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LATERAL DIFFUSION NEAR THE INTERFACE OF TWO IMMISCIBLE LIQUIDS*

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Dedicated to Professor Peter Talkner on the occasion of his 60th birthday

Diffusion coefficient of a particle diffusing near the interface of two immiscible liquids varies when the particle crosses the interface. We show how the problem of lateral diffusion in such a system can be reduced to that of finding the distribution of the cumulative residence time spent by the particle in one of the layers. The latter problem can be solved with relative ease since the distribution is determined by the one-dimensional motion in the direction normal to the interface. The approach is utilized to find an exact solution for the Fourier–Laplace transform of the lateral propagator, the effective medium approximation for this propagator, and the time-dependent behavior of the lateral diffusion coefficient in several special cases.

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1. Introduction

Lateral diffusion in multilayer media plays an important role in different biophysical problems [1–8]. Specific feature of lateral diffusion in such media is that the lateral diffusion coefficient changes when the particle goes from one layer to the other. In the present paper I continue our analysis of lateral diffusion in multilayer media initiated in Ref. [9]. The major idea of the approach developed in that reference is to reduce the problem of lateral diffusion to that of finding the distribution of the cumulative residence times

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spent by the particle in different layers. The latter problem can be solved with relative ease since the distribution is determined by the one-dimensional motion of the particle in the direction normal to the layers.

Our analysis in Ref. [9] is mainly focused on a situation in which neighboring layers are separated by membranes, and transitions between them can be described by a kinetic scheme. This scheme is used to find the distribution of the cumulative residence times. In the absence of the separating membranes (*i.e.*, formally when permeability of the membranes tend to infinity) the formalism based on the kinetic scheme fails, and another approach should be used to find the distribution of the cumulative residence times and then the lateral propagator and the lateral diffusion coefficient.

This is the subject of the present paper. In the next section we show how this can be done for two semi-infinite layers of immiscible liquids when a diffusing particle starts from the interface. One of the main results of this section is an exact solution for the Fourier–Laplace transform of the lateral propagator given in Eq. (9). This is derived using the expression for the Fourier–Laplace transform of the propagator in terms of the double Laplace transform of the probability density of the cumulative residence time in Eq. (5) and the expression for the double Laplace transform of this probability density in Eq. (8), which is derived in Appendix. The result in Eq. (9) is then used to find the effective medium approximation for the lateral propagator, Eq. (12), and the expression in Eq. (11), which gives the effective lateral diffusion coefficient as a function of the particle diffusion coefficients in the two layers. Some of the results obtained in Section 2 are generalized in Section 3, where we also discuss the relation between lateral and normal diffusion of the particle.

This paper deals with fluctuations of the diffusion coefficient due to the particle transitions between the two layers with different viscosities. It is worth noting that a more general problem of diffusion of Brownian particles in a fluid with fluctuating viscosity has been studied recently by Talkner and his colleagues Luczka, Hanggi, and Rozenfeld [10, 11].

2. Formalism

Consider a diffusing particle that starts from the interface of two semi-infinite layers of immiscible liquids (Fig. 1). The system of coordinates is chosen so that the particle starts from the origin, and the interface corresponds to the plane $z = 0$. Layers with positive and negative values of the z-coordinate will be referred to as layers 1 and 2, respectively. The particle diffusion coefficients in the two layers are denoted as D_1 and D_2. For certainty we assume that $D_1 > D_2$.

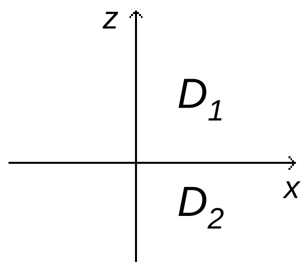

Fig. 1. Schematic representation of two semi-infinite layers of immiscible liquids. The plane $z = 0$ corresponds to the separating interface.

