially the same as obtained two years later by Berezhkovskii and Szabo [21],

$$D(x) \approx D_{\mathrm{BS}}(x) = \frac{D_0}{1 + y_0'(x)^2 + \frac{1}{12}w'(x)^2}. \tag{7}$$

In this work we briefly summarize the results on the projection of two-dimensional diffusion to a one-dimensional effective dimension on a curved surface through the KP's method, that was first presented in [22].

2. Effective one dimensional diffusion on a curved surface

In order to study the diffusion in narrow channels, Kalinay and Percus [14] proposed a projection procedure that allows us to obtain corrections in terms of an expansion parameter namely, $\lambda = D_1/D_2$, being $D_1 \neq D_2$, the constant diffusion coefficient in the transversal and longitudinal directions. In order to proceed with the method, we first have to consider anisotropic diffusion, physically this means that the transverse contribution is transient and can be separated from the longitudinal one, projecting by integration.

First of all we need to write the diffusion equation for the anisotropic case on a two-dimensional Riemannian surface. This was achieved by using the corresponding operators for gradient and the divergence on the surface [23, 24]

$$\frac{\partial \tilde{C}}{\partial t} = \frac{1}{\sqrt{g}} \frac{\partial}{\partial x^\alpha} \left(\sqrt{g} D^\alpha_\beta g^{\beta\gamma} \frac{\partial \tilde{C}}{\partial x^\gamma} \right), \tag{8}$$

where x^α are the local coordinates on the surface, and g is the determinant of the metric $g_{\alpha\beta}$. Notice that in eq. (8) we are considering $\tilde{C} = \sqrt{g} C$ instead of C for the concentration, this is to guarantee the right scalar structure of the equation [23, 24]. The anisotropic diffusion tensor appearing in (8) is defined with respect to the local coordinates on the surface as,

$$[D^\alpha_\beta] = \begin{pmatrix} D_1 & 0 \\ 0 & D_2 \end{pmatrix}. \tag{9}$$

Let us notice that in principle it does not depend on the metric components because we are considering the case when diffusion is anisotropic over the orthogonal directions of the local coordinates on the surface. For isotropic diffusion $D^\alpha_\beta = D_0\, \delta^\alpha_\beta$, and Eq. (9) reduces to an equation with the Laplace-Beltrami operator [25, 26, 27]. In the literature there are several studies that show similar results obtained by this parameterization [28, 29, 30, 31].

Let us now consider the case of a metric that describes symmetrical surfaces with local coordinates (χ, η), which is diagonal, and whose components only dependent on one of the local coordinates. In this case its metric can be written as,

$$[g_{\alpha\beta}] = \begin{pmatrix} g_1(\chi) & 0 \\ 0 & g_2(\chi) \end{pmatrix}, \quad \left[g^{\alpha\beta}\right] = \begin{pmatrix} g_1^{-1}(\chi) & 0 \\ 0 & g_2^{-1}(\chi) \end{pmatrix}. \tag{10}$$

Although it looks like a very restrictive condition, there are many surfaces of interest that fulfill this requirement [32, 33]. In this way the diffusion equation only contains square roots of the metric components and is reduced to the following,

$$\frac{\partial \tilde{C}(\chi,\eta,t)}{\partial t} = \frac{D_\chi}{\sqrt{g_1 g_2}} \frac{\partial}{\partial \chi} \left[\sqrt{\frac{g_2}{g_1}} \frac{\partial}{\partial \chi} \tilde{C}(\chi,\eta,t) \right] + \frac{D_\eta}{\sqrt{g_1 g_2}} \sqrt{\frac{g_1}{g_2}} \frac{\partial^2}{\partial \eta^2} \tilde{C}(\chi,\eta,t). \tag{11}$$

The next step in the procedure is to integrate (11) on the transient variable η, to this end we first have to define the one-dimensional marginal concentration, namely,

$$c(\chi,t) \equiv \int_{f_1(\chi)}^{f_2(\chi)} \tilde{C}(\chi,\eta,t) d\eta, \tag{12}$$

where the geometry of the channel is given by the boundaries defined by the functions $f_1(\chi)$ and $f_2(\chi)$.

