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Translational and rotational drag coefficients for a disk moving in a liquid membrane associated with a rigid substrate

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A simple phenomenological drag relation is used to characterize weak dynamic coupling of a liquid membrane to an adjacent solid substrate. With this linear velocity-dependent drag relation, the inertialess equations of motion for membrane flow are easily solved for steady translation and rotation of a disk-like particle. The resulting drag coefficients exhibit functional dependencies on the dimensionless particle size very similar to the relations obtained for particle motion in a membrane bounded by semi-infinite liquid domains (Hughes *et al.* 1981), although the scaling of particle size is different. Within this phenomenological approach, diffusivities of molecular probes in membranes can be used to investigate the intrinsic molecular drag at a solid–liquid membrane interface and to estimate properties of thin lubricating liquid layers between membrane and substrate.

1. Introduction

Recently, there has been significant research and technological interest in coupling molecularly thin liquid-crystal membranes to solid substrates (Barlow 1980; Roberts & Pitt 1983). One technique, known as Langmuir–Blodgett film deposition, utilizes a monolayer film balance and immersion/withdrawal of the substrate through the monolayer interface to produce one or more condensed layers on the substrate (Blodgett & Langmuir 1937). This technique (as well as others which involve chemical treatment of the substrate for subsequent bonding of molecular layers) takes advantage of the amphiphilic (surfactant) character of these molecules. A specific approach used to study the dynamic interaction between membrane and substrate is to measure the translational (surface) diffusivity of molecular probes incorporated in the membrane (Seul & McConnell 1986). Changes observed in diffusivity between a free (unsupported) membrane and one associated with a rigid substrate reflect both the degree of membrane coupling to the substrate and alterations in membrane state in the presence of the substrate. Hence, we have set out to derive relations for particle mobility (translational and rotational) in an anisotropic fluid layer as a function of an intrinsic frictional parameter which can be used to characterize weak membrane–substrate interactions.

Self-diffusion of molecular probes in molecularly thin membranes has been studied for nearly twenty years by biophysical scientists as a method for determination of

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viscous properties of both synthetic and natural (cell) membranes (Edidin 1974; Vaz, Derzko & Jacobson 1982). Einstein kinetic relations have been used to derive estimates of molecular mobility (velocity/drag) from measurements of particle diffusivities ($D = kT \times$ mobility). The difficulty has been that analysis of the fluid mechanics of particle motion in an anisotropic (two-dimensional) liquid cannot yield a simple linear velocity dependence for drag in the inertialess limit if the velocity field is required to approach zero at infinite distance from the particle. Further, theoretical physicists have concluded that viscosity for a two-dimensional condensed liquid should diverge weakly as the logarithm of size of the domain (Forster, Nelson & Stephen 1977). Saffman (1976) recognized that this problem of two-dimensional symmetry will always be broken by transfer of momentum to an adjacent bulk (three dimensional) liquid. Even though the 'Stokes paradox' and the divergence of surface viscosity are eliminated by coupling to the third dimension, solution of the inertialess Navier–Stokes equations is no simple task because of the dual boundary conditions (i.e. interfacial drag of the bulk liquid on the membrane exterior to the particle and the no-slip requirement of uniform velocity for the liquid over the disk surface of the particle). Saffman examined the limit of large membrane viscosity (or low viscosity in the adjacent bulk liquid) with a singular perturbation approximation. His results show that in this limit there is essentially no dependence of drag on particle size (only a weak logarithmic effect). Hughes, Pailthorpe & White (1981) undertook the heroic task of obtaining the exact solution to the equations of motion for any combination of membrane and adjacent liquid viscosities. Even with their elegant results, it is necessary to introduce a series expansion to approximate the drag coefficients. In these analyses, the important parameter is a characteristic length δ for the flow-field region dominated by membrane dissipation, which is given by the ratio of membrane (two-dimensional) viscosity η_m (dyn s/cm) to the adjacent liquid viscosity μ (dyn s/cm^2), i.e. $\delta \equiv \eta_m/\mu$. The extensive (size-dependent) properties of the drag coefficients reduce to functions of the ratio of the particle radius a to the length δ. (Note: in the analyses of Saffman and Hughes *et al.* the membrane is treated as a thin liquid region of viscosity μ_m thickness h_m, with constant velocity over any cross-section. Without gradients across the membrane, thickness is of no consequence and the material can be treated as anisotropic with a surface viscosity $\eta_m \equiv \mu_m h_m$.)