The lateral propagator at time t depends on how much time the particle has spent in each layer. If the cumulative residence time spent by the particle in layer 1 is τ and the corresponding time spent in layer 2 is $(t - \tau)$, the lateral propagator is

$$g(x|\tau, t-\tau) = \frac{\exp\left(-\frac{x^2}{4[D_1\tau + D_2(t-\tau)]}\right)}{\sqrt{4\pi[D_1\tau + D_2(t-\tau)]}}\,. \qquad (1)$$

Time τ is a random variable. Its distribution is determined by diffusion along the z-coordinate. Let $f(\tau|t)$ be the probability density for time τ on condition that the total observation time is t. The lateral propagator at time t is the propagator in Eq. (1) averaged over τ

$$g(x,t) = \int_0^t g(x|\tau, t-\tau) f(\tau|t) d\tau\,. \qquad (2)$$

The Fourier transform of this propagator is given by

$$g(k,t) = \int_{-\infty}^{\infty} e^{ikx} g(x,t) dx = \int_0^t e^{-k^2[D_1\tau + D_2(t-\tau)]} f(\tau|t) d\tau\,. \qquad (3)$$

Introducing the double Laplace transform of the probability density $f(\tau|t)$

$$\hat{f}(\lambda|\sigma) = \int_0^\infty e^{-\sigma t} dt \int_0^t e^{-\lambda\tau} f(\tau|t) d\tau\,, \qquad (4)$$

we can write the Laplace transform of the propagator in Eq. (3) in the form

$$\hat{g}(k,s) = \int_0^\infty e^{-st} g(k,t) dt = \hat{f}(\lambda = (D_1 - D_2)k^2 | \sigma = s + D_2 k^2)\,. \qquad (5)$$

This expression shows that the Fourier–Laplace transform of the lateral propagator is the double Laplace transform of the probability density $f(\tau|t)$ with correctly chosen values of the transform parameters, $\lambda = (D_1 - D_2)k^2$ and $\sigma = s + D_2 k^2$.

Thus the problem of finding the lateral propagator can be reduced to that of finding the transform $\hat{f}(\lambda|\sigma)$, which is a one-dimensional problem that can be solved with relative ease. To find this transform assume that being in layer 1 the particle annihilates with the rate λ. Then the particle survival probability, $S_\lambda(t)$, is given by

$$S_\lambda(t) = \int_0^t e^{-\lambda\tau} f(\tau|t) d\tau \, . \tag{6}$$

The double transform of interest is the Laplace transform of the survival probability

$$\hat{f}(\lambda|\sigma) = \int_0^\infty e^{-\sigma t} S_\lambda(t) dt = \hat{S}_\lambda(\sigma) \, . \tag{7}$$

The Laplace transform $\hat{S}_\lambda(\sigma)$ is found in Appendix A. Using this transform we obtain

$$\hat{f}(\lambda|\sigma) = \hat{S}_\lambda(\sigma) = \frac{\sqrt{D_1\sigma} + \sqrt{D_2(\lambda+\sigma)}}{\sqrt{\sigma(\lambda+\sigma)}\left[\sqrt{D_1(\lambda+\sigma)} + \sqrt{D_2\sigma}\right]} \, . \tag{8}$$

Since the transform $\hat{f}(\lambda|\sigma)$ is known, we can find the propagator in Eq. (5), which can be written in the form convenient for further discussion

$$\hat{g}(k,s) = \frac{1}{s + k^2 \frac{\sqrt{D_1^3(s+D_2k^2)} + \sqrt{D_2^3(s+D_1k^2)}}{\sqrt{D_1(s+D_2k^2)} + \sqrt{D_2(s+D_1k^2)}}} \, . \tag{9}$$

One can see that $\hat{g}(0,s) = 1/s$ as it should be because of the probability conservation. Retaining the lowest order terms in small-s and small-k expansion we obtain the effective medium approximation for the propagator

$$\hat{g}_{\mathrm{EM}}(k,s) = \frac{1}{s + D_{\mathrm{eff}} k^2} \, , \tag{10}$$

where D_{eff} is the effective diffusion coefficient given by

$$D_{\mathrm{eff}} = \frac{D_1^{3/2} + D_2^{3/2}}{D_1^{1/2} + D_2^{1/2}} \, . \tag{11}$$