Equation (11) can be integrated by means of the fundamental theorem of Calculus applying the Leibniz rule [15]. Then imposing boundary conditions that produce a flow parallel to the channel walls, and keeping leading orders in parameter $\lambda = D_\chi/D_\eta$, a Fick-Jacobs type equation on a curved symmetric surface is obtained [22],

$$\frac{\partial c(\chi,t)}{\partial t} = \frac{D_\chi}{\sqrt{g_1 g_2}} \frac{\partial}{\partial \chi} \left(\sqrt{\frac{g_2}{g_1}} w(\chi) \frac{\partial}{\partial \chi} \frac{c(\chi,t)}{w(\chi)} \right). \tag{13}$$

From this last equation it is possible to apply the Kalinay and Percus' perturbation method [14] by expanding the concentration in the parameter λ and then by an iterative process, allowing us to obtain systematically higher order terms in λ. These corrections are contained in a generalization of the diffusion coefficient which can now be considered as dependent on the longitudinal coordinate χ. After this cumbersome procedure, it is possible to recognize the series that comes form the recurrence scheme, reaching the generalization of the expression found by Dagdug and Pineda [15] for two dimensional asymmetric channels, now for a curved surface,

$$D(\chi) = \frac{D_0}{w'(\chi)}\sqrt{\frac{g_1}{g_2}}\left\{ \arctan\left[\sqrt{\frac{g_2}{g_1}}\left(y_0'(\chi) + \frac{w'(\chi)}{2}\right)\right] - \arctan\left[\sqrt{\frac{g_2}{g_1}}\left(y_0'(\chi) - \frac{w'(\chi)}{2}\right)\right]\right\}. \tag{14}$$

Up to the date, this is the most general effective diffusion coefficient for 2D narrow channels that was obtained by the KP procedure.

3. Diffusion on narrow channels on the surface of a torus

In this section we study the effective diffusion of a narrow channel embedded on a torus. These kind of surfaces have an important role in physics as well as in some biophysical systems [34]. A scheme of a channel embedded on this surface is show in figure 1. A torus is a surface of revolution generated by rotating a circle in three-dimensional space about an axis which is outside the circle. The torus can be parameterized by two angles θ and φ, both varying form 0 to 2π; and two constant radii $b < a$, corresponding to the inner and outer circles respectively. With this parametrization the line element is given by,

$$ds^2 = (a + b\cos\theta)^2 \, d\varphi^2 + b^2 d\theta^2. \tag{15}$$

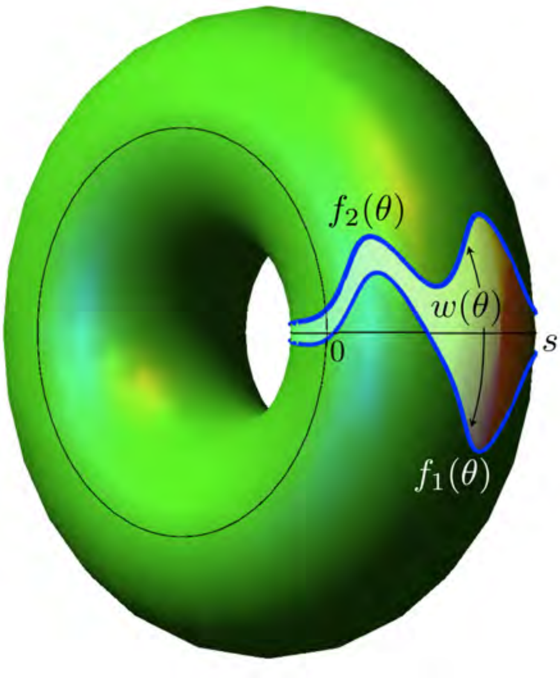

Figure 1: A schematic representation of an asymmetric channel on the surface of the torus is presented. The boundary functions $f_1(\theta)$ and $f_2(\theta)$, as well as the width function $w(\theta)$ are shown.