In this paper, we examine the effect of a rigid substrate in close proximity to a liquid membrane. We assume that transport of momentum into the third dimension is dominated by the presence of the substrate and can be modelled by simple interfacial drag which is proportional to membrane velocity. It will be seen that this substrate drag relation leads to ready solution of the two-dimensional equations of motion of the inertialess fluid membrane. The drag coefficients, obtained as simple analytical expressions valid for all particle sizes, provide useful relations for estimating frictional coefficients at molecular interfaces from diffusivity measurements. If a thin lubricating layer (thickness h) of bulk liquid exists between the membrane and substrate, the same drag coefficients can be used with a surface frictional coefficient given by μ/h provided that the layer thickness $h \ll \delta$. It is also interesting to note that the drag coefficients for a thin liquid layer between membrane and substrate closely approximate the results of Hughes *et al.* (1981) when the layer thickness is arbitrarily set equal to δ. As such, it is possible to estimate the effect of a semi-infinite bathing liquid above the membrane–solid substrate complex.

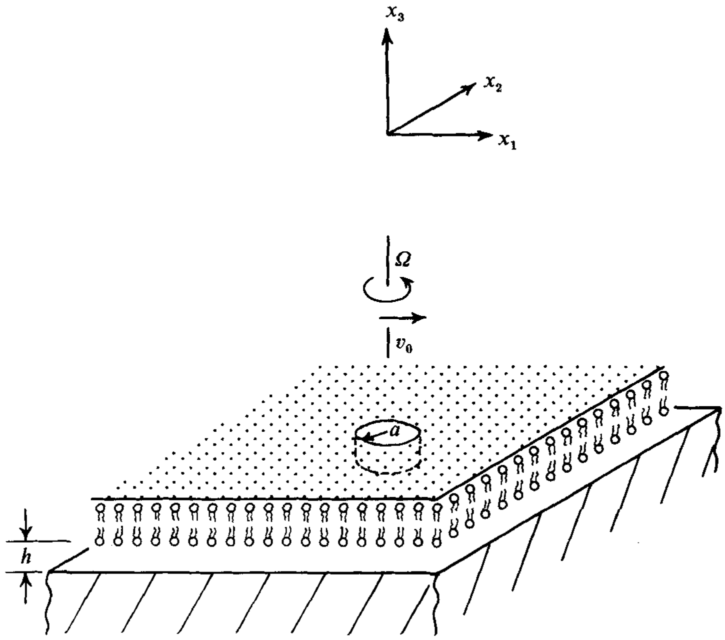

FIGURE 1. Schematic illustration of a disk-like particle (moving by either translation or rotation) in a condensed liquid membrane adsorbed to a solid substrate. Here, a thin lubricating layer of liquid is shown between the membrane and substrate.

2. Surface flow fields for a disk moving (by translation or rotation) in a liquid membrane which drags on a solid substrate

Figure 1 schematically illustrates a surfactant double-layer membrane adsorbed onto a solid substrate with a moving disk-like probe. Bilayer membranes are formed as condensed liquid crystals with very small area compressibility (Evans & Needham 1987); hence, the divergence of the velocity field in the membrane plane is zero,

$$\bar{\nabla}\cdot\mathbf{v} = \frac{\partial v_1}{\partial x_1}+\frac{\partial v_2}{\partial x_2} \equiv 0. \tag{2.1}$$

Here, $\bar{\nabla}$ defines the gradient operator in the plane of the surface and (v_1, v_2) are the components of velocity in the surface at a local point (x_1, x_2) as shown in figure 1. For steady inertialess flow, the equations of motion are given by the Stokes approximation to the momentum equation for the fluid membrane subject to interfacial drag (shear stress $\boldsymbol{\sigma}$) from the solid substrate,

$$\bar{\nabla}\tau_0+\eta_{\mathrm{m}}\bar{\nabla}^2 v = \boldsymbol{\sigma}, \tag{2.2}$$

where τ_0 is the two-dimensional isotropic tension (dyn/cm) in the membrane (i.e. negative surface pressure); η_{m} is the membrane surface viscosity (dyn s/cm). We consider that the association of the membrane with the substrate is weak (not strongly bonded); thus, a reasonable approximation is to assume that the interfacial drag of the membrane on the substrate is proportional to the local velocity of the membrane relative to the substrate, i.e.

