

## FAST TRACK COMMUNICATION

**Concentration fluctuations in binary fluid membranes****K Seki<sup>1</sup>, S Komura<sup>2,4</sup> and M Imai<sup>3</sup>**<sup>1</sup> National Institute of Advanced Industrial Science and Technology (AIST), Ibaraki 305-8565, Japan<sup>2</sup> Department of Chemistry, Faculty of Science, Tokyo Metropolitan University, Tokyo 192-0397, Japan<sup>3</sup> Department of Physics, Faculty of Science, Ochanomizu University, Tokyo 112-0012, JapanE-mail: [komura@comp.metro-u.ac.jp](mailto:komura@comp.metro-u.ac.jp)

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Online at [stacks.iop.org/JPhysCM/19/072101](http://stacks.iop.org/JPhysCM/19/072101)**Abstract**

We investigate the dynamics of critical fluctuations in binary fluid membranes using a two-dimensional hydrodynamic model with momentum decay to the surrounding water. In particular, the decay rate of concentration fluctuations is obtained analytically. In the limit of small wavenumber  $q$  with respect to the correlation length, the decay rate is proportional to  $q^2$ , as usual. In the large- $q$  limit, however, the effective diffusion coefficient increases only logarithmically with  $q$ .

Biological membranes typically contain various components such as lipid mixtures, sterols, and proteins that are indispensable to cell functions [1]. Rather than being uniformly distributed in the membrane, there are growing evidences that some intra-membrane components are incorporated in domains arising from lateral lipid segregation in membranes. This phenomenon has attracted great interest in the context of ‘rafts’ [2], i.e., liquid domains rich in cholesterol, saturated lipids (typically sphingomyelin lipids), and in some cases particular proteins [3]. Cholesterol-rich domains have been directly observed in model membranes composed of lipid mixtures and cholesterol, using advanced fluorescence microscopy [4–6].

Recent researches have focused on the dynamical aspect of lateral phase separation in mixed membranes. For example, Veatch and Keller observed domain formation in giant vesicles consisting of dipalmitoyl-phosphatidylcholine (DPPC), dioleoyl-phosphatidylcholine (DOPC), and cholesterol [7]. Depending on the composition, phase separation occurs through either domain ripening or spinodal decomposition. Saeki *et al* reported that the average domain size develops with time as  $\sim t^{0.15}$ , which is a very slow process [8]. In the experiment by Yanagisawa *et al*, two different types of domain coarsening were observed, i.e., ‘normal coarsening’ and the ‘trapped coarsening’ [9]. In the former case, the growth law is found to be  $\sim t^{2/3}$ , whereas in the latter case, domain coarsening is suppressed at a certain size. Moreover,

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domain growth and budding in phase-separating fluid vesicles were investigated by means of several simulation methods such as the time-dependent Ginzburg–Landau approach [10], Monte Carlo simulation [11], or dissipative particle dynamics [12].

In this letter, we investigate the dynamics of two-component fluid membranes from the theoretical point of view. Taking into account the effect of hydrodynamic interaction within the membrane, we calculate the decay rate of the time-correlation function of the concentration fluctuations. Since fluid membranes are sandwiched by surrounding water, we use a two-dimensional hydrodynamic equation with momentum decay to this water. Such an equation has previously been used to calculate the diffusion constant of proteins [13–16] or polymers [17] moving in membranes. Our formulation closely follows that in [18], which deals with the dynamics of microemulsions. An effective diffusion coefficient is calculated in the entire range of wavenumber. In the large-wavenumber limit, the effective diffusion coefficient depends logarithmically on the wavenumber, which is in contrast to the case for three-dimensional critical fluids. Although there have been no experimental reports on concentration fluctuations in membranes, they could be measured in the near future since the critical point has been determined in various phase diagrams of multi-component membranes [4, 6, 7].

We consider a two-component fluid membrane composed of lipid A and lipid B (or cholesterol) whose local area fractions are denoted by  $\phi_A(\mathbf{r})$  and  $\phi_B(\mathbf{r})$ , respectively ( $\mathbf{r}$  is the two-dimensional vector). Since the relation  $\phi_A(\mathbf{r}) + \phi_B(\mathbf{r}) = 1$  holds, we introduce a variable defined by  $\psi(\mathbf{r}) \equiv \phi_A(\mathbf{r}) - \phi_B(\mathbf{r})$ . The simplest form of the free-energy functional  $F\{\psi\}$  describing the fluctuation around the homogeneous state is

$$F\{\psi\} = \int d\mathbf{r} \left[ \frac{a}{2} \psi^2 + \frac{c}{2} (\nabla \psi)^2 - \mu \psi \right], \quad (1)$$

where  $a > 0$  is proportional to the temperature difference with respect to the critical temperature,  $c > 0$  is related to the line tension, and  $\mu$  is the chemical potential.