Torus' curvature depend on the corresponding angle θ and is given by $K = \cos\theta/[b\,(a + b\cos\theta)]$ [32, 33]. From (15) we notice that just one of the metric coefficients is function of local coordinates [22]. The longitudinal variable is identified with $b\theta$. The metric coefficients can be written as follows,

$$g_1(\theta) = b^2, \qquad g_2(\theta) = (a + b\cos\theta)^2 \,. \tag{16}$$

This last two relations allow us to write out the following ratio,

$$\sqrt{\frac{g_1(\theta)}{g_2(\theta)}} = \frac{1}{\frac{a}{b} + \cos\theta}, \tag{17}$$

On plugging the above expression into (14) we obtain that the effective diffusion coefficient dependent on the longitudinal local coordinate is given by,

$$\begin{aligned} D_{torus}(\theta) = \quad & \frac{D_0}{w'(\theta)} \frac{1}{\frac{a}{b} + \cos\theta} \left\{ \arctan\left[\left(\frac{a}{b} + \cos\theta\right) \left(y_0'(\theta) + \frac{w'(\theta)}{2} \right) \right] \right. \\ & \left. - \arctan\left[\left(\frac{a}{b} + \cos\theta\right) \left(y_0'(\theta) - \frac{w'(\theta)}{2} \right) \right] \right\}. \end{aligned} \tag{18}$$

As an illustrative example of how particles diffuse in this surface, let us study a simple asymmetrical conical channel made up by two straight lines $\varphi_2 = m_2\theta - \varphi_0$ and $\varphi_1 = m_1\theta + \varphi_0$. For this channel the effective diffusion coefficient is as follows,

$$D_{torus}(\theta) = \frac{D_0}{m_1 - m_2} \frac{1}{\frac{a}{b} + \cos\theta} \left\{ \arctan\left[m_1 \left(\frac{a}{b} + \cos\theta\right) \right] - \arctan\left[m_2 \left(\frac{a}{b} + \cos\theta\right) \right] \right\}. \tag{19}$$

In figure 2, we show a diagram of this channel and the effective diffusion coefficient is plotted. We keep constant the upper slope m_2, while varying the lower m_1. As expected [22], the diffusion coefficient increases with increasing radius.

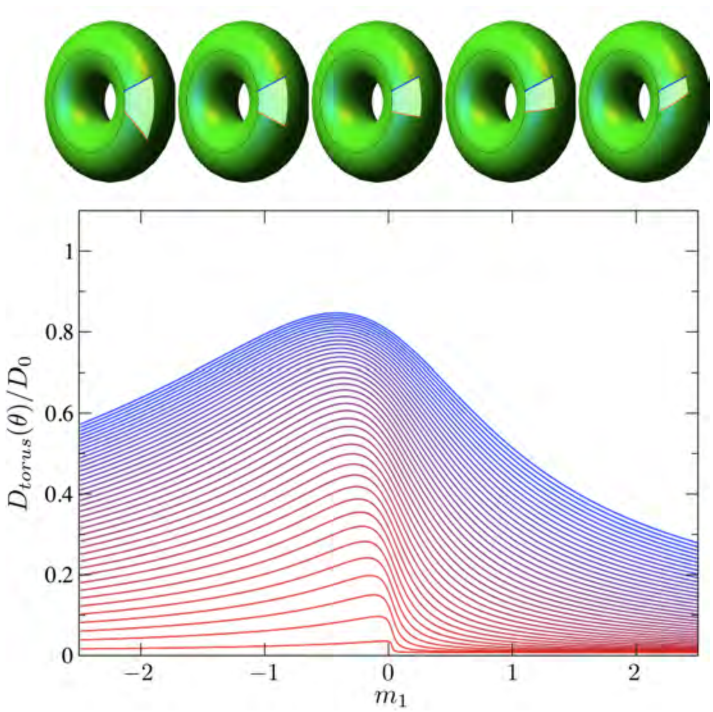

Figure 2: An asymmetric channel formed by straight walls. The slope of the upper boundary is fixed at $m_2 = 1$ and the lower one varies from $-2 \leq m_1 \leq 2$. The effective diffusion coefficient is plotted at $\theta \approx 0$ We keep the large radius fixed at $a = 1$, while the small radius takes values $0.01 \leqslant b \leqslant 1$. The increase in the small radius is shown with the change from red to blue hues.