$$\boldsymbol{\sigma} \equiv b_{\mathrm{s}}\,\boldsymbol{v}, \tag{2.3}$$

where b_s (dyn s/cm^3) is an intrinsic coefficient of friction between the membrane and substrate. Here, the frictional interaction is treated as isotropic in the surface plane; however, it is possible to introduce directional properties for a structured substrate.

By taking the two-dimensional curl of (2.2) and introducing the vorticity component for surface flow,

$$\omega \equiv \left(\frac{\partial v_1}{\partial x_2} - \frac{\partial v_2}{\partial x_1}\right),$$

the equations of motion reduce to a single scalar equation,

$$\bar{\nabla}^2 \omega = \left(\frac{b_s}{\eta_m}\right)\omega, \tag{2.4}$$

with solutions for cylindrical symmetry given in part by modified Bessel functions and angular harmonics, which are regular at infinite distance from the disk.

For steady translation of the disk, the velocity field in cylindrical coordinates is of the form

$$v_r = \frac{1}{r} f(r) \cos\phi, \quad v_\phi = -\frac{\partial f(r)}{\partial r} \sin\phi, \quad \omega = f_\omega(r) \sin\phi, \tag{2.5}$$

where
$$f_\omega(r) \equiv -\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial f}{\partial r}\right) + \frac{f}{r^2}.$$

Therefore, the solution for $r \geqslant a$, which satisfies the condition that the velocity approach zero at large distances, is given by

$$\left.\begin{aligned} v_r &= \left\{\frac{C_1}{r^2} + \frac{C_2}{r} K_1\left(\frac{\epsilon r}{a}\right)\right\} \cos\phi, \\ v_\phi &= \left\{\frac{C_1}{r^2} + \frac{C_2}{r}\left[\left(\frac{\epsilon r}{a}\right) K_0\left(\frac{\epsilon r}{a}\right) + K_1\left(\frac{\epsilon r}{a}\right)\right]\right\} \sin\phi, \end{aligned}\right\} \tag{2.6}$$

where K_0 and K_1 are modified Bessel functions of the second kind, orders zero and one; a is the radius of the disk; and ϵ is a dimensionless parameter defined by

$$\epsilon \equiv a\left(\frac{b_s}{\eta_m}\right)^{\frac{1}{2}}. \tag{2.7}$$

The coefficients (C_1, C_2) are determined by the no-slip velocity boundary condition at $r = a$,

$$v_r = v_0 \cos\phi, \quad v_\phi = -v_0 \sin\phi,$$

which leads to

$$C_1 = v_0 a^2\left[1 + \frac{2K_1(\epsilon)}{\epsilon K_0(\epsilon)}\right], \quad C_2 = -\frac{2v_0 a}{\epsilon K_0(\epsilon)}. \tag{2.8}$$

Equations (2.6) and (2.8) specify the surface flow field for the disk translating with steady velocity v_0.

Similarly, the solution for steady rotation of the disk, which satisfies the approach to zero at large distances, is given by the simple result,

$$v_r \equiv 0, \quad v_\phi = C_1 K_1\left(\frac{\epsilon r}{a}\right), \tag{2.9}$$

where the coefficient C_1 is obtained from the no-slip velocity boundary condition at $r = a$,

$$v_\phi = \Omega a, \quad C_1 = \frac{\Omega a}{K_1(\epsilon)}. \tag{2.10}$$

The next task is to determine the drag force and torque that is applied to the disk by the liquid membrane.