Next we discuss the hydrodynamics of the fluid membrane. Introducing the two-dimensional local velocities  $\mathbf{v}_A$  and  $\mathbf{v}_B$  for each component, we define the average local velocity by  $\mathbf{v} = \phi_A \mathbf{v}_A + \phi_B \mathbf{v}_B$ , which satisfies the incompressibility condition:

$$\nabla \cdot \mathbf{v} = 0. \quad (2)$$

Although a lipid membrane can be regarded as a two-dimensional viscous fluid, it is not an isolated system since lipids are coupled to the adjacent water. Accordingly, the momentum within the membrane can leak to the outer fluid. Such an effect can be phenomenologically taken into account through a momentum relaxation term in the two-dimensional hydrodynamic equation [13–17]. Moreover, a thermodynamic force due to the concentration fluctuations should also be included [19]. Then the hydrodynamic equation for a mixed fluid membrane can be written as

$$\rho \left[ \frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} \right] = \eta \nabla^2 \mathbf{v} - \nabla p - \lambda \mathbf{v} - \psi \nabla \frac{\delta F}{\delta \psi}, \quad (3)$$

where all the vector operators are two dimensional. In the above,  $p$  is the pressure, and the constants  $\rho$  and  $\eta$  are the density and dynamic viscosity of the lipid membrane, respectively<sup>5</sup>. The phenomenological parameter  $\lambda$  is inversely proportional to the momentum relaxation time which reflects the coupling strength between the membrane and the surrounding water. Within the present treatment, thermal fluctuations of the membrane shape are ignored. Details of the physical meaning of  $\lambda$  will be discussed later.

<sup>5</sup> The dimension of  $\eta$  is that of three-dimensional viscosity times length such as the membrane thickness.

The time evolution of concentration in the presence of hydrodynamic flow is given by the time-dependent Ginzburg–Landau equation for a conserved order parameter [18]:

$$\frac{\partial \psi}{\partial t} + \nabla \cdot (\mathbf{v}\psi) = L \nabla^2 \frac{\delta F}{\delta \psi}, \quad (4)$$

where  $L$  is the kinetic coefficient. Equations (1)–(4) provide the complete set of differential equations to be solved.

In the following, we assume that the relaxation of velocity  $\mathbf{v}$  is much faster than that of concentration  $\psi$ , so the left-hand side of equation (3) can be neglected. In this case, the velocity can be formally solved from equations (2) and (3) as

$$v^\alpha(\mathbf{r}, t) = \int d\mathbf{r}' T^{\alpha\beta}(\mathbf{r}, \mathbf{r}') (\nabla'_\beta \psi) \frac{\delta F}{\delta \psi(\mathbf{r}')}, \quad (5)$$

where  $T^{\alpha\beta}(\mathbf{r}, \mathbf{r}')$  is the two-dimensional Oseen tensor which takes into account the extra dissipation due to the surrounding fluid. Defining the Fourier transform by

$$T^{\alpha\beta}(\mathbf{r}) = \int \frac{d\mathbf{q}}{(2\pi)^2} T_{\mathbf{q}}^{\alpha\beta} e^{i\mathbf{q}\cdot\mathbf{r}}, \quad (6)$$

we obtain

$$T_{\mathbf{q}}^{\alpha\beta} = \frac{1}{\eta(q^2 + \kappa^2)} \left( \delta_{\alpha\beta} - \frac{q^\alpha q^\beta}{q^2} \right), \quad (7)$$

where  $q = |\mathbf{q}|$  and  $\kappa^{-1} \equiv (\eta/\lambda)^{1/2}$  is the hydrodynamic screening length. Equation (7) reduces to the usual Oseen tensor when  $\kappa \rightarrow 0$ . Since we are interested in the concentration fluctuations around the homogeneous state, we define  $\delta\psi(\mathbf{r}, t) = \psi(\mathbf{r}, t) - \bar{\psi}$ , where the bar indicates a spatial average. Then the free-energy functional (1) is expanded in powers of  $\delta\psi$  as

$$F\{\delta\psi\} = \int d\mathbf{r} \left[ \frac{a}{2} (\delta\psi)^2 + \frac{c}{2} (\nabla \delta\psi)^2 \right]. \quad (8)$$