Is worth noticing that the factor (17) can be rewritten in terms of the Gaussian curvature instead of the angle. Although this gives no gain to simplify calculations, it helps us to see how the effective diffusion coefficient is modified in terms of the curvature,

$$\sqrt{\frac{g_1(\theta)}{g_2(\theta)}} = \frac{b}{a}\left(1 - b^2 K\right), \tag{20}$$

Explicitly for (19) we have,

$$D_{torus}(K) = \frac{D_0}{m_1 - m_2}\frac{b}{a}\left(1 - b^2K\right)\left\{\arctan\left[m_1\left(\frac{a}{b\left(1-b^2K\right)}\right)\right]\right.$$
$$\left. - \arctan\left[m_2\left(\frac{a}{b\left(1-b^2K\right)}\right)\right]\right\}. \tag{21}$$

Equation (21) gives us an insight of how the curvature of the manifold changes the dynamics of the system and in particular, how it influences the effective diffusion in the confined geometry. Indeed we can notice that, as the curvature varies $-1/[b(a-b)] \le K \le 1/[b(a+b)$, the effective diffusion coefficient changes.This dependence is shown on figure 3. From $\theta = 0$ (fig. 2) to $\theta = \pi/2$ (fig. 3(a)), the effective diffusion coefficient decreases. From $\theta = \pi/2$ to $\theta = \pi$ (fig. 3(b)) it grows and continues growing till $\theta = 3\pi/2$ (fig. 3(c)). From that point it begins to decrease again to $\theta = 2\pi$ (fig. 3(d)).