3. Drag force and torque applied to the disk due to motion in the liquid membrane

For uniform translational motion of the disk through the membrane, the membrane stress resultants that act on the disk are given by

$$\left.\begin{aligned} \tau_{rr} &= \left\{f_0 + 2\eta_m\left[\frac{1}{r}\frac{\partial f}{\partial r} - \frac{f}{r^2}\right]\right\}\cos\phi, \\ \tau_{r\phi} &= -\eta_m\left\{\frac{\partial^2 f}{\partial r^2} - \frac{1}{r}\frac{\partial f}{\partial r} + \frac{f}{r^2}\right\}\sin\phi, \end{aligned}\right\} \tag{3.1}$$

where the membrane isotropic tension τ_0 is $f_0 \cos\phi$ and is chosen to satisfy the equation of motion (2.2), i.e.

$$f_0 = \eta_m r\frac{\partial f_\omega}{\partial r} + b_s r\frac{\partial f}{\partial r}$$

and again

$$f_\omega \equiv -\nabla_r^2 f.$$

With (2.6) and (2.8), the stress resultant at $r = a$ are expressed as

$$\left.\begin{aligned} \frac{\tau_{rr}}{\eta_m \cos\phi} &= -\frac{v_0\,\epsilon}{a}\left\{\epsilon + \frac{2K_1(\epsilon)}{K_0(\epsilon)}\right\}, \\ \frac{\tau_{r\phi}}{\eta_m \sin\phi} &= \frac{2v_0\,\epsilon K_1(\epsilon)}{aK_0(\epsilon)}. \end{aligned}\right\} \tag{3.2}$$

The drag on the disk by the surrounding liquid membrane is the integral of the action of stress resultants around its perimeter,

$$F_D = -2a\int_0^\pi [\tau_{rr}\cos\phi - \tau_{r\phi}\sin\phi]\,d\phi.$$

From (3.2), the result is

$$F_D = \pi\eta_m\,\epsilon v_0\left[\epsilon + \frac{4K_1(\epsilon)}{K_0(\epsilon)}\right],$$

which gives the drag coefficient, λ_T, for translation:

$$\lambda_T = 4\pi\eta_m\left[\tfrac{1}{4}\epsilon^2 + \frac{\epsilon K_1(\epsilon)}{K_0(\epsilon)}\right].$$

If there is intrinsic drag of the disk on the substrate (again assumed to be proportional to particle velocity), the drag coefficient will be modified:

$$\lambda_T = 4\pi\eta_m\left[\tfrac{1}{4}\epsilon^2(1 + b_p/b_s) + \frac{\epsilon K_1(\epsilon)}{K_0(\epsilon)}\right] \tag{3.3}$$

where the intrinsic particle drag is assumed to be

$$F_{D_p} \equiv \pi a^2 b_p v_0.$$

Similarly, for uniform rotation of the disk, the relevant membrane stress resultant that acts on the disk is given by

$$\tau_{r\phi} = \eta_m \left(\frac{\partial v_\phi}{\partial r} - \frac{v_\phi}{r} \right),$$

and the torque imparted by the liquid membrane to the disk is

$$M = -\tau_{r\phi} 2\pi a^2.$$

From (2.9) and (2.10), we find

$$M = 2\pi a^2 \eta_m \Omega \left[2 + \frac{\epsilon K_0(\epsilon)}{K_1(\epsilon)} \right], \tag{3.4}$$

which leads to the rotational drag coefficient λ_R:

$$\lambda_R = 4\pi a^2 \eta_m \left[1 + \frac{\epsilon K_0(\epsilon)}{2K_1(\epsilon)} \right].$$

Again, if there is intrinsic drag of the particle on the substrate, the rotational drag coefficient is expressed as

$$\lambda_R = 4\pi a^2 \eta_m \left[1 + \frac{\epsilon K_0(\epsilon)}{2K_1(\epsilon)} + \left(\frac{b_p}{8b_s} \right) \epsilon^2 \right]. \tag{3.5}$$

If we neglect the intrinsic particle drag on the substrate, the drag coefficients λ_T and λ_R, normalized by $4\pi\eta_m$ and $4\pi\eta_m a^2$ respectively, depend only on ϵ, which is the dimensionless particle radius $\epsilon \equiv a(b/\eta_m)^{\frac{1}{2}}$.