Inverting the Fourier–Laplace transform in Eq. (10) we obtain

$$g_{\mathrm{EM}}(x,t) = \frac{1}{\sqrt{4\pi D_{\mathrm{eff}} t}} \exp\left(-\frac{x^2}{4D_{\mathrm{eff}} t}\right) \tag{12}$$

which is the effective medium approximation for the lateral propagator in the x-, t-variables.

Effective diffusion coefficient in Eq. (11) has a transparent physical interpretation. To show this we introduce the average time spent by the diffusing particle in layer $1, \bar{\tau}(t)$. Formally this time is defined by

$$\bar{\tau}(t) = \int_0^t \tau f(\tau|t) d\tau \,. \tag{13}$$

The Laplace transform of this time can be written in terms of the double transform $\hat{f}(\lambda|\sigma)$

$$L\left\{\bar{\tau}(t)\right\} = \int_0^\infty e^{-\sigma t} \bar{\tau}(t) dt = -\left.\frac{\partial \hat{f}(\lambda|\sigma)}{\partial \lambda}\right|_{\lambda=0} . \tag{14}$$

Using the expression for $\hat{f}(\lambda|\sigma)$ in Eq. (8) we obtain

$$L\left\{\bar{\tau}(t)\right\} = \frac{D_1^{1/2}}{\left(D_1^{1/2} + D_2^{1/2}\right)\sigma^2} \,. \tag{15}$$

Inverting this transform we find

$$\bar{\tau}(t) = \frac{D_1^{1/2}}{D_1^{1/2} + D_2^{1/2}} t \,. \tag{16}$$

This allows us to write D_{eff} in Eq. (11) in the form

$$D_{\mathrm{eff}} = \frac{1}{t}[D_1 \bar{\tau}(t) + D_2(t - \bar{\tau}(t))] \tag{17}$$

which represents D_{eff} as a weighted sum of D_1 and D_2 with the weight factors given by the fractions of the observation time that the diffusing particle has spent in each layer.

When the particle starts from the interface of two semi-infinite layers, the lateral diffusion coefficient is independent of time and given by the expression in Eq. (11) for the entire range of time from zero to infinity. To see this, one can use the propagator in Eq. (9) to find the Laplace transform of the mean-squared displacement $\langle x^2(t)\rangle$ which is given by

$$L\left\{\langle x^2(t)\right\} = -\left.\frac{\partial^2 \hat{g}(k,s)}{\partial k^2}\right|_{k=0} = \frac{2D_{\mathrm{eff}}}{s^2} \tag{18}$$

with D_{eff} given in Eq. (11). Inverting this, one finds that $\langle x^2(t)\rangle = 2D_{\mathrm{eff}}t$ for all times.

In general, diffusion coefficients in non-uniform media are functions of time. For example, when the particle starts not from the interface, its diffusion coefficient is equal to the diffusion coefficient in the initial layer at short times and approaches D_{eff} as $t \to \infty$. As another example consider the case of layers of finite thickness, L_1 and L_2, respectively. Here the diffusion coefficient of the particle, which starts from the interface, changes from D_{eff} in Eq. (11) at short times (when the particle does not feel that the layers have finite thickness) to the long-time asymptotic value D_∞ given by

$$D_\infty = \frac{D_1 L_1 + D_2 L_2}{L_1 + L_2}\,. \tag{19}$$

This is analogous to the expression for D_{eff} in Eq. (17) in the sense that it gives D_∞ as a weighted sum of D_1 and D_2. Moreover, the weight factors, $L_1/(L_1+L_2)$ and $L_2/(L_1+L_2)$, are the equilibrium probabilities of finding the particle in each layer and, hence, fractions of the long observation time, which the particle spends in the layers. We discuss transient behavior of the diffusion coefficient in the next section.