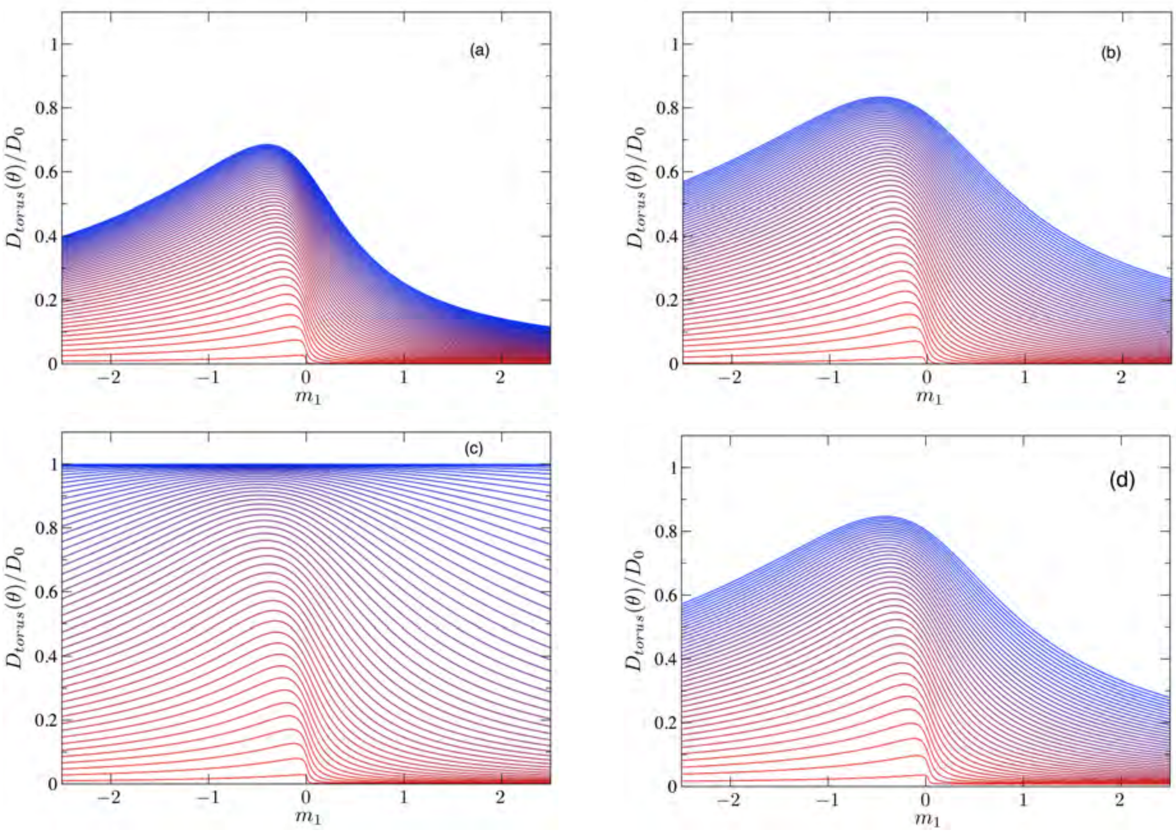

Figure 3: Effective diffusion coefficient for an asymmetric channel formed by straight walls with upper boundary fixed at $m_2 = 1$ and the lower one varying from $-2 \le m_1 \le 2$. The large radius was fixed at $a = 1$, and the small radius was chosen to vary $0.01 \leqslant b \leqslant 1$. The increase in b is shown with the change from red to blue. We plot the ratio $D_{torus}(\theta)/D_0$ at different angles (a) $\theta = \pi/2$, (b) $\theta = \pi$, (c) $\theta = 3\pi/2$ and $\theta = 2\pi$.

4. Summary and conclusions

In this work we present the corresponding Fick-Jacobs equation for a two-dimensional narrow asymmetric channel with varying width $w(\chi)$ and a non-straight midline $y_0(\chi)$, embedded on a symmetric curved surface. The effective diffusion coefficient that depends on the longitudinal local coordinate, is given by Eq. 14), and was obtained by using the Kalinay and Percus' projection method. For a flat surface, this expression reduces to DP's result for a general asymmetric channel given in Eq. (3), which also contains all previous known results.

In particular we present an asymmetric conical channel embedded on torus' surface. We recovered the results obtained by DP with additional dependence of the effective diffusion coefficient on both, the radius a and b, and the angle θ. For the asymmetric conical channel, the diffusion coefficient for a fixed slope, first decreases from 0 to $\pi/2$, then starts to increase up to the value $\theta = 3\pi/2$, where it begins to decrease again till 2π. Varying the small radius the effective diffusion coefficient decreases as b increases as was already shown in [22] for the sphere. We also notice that in eq. (21) the effective diffusion coefficient can be written in terms of the curvature of the surface.

5. Acknowledgments

This work was partially supported by PROMEP under Grant No. 47510283 (GCA), and by CONACyT under Grant No. 176452. We also thank the organizers of Diffusion Fundamentals V, for their hospitality and for the very nice conference that allowed an environment of scientific exchange.

References

[1] J. Kärger,D. M. Ruthven, and D. N. Theodorou, *Diffusion in Nanoporous Materials Vols. 1 & 2* (Wiley-VHC, Weinheim, 2012).

[2] B. Alberts, A. Johnson, J. Lewis, M. Raff, K. Roberts, and P. Walter, *Molecular Biology of the Cell* (Garland Science, New York, 2007).

[3] T. Fujiwara, K. Ritchie, H. Murakoshi, K. Jacobson, and A. Kusumi, J. Cell Biol. **157**, 1071 (2002); R. G. Parton and J. F. Hancock, TRENDS in Cell Biology **14**, 41 (2004); S. Semrau and T. Schmidt, Soft Matter **5**, 3174 (2009).

[4] M. J. Saxton, Biophys. J. **39**, 165 (1982); A. Kusumi, Y. Sako, and M. Yamamoto, Biophys. J. **65**, 2021 (1993); A. D. Douglass and R. D. Vale, Cell **121**, 937 (2005); C. A. Day and A. K. Kenworthy, Biochimica et Biophysica Acta **1788**, 245 (2009).