4. Effect of thin layer of bulk liquid between membrane and substrate

It is expected that for a thin lubricating layer of liquid between the membrane and solid substrate, the radial and angular dependence of the velocity field will be dominated by the membrane flow field and decrease linearly across the liquid layer (from the value at the membrane to zero at the substrate surface). As such, the frictional coefficient b for coupling of the membrane to the substrate is simply given by $b_s = \mu/h$, where μ is the (three-dimensional) viscosity of the liquid and h is the thickness of the layer. This lubrication approximation is valid when the layer thickness h is much less than the characteristic length δ, i.e. $h \ll \eta_m/\mu$. Similarly, for a membrane between two solid surfaces with two lubricating liquid layers, the frictional coefficient b_s will be the sum of coefficients defined for each liquid layer. When h is arbitrarily set equal to $\delta \equiv \eta_m/(\mu_1+\mu_2)$ in the lubrication approximation, the dimensionless particle radius becomes identical to the definition used by Hughes *et al.* in their analysis of a membrane between semi-infinite liquid regions, i.e.

$$\epsilon = a\left(\frac{(\mu_1+\mu_2)}{\eta_m \delta} \right)^{\frac{1}{2}} = \frac{a(\mu_1+\mu_2)}{\eta_m}.$$

5. Results and discussion

Dimensionless translational and rotational mobilities ($4\pi\eta_m/\lambda_T$ and $4\pi\eta_m a^2/\lambda_R$ respectively) are plotted in figure 2 versus the dimensionless particle radius ϵ as