3. Generalizations and discussion

In this section we discuss three generalizations of the results derived in Section 2. We begin with the case when the particle starts not from the interface. Suppose, it starts in layer 1 at distance z_0 from the interface. We derive the lateral diffusion coefficient of the particle as a function of time. The result can be obtained following the way discussed in Section 2. Instead we use another approach which is simpler because it deals with the mean-squared displacement, $\langle x^2(t|z_0)\rangle$, and not with the propagator.

Let $\varphi(t|z_0)$ be the probability density of the first passage time from z_0, $z_0 > 0$, to the interface

$$\varphi(t|z_0) = \frac{z_0}{\sqrt{4\pi D_1 t^3}} \exp\left(-\frac{z_0^2}{4D_1 t}\right)\,. \tag{20}$$

Then

$$\Phi(t|z_0) = \int_t^\infty \varphi(t'|z_0)dt' = \operatorname{erf}\left(\frac{z_0}{\sqrt{4D_1 t}}\right)\,, \tag{21}$$

where $\mathrm{erf}(z)$ is the error function [12], is the probability that the particle has spent the observation time t in layer 1. Using these two functions we can write the mean-squared displacement as

$$\langle x^2(t|z_0)\rangle = 2D_1 t\,\Phi(t|z_0) + 2\int_0^t \varphi(t'|z_0)[D_1 t' + D_{\mathrm{eff}}(t-t')]dt' \tag{22}$$

and explicitly

$$\langle x^2(t|z_0)\rangle = 2D_{\mathrm{eff}}t + 2(D_1 - D_{\mathrm{eff}})\int_0^t \mathrm{erf}\left(\frac{z_0}{\sqrt{4D_1 t'}}\right)dt'\,. \tag{23}$$

We define the time-dependent diffusion coefficient, $D(t|z_0)$, by

$$D(t|z_0) = \frac{1}{2}\frac{d\langle x^2(t|z_0)\rangle}{dt}\,. \tag{24}$$

Substituting here $\langle x^2(t|z_0)\rangle$ given in Eq. (23) we obtain

$$D(t|z_0) = D_{\mathrm{eff}} + (D_1 - D_{\mathrm{eff}})\mathrm{erf}\left(\frac{z_0}{\sqrt{4D_1 t}}\right)\,. \tag{25}$$

This describes transient behavior of the lateral diffusion coefficient, which varies from D_1 to D_{eff} as t goes from zero to infinity.

Now we generalize the results obtained in Section 2 to the case of finite thickness of the layers. We denote the thickness by L_1 and L_2 and assume that the planes $z = L_1$ and $z = -L_2$ may be considered as reflecting boundaries. We also assume that the particle starts from the interface at $z = 0$. We again can find the double Laplace transform of the probability density of the cumulative residence time in layer 1 following the way discussed in Appendix. This leads to

$$\hat{f}(\lambda|\sigma) = \frac{\sqrt{D_1\sigma}\coth\left(\sqrt{\frac{\sigma}{D_2}}L_2\right) + \sqrt{D_2(\lambda+\sigma)}\coth\left(\sqrt{\frac{\lambda+\sigma}{D_1}}L_1\right)}{\sqrt{\sigma(\lambda+\sigma)}\left[\sqrt{D_1(\lambda+\sigma)}\coth\left(\sqrt{\frac{\sigma}{D_2}}L_2\right) + \sqrt{D_2\sigma}\coth\left(\sqrt{\frac{\lambda+\sigma}{D_1}}L_1\right)\right]}\,. \tag{26}$$

When L_1 and L_2 tend to infinity this $\hat{f}(\lambda|\sigma)$ takes the asymptotic form given in Eq. (8).