[5] J. Hwang, L. A. Gheber, L. Margolis, and M. Edidin, Biophys. J. **74**, 2184 (1998); J. Capoulade, M. Wachsmuth, L. Hufnagel, and M. Knop, Nat. Biotechnol. **29**, 835 (2011).

[6] J. Lippincott-Schwartz, E. Snapp, and A. Kenworthy, Nat. Rev. Mol. Cell. Biol. **2**, 444 (2001).

[7] T. Yoshigaki, Phys. Rev. E. **75**, 041901 (2007).

[8] R. J. Jackman, J. L. Wilbur, and G. M. Whitesides, Science, **269**, 664 (1995).

[9] M. Wu, C. Li, C. Hu, Y. Chang, Y. Liaw, L. Hunag, C. Chang, T. Tsong, T. Hsu, Phys. Rev. B **74**, 125424 (2006).

[10] P. Ferrin, S. Kandoi, A. U. Nilekar, and M. Mavrikakis, Surface Science **606**, 679 (2012).

[11] P. S. Burada, P. Hänggi, F. Marchesoni, G. Schmid, and P. Talkner, Chem. Phys. Chem. **10**, 45 (2009).

[12] R. Zwanzig, J. Chem. Phys. **96**, 3926 (1992).

[13] M. H. Jacobs, *Diffusion Processes*, (Springer, New York, 1967).

[14] P. Kalinay and J. K. Percus, J. Chem. Phys. **122**, 204701 (2005); Phys. Rev. E **74**, 041203 (2006).

[15] L. Dagdug and I.Pineda, J. Chem. Phys. **137**, 024107 (2012).

[16] I. Pineda, J. Alvarez-Ramirez and L. Dagdug, J. Chem. Phys. **137**, 174103 (2012).

[17] F. Li and B. Ai, Phys. Rev. E **87**, 062128 (2013).

[18] B. Ai and J. Wu, J. Chem. Phys. **139**, 034114 (2013).

[19] E. Yariv, H. Brenner, and S. Kim, SIAM J. Appl. Math. **64**, 1099 (2004).

[20] R. M. Bradley, Phys. Rev. E **80**, 061142 (2009).

[21] A. M. Berezhkovskii and A. Szabo, J. Chem. Phys. **135**, 074108 (2011).

[22] G. Chacón-Acosta, I. Pineda and L. Dagdug, J. Chem. Phys. In press (2013).

[23] H. Risken, *The Fokker-Planck equation. Methods of solution and applications*, (Springer, Berlin,1989).

[24] N. G. van Kampen, J. Stat. Phys. **44**, 1 (1986).

[25] P. Castro-Villareal, J. Stat. Mech. **P08006**, 1742 (2010).

[26] J. Balakrishnan, Phys. Rev. E, **61**, 4648 (2000).

[27] J. Faraudo, J. Chem. Phys., **116**, 5831 (2002).

[28] C. Chevalier, F. Debbasch, J. Stat. Phys. **131**, 717 (2008); F. Debbasch, C. Chevalier, IMS Collection **4**, 85 (2008).

[29] P. H. Roberts and H. D. Ursell, Phil. Trans. R. Soc. Lond. A **252**, 317 (1960).

[30] B. M. Aizendub, N. D. Gershon, Biophys. J., **38**, 287 (1982); S. Gustafsson, B. Halle, J. Chem. Phys., **106**, 1880 (1997).

[31] A. Naji, F. L. H. Brown, J. Chem. Phys., **126**, 235103 (2007).

[32] M. P. do Carmo, *Differential Geometry of Curves and Surfaces*, (Prentice-Hall, 1976).

[33] V. A. Toponogov, *Differential Geometry of Curves and Surfaces. A Concise Guide*, (Birkhäuser, 2006).

[34] R. Lipowsky E. Sackmann, *Structure and Dynamics of Membranes*, (Handbook of Biological Physics, Elsevier Science B.V. 1995).