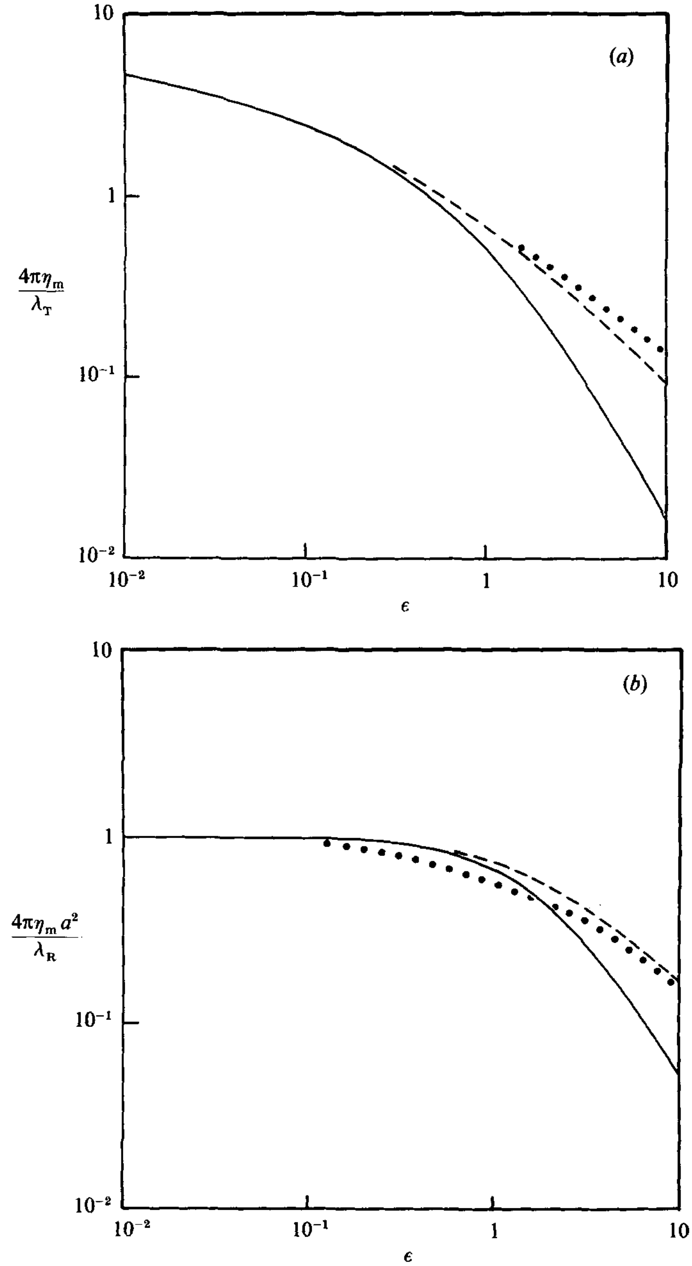

FIGURE 2. Dimensionless particle mobilities for (*a*) translation $(4\pi\eta_m/\lambda_T)$ and (*b*) rotation $(4\pi\eta_m a^2/\lambda_R)$ of a disk-like particle in a membrane are plotted versus the dimensionless particle radius ϵ. The solid curves are the results of this analysis given in (3.3) and (3.5) for a membrane weakly adsorbed to a solid substrate; the dashed curves are the results from (3.3) and (3.5) *excluding* the ϵ^2 terms; the dotted curves are results obtained previously by Hughes *et al.* (1981) for a membrane immersed in an infinite liquid domain.

given by (3.3) and (3.5). Also shown in figure 2 are the results obtained by Hughes *et al.* as a function of dimensionless radius (scaled differently as mentioned above). It is apparent that our results are the same as those of Hughes *et al.* for $\epsilon < 1$. Indeed, the drag coefficients remain close even for large $\epsilon (> 1)$ if we exclude the ϵ^2 terms in

(3.3) and (3.5), as shown in figure 2. The ϵ^2 terms are the direct particle drag on the substrate *and* the image interaction of the particle with the substrate which is produced by the potential (inviscid) flow in the membrane. The weak logarithmic dependence of the translational drag coefficient on particle size for small values of $\epsilon(<0.1)$ is common to all membrane analyses; this behaviour was initially derived and examined by Saffman (1976). The correlation between our results and those of Hughes *et al.* over a wide range of ϵ-values is not surprising; it shows that the kinematics of flow in the membrane surface (two-dimensional) strongly influences the flow field in the third dimension (this feature was also recognized by Hughes *et al.*).

Because of the similarity in functional characteristics of the drag coefficients for a membrane subject to simple substrate drag and a membrane immersed in an infinite liquid region, it is reasonable to make the *ad hoc* assumption that the presence of a semi-infinite bathing liquid on one side of a membrane closely associated with a solid substrate can be represented approximately by a simple frictional coefficient:

$$b_\infty \equiv \mu/\delta.$$

Therefore, the total drag from the substrate plus the semi-infinite bathing liquid is approximated by $b = (b_s + b_\infty)$, where b_s is the substrate–membrane frictional coefficient. The dimensionless particle radius ϵ is given by

$$\epsilon \approx a\left(\frac{b_s + b_\infty}{\eta_m}\right)^{\frac{1}{2}} = a\left(\frac{b_s + \mu/\delta}{\eta_m}\right)^{\frac{1}{2}}. \tag{5.1}$$

For a thin lubricating layer of liquid between membrane and substrate, this relation becomes

$$\epsilon \approx a\left(\frac{\mu(1/h + 1/\delta)}{\eta_m}\right)^{\frac{1}{2}}$$

when the liquid layer has the same viscosity as the semi-infinite bathing liquid. Consequently for $h \ll \delta$, the obvious result is that the presence of the bathing liquid can be neglected.

To illustrate the distance scales expected experimentally, consider bilayer membranes composed of diacyl lipid (surfactant) molecules for which a great deal of diffusion data exist. Translational diffusivities for probes of about 10^{-7} cm diameter in lipid bilayers are in the range of 10^{-8} to 10^{-7} cm^2/s when the bilayers are in the liquid state (Vaz *et al.* 1982). From the Einstein relation for particle diffusivity, $D = kT/\lambda_T$, the surface viscosity of a lipid bilayer membrane is estimated to be on the order of 10^{-6} to 10^{-5} dyn s/cm (P cm). As such, the characteristic length δ is greater than 10^{-4} cm (10^4 Å) in water. For bilayer membranes weakly adsorbed to solid substrates, separation distances are not yet known; however, preliminary studies with optical techniques indicate that lubricating liquid layers are much less than 10^{-6} cm (100 Å). Hence, the drag relations (3.3) and (3.5) are appropriate models and the superficial liquid can be neglected.

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