Then the static correlation function is given by

$$\chi_{\mathbf{q}} = \langle \delta\psi_{\mathbf{q}} \delta\psi_{-\mathbf{q}} \rangle = \frac{k_B T}{c(q^2 + \xi^{-2})}, \quad (9)$$

where  $k_B$  is the Boltzmann constant,  $T$  the temperature, and  $\xi \equiv (c/a)^{1/2}$  the correlation length. Substituting equation (5) into (4), we obtain

$$\frac{\partial \delta\psi(\mathbf{r}, t)}{\partial t} = L \nabla^2 \frac{\delta F}{\delta (\delta\psi)} - \int d\mathbf{r}' (\nabla_\alpha \delta\psi(\mathbf{r})) T^{\alpha\beta}(\mathbf{r}, \mathbf{r}') (\nabla'_\beta \delta\psi(\mathbf{r}')) \frac{\delta F}{\delta (\delta\psi(\mathbf{r}'))}, \quad (10)$$

for the dynamics of the concentration fluctuations.

Using the above equation, we now consider the dynamics of the time-correlation function defined by

$$S(\mathbf{r}, t) = \langle \delta\psi(\mathbf{r}_1, t) \delta\psi(\mathbf{r}_2, 0) \rangle, \quad (11)$$

where  $\mathbf{r} = \mathbf{r}_2 - \mathbf{r}_1$ . With the use of the factorization approximation<sup>6</sup>, the spatial Fourier transform of  $S(\mathbf{r}, t)$  satisfies the equation

$$\frac{\partial S_{\mathbf{q}}(t)}{\partial t} = -(\Gamma_{\mathbf{q}}^{(1)} + \Gamma_{\mathbf{q}}^{(2)}) S_{\mathbf{q}}(t). \quad (12)$$

The first part of the relaxation rate  $\Gamma_{\mathbf{q}}^{(1)}$  corresponds to the so-called van Hove part, which is simply given by

$$\Gamma_{\mathbf{q}}^{(1)} = L k_B T q^2 \chi_{\mathbf{q}}^{-1} = L c q^2 (q^2 + \xi^{-2}). \quad (13)$$

<sup>6</sup> More precisely, the fourth-order correlations are approximated as  $\langle (\nabla_\alpha \delta\psi(\mathbf{r}_1, t)) (\nabla_\beta \delta\psi(\mathbf{r}_2, t)) \delta\psi(\mathbf{r}_3, t) \delta\psi(\mathbf{r}_4, 0) \rangle \approx \langle (\nabla_\alpha \delta\psi(\mathbf{r}_1, t)) (\nabla_\beta \delta\psi(\mathbf{r}_2, t)) \rangle \langle \delta\psi(\mathbf{r}_3, t) \delta\psi(\mathbf{r}_4, 0) \rangle$ . See [18] for details.

The second part of the relaxation rate,  $\Gamma_{\mathbf{q}}^{(2)}$ , is the hydrodynamic part that is given by an integral

$$\Gamma_{\mathbf{q}}^{(2)} = \frac{k_B T}{\eta \chi_{\mathbf{q}}} \int \frac{d\mathbf{p}}{(2\pi)^2} \chi_{\mathbf{p}} \frac{1}{(\mathbf{p} - \mathbf{q})^2 + \kappa^2} \frac{q^2 p^2 - (\mathbf{q} \cdot \mathbf{p})^2}{(\mathbf{p} - \mathbf{q})^2}. \quad (14)$$

We first carry out the integral over the angle between  $\mathbf{p}$  and  $\mathbf{q}$ :

$$\int_0^{2\pi} d\theta \frac{p^2 q^2 \sin^2 \theta}{(p^2 + q^2 - 2pq \cos \theta)(p^2 + q^2 - 2pq \cos \theta + \kappa^2)} = \frac{\pi}{2} \left( -1 + \sqrt{[(p+q)^2/\kappa^2 + 1][(p-q)^2/\kappa^2 + 1]} - |q^2 - p^2|/\kappa^2 \right). \quad (15)$$

Introducing the dimensionless quantities  $Q \equiv q\xi$  and  $\Lambda \equiv \kappa\xi$ , we further perform the integral over the magnitude of  $\mathbf{p}$  to obtain the result

$$\Gamma_{\mathbf{q}}^{(2)} = q^2 D_Q(\Lambda), \quad (16)$$

with an *effective diffusion coefficient*

$$D_Q(\Lambda) = \frac{k_B T}{8\pi\eta} \frac{Q^2 + 1}{Q^2} \left[ \frac{Q^2 + 1}{\Lambda^2} \ln \left( \frac{\Lambda}{Q^2 + 1} \right) - \ln \Lambda + \frac{\Omega}{2\Lambda^2} \ln \left( \frac{Q_+^4 + Q_-^2 + Q_+^2 \Omega}{\Omega - Q_-^2 - 1} \right) \right]. \quad (17)$$