The double Laplace transform in Eq. (26) can be used to find the Fourier–Laplace transform of the propagator by means of Eq. (5). Then the transform of the propagator, in its turn, can be used to find the Laplace transform of the time-dependent diffusion coefficient, $D(t)$, by the relation

$$\hat{D}(s) = -\frac{1}{2}\left.\frac{\partial^3 \hat{g}(k,s)}{\partial k^2 \partial s}\right|_{k=0}. \tag{27}$$

This leads to

$$\hat{D}(s) = \frac{D_1^{3/2}\tanh\left(\sqrt{\frac{s}{D_1}}L_1\right) + D_2^{3/2}\tanh\left(\sqrt{\frac{s}{D_2}}L_2\right)}{s\left[D_1^{1/2}\tanh\left(\sqrt{\frac{s}{D_1}}L_1\right) + D_2^{1/2}\tanh\left(\sqrt{\frac{s}{D_2}}L_2\right)\right]}. \tag{28}$$

The particle does not feel the presence of the boundaries at short times, and its lateral diffusion coefficient is equal to D_{eff} given in Eq. (11). One can see this from Eq. (28) since $\hat{D}(s) = D_{\text{eff}}/s$ as $s \to \infty$. The diffusion coefficient approaches its asymptotic value D_∞ given in Eq. (19) as $t \to \infty$. This follows from the small-s behavior of $\hat{D}(s)$ in Eq. (28), $\hat{D}(s) \approx D_\infty/s$ as $s \to 0$. Inverting the transform in Eq. (28) numerically one can find transient behavior of $D(t)$ over the entire range of time.

The formalism developed in Section 2 can be generalized to the case when the particle annihilates with the rates γ_1 and γ_2 in layers 1 and 2, respectively. In this case the propagator in Eq. (1) should be replaced by

$$g(x|\tau, t-\tau) = \frac{\exp\left(-\frac{x^2}{4[D_1\tau + D_2(t-\tau)]} - \gamma_1\tau - \gamma_2(t-\tau)\right)}{\sqrt{4\pi[D_1\tau + D_2(t-\tau)]}}. \tag{29}$$

The Fourier–Laplace transform of this propagator averaged over τ again can be expressed in terms of the double Laplace transform of $f(\tau|t)$. The result is

$$\hat{g}(k,s) = \hat{f}(\lambda = (D_1 - D_2)k^2 + \gamma_1 - \gamma_2 | \sigma = s + D_2 k^2 + \gamma_2), \tag{30}$$

where $\hat{f}(\lambda|\sigma)$ is given in Eq. (8). Taking small-s and small-k limit of this propagator and inverting the result we obtain the effective medium approximation for the propagator

$$g_{\text{EM}}(x,t) = \frac{1}{\sqrt{4\pi D_{\text{eff}} t}} \exp\left(-\frac{x^2}{4D_{\text{eff}} t} - \gamma_{\text{eff}} t\right). \tag{31}$$

Here γ_{eff} is the effective annihilation rate given by

$$\gamma_{\text{eff}} = \frac{1}{t}[\gamma_1 \bar{\tau}(t) + \gamma_2(t - \bar{\tau}(t))] = \frac{\gamma_1 D_1^{1/2} + \gamma_2 D_2^{1/2}}{D_1^{1/2} + D_2^{1/2}}, \tag{32}$$

where we have used the result for the average residence time $\bar{\tau}(t)$ given in Eq. (16). The propagator in Eq. (31) is a generalization of the propagator in Eq. (12) to the case of diffusion in absorbing media.

Finally, we compare lateral diffusion of the particle with that in the direction normal to the interface separating two semi-infinite layers of immiscible liquids (Fig. 1) assuming that the particle starts from the interface. The particle propagator in the direction normal to the interface, $G_z(z,t)$, is given by

$$G_z(z,t) = \frac{\exp\left(-\frac{z^2}{4D_1 t}\right) H(z) + \exp\left(-\frac{z^2}{4D_2 t}\right) H(-z)}{\sqrt{\pi t}\left(\sqrt{D_1} + \sqrt{D_2}\right)}, \tag{33}$$

where $H(z)$ is the Heaviside step function. Note that the average time spent by the particle in layer 1, $\bar{\tau}(t)$, given in Eq. (16) can be found using the propagator in Eq. (33) by the relation [13]

$$\bar{\tau}(t) = \int_0^t dt' \int_0^\infty G_z(z,t')dt' = \frac{D_1^{1/2}}{D_1^{1/2} + D_2^{1/2}} t\,. \tag{34}$$

In contrast to the lateral diffusion the average displacement in normal direction, $\langle z(t) \rangle$, is not equal to zero and is given by

$$\langle z(t) \rangle = \int_{-\infty}^{\infty} z G_z(z,t) dz = \frac{2}{\sqrt{\pi}} \left(D_1^{1/2} - D_2^{1/2}\right) \sqrt{t}\,. \tag{35}$$

However, the second moments of the displacement in both directions are equal to

$$\langle z^2(t) \rangle = \int_{-\infty}^{\infty} z^2 G_z(z,t) dz = 2D_{\mathrm{eff}} t = \langle x^2(t) \rangle\,. \tag{36}$$

One can easily find and compare higher moments of the displacement in both directions using the propagators in Eqs. (9) and (33).

In summary, in this paper we analyze lateral diffusion near the interface of two immiscible liquids. The approach we use is based on the reduction of the problem of lateral diffusion to that of finding the distribution of the cumulative residence time spent by the diffusing particle in one of the layers. The latter problem can be solved with relative ease since the distribution is determined by the one-dimensional motion of the particle in the direction normal to the layers.

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Appendix A

The Laplace transform of the survival probability $S_\lambda(t)$

To find the Laplace transform of the survival probability $S_\lambda(t)$ consider the particle diffusion along the z-coordinate assuming that the particle annihilates with the rate λ when $z > 0$. The particle survival probability $S_\lambda(t)$ can be written in terms of its Green's function, $G_\lambda(z,t)$, which satisfies

$$\frac{\partial G_\lambda}{\partial t} = \frac{\partial}{\partial z}\left\{[D_1 H(z) + D_2 H(-z)]\frac{\partial G_\lambda}{\partial z}\right\} - \lambda H(z) G_\lambda\,, \tag{A.1}$$

where $H(z)$ is the Heaviside step function. The Green's function vanishes as $|z| \to \infty$ and approaches $\delta(z)$ as $t \to 0$. The survival probability is given by

$$S_\lambda(t) = \int_{-\infty}^{\infty} G_\lambda(z,t)dz\,. \tag{A.2}$$

The Laplace transform of the propagator

$$\hat{G}_\lambda = \hat{G}_\lambda(z,\sigma) = \int_0^\infty e^{-\sigma t} G_\lambda(z,t)dt \tag{A.3}$$

satisfies

$$\sigma\hat{G}_\lambda - \delta(z) = \frac{d}{dz}\left\{[D_1 H(z) + D_2 H(-z)]\frac{d\hat{G}_\lambda}{dz}\right\} - \lambda H(z)\hat{G}_\lambda\,. \tag{A.4}$$

Solving this equation we find

$$\hat{G}_\lambda = \frac{\exp\left(-\sqrt{\frac{\lambda+\sigma}{D_1}}z\right)H(z) + \exp\left(\sqrt{\frac{\sigma}{D_2}}z\right)H(-z)}{\sqrt{D_1(\lambda+\sigma)} + \sqrt{D_2\sigma}}\,. \tag{A.5}$$

Using this solution we obtain

$$\hat{S}_\lambda(\sigma) = \int_{-\infty}^{\infty} \hat{G}_\lambda(z,\sigma)dz = \frac{\sqrt{D_1\sigma} + \sqrt{D_2(\lambda+\sigma)}}{\sqrt{\sigma}\sqrt{\lambda+\sigma}\left[\sqrt{D_1(\lambda+\sigma)} + \sqrt{D_2\sigma}\right]} \,. \qquad \text{(A.6)}$$

This allows us to find the double transform $\hat{f}(\lambda|\sigma)$ given in Eq. (8).

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