Here we have used the following abbreviations:

$$\Omega = \sqrt{(Q^2 + \Lambda^2 - 1)^2 + 4Q^2}, \quad Q_{\pm} = \sqrt{Q^2 \pm \Lambda^2}. \quad (18)$$

For three-dimensional binary critical fluids, an analogous calculation yields the famous Kawasaki function [20]. Its validity has been successfully confirmed by several dynamic light-scattering experiments [21]. Equation (17) is the two-dimensional analogue of the Kawasaki function, provided that the momentum can leak to the surrounding fluid.

In figure 1, we plot  $D_Q(\Lambda)$  (scaled by  $k_B T/4\pi\eta$ ) as a function of  $Q = q\xi$  for various values of  $\Lambda$ . Since  $\Gamma_{\mathbf{q}}^{(2)}$  can be generally expanded in terms of  $Q^2$  in the isotropic state,  $D_Q(\Lambda)$  is constant in the limit of  $Q \ll 1$ . The effective diffusion coefficient starts to increase when  $Q \approx 1$ . All the curves are generally S-shaped, and an inflection point appears in the intermediate  $Q$ -region. In the limit of  $Q \gg 1$ , the asymptotic form of equation (17) can be approximately given by

$$D_Q(\Lambda) \approx \frac{k_B T}{8\pi\eta} \left[ \ln \left( \frac{Q^2 + \Lambda^2}{\Lambda^2} \right) + \frac{Q^2}{\Lambda^2} \ln \left( \frac{Q^2 + \Lambda^2}{Q^2} \right) \right]. \quad (19)$$

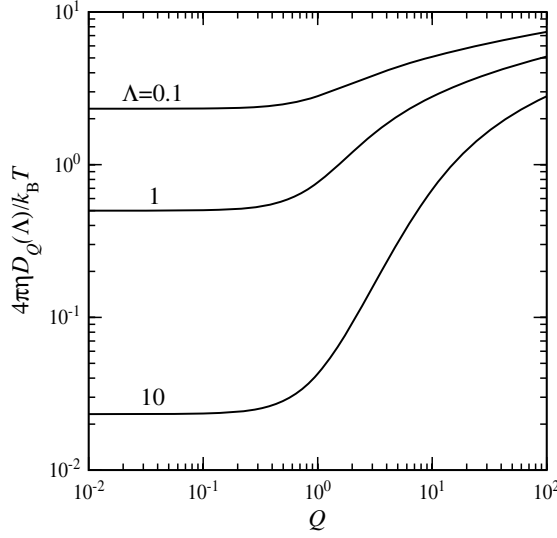
If we plot this equation in figure 1 for different  $\Lambda$  values (not shown), they all coincide with the exact result as long as  $Q > 1$ . Equation (19) can be further approximated as

$$D_Q(\Lambda) \approx \frac{k_B T}{4\pi\eta} \ln(Q/\Lambda), \quad (20)$$

when  $Q \gg \Lambda$ , or

$$D_Q(\Lambda) \approx \frac{k_B T}{4\pi\eta} (Q/\Lambda)^2, \quad (21)$$

when  $Q \ll \Lambda$  (except the logarithmic correction). Equation (20) shows that  $D_Q(\Lambda)$  depends only logarithmically on  $Q$  in the large- $Q$  limit, i.e.,  $Q \gg 1$  and  $Q \gg \Lambda$ . This is an important result of this letter. Such a weak wavenumber dependence should be contrasted with that in three-dimensional critical fluids for which the effective diffusion coefficient increases linearly with  $q$  (or the relaxation rate is proportional to  $q^3$ ) in the large- $q$  limit [20].



**Figure 1.** Scaled effective diffusion coefficient  $D_Q(\Lambda)$  as a function of the dimensionless wavenumber  $Q = q\xi$  for  $\Lambda = 0.1, 1, 10$  from top to bottom.

Next we argue the dependence of  $D_Q(\Lambda)$  on  $\Lambda$ , which measures the coupling strength between the membrane and the water. In figure 2, we plot  $D_Q(\Lambda)$  against  $\Lambda = \kappa\xi$  for various values of  $Q$ . When  $\Lambda$  is large, the momentum transfer from the membrane to the water becomes faster. Under such conditions, the concentration fluctuations decay slowly because the velocity field in the membrane is damped out quickly by the momentum transfer. Therefore, the effective diffusion coefficient  $D_Q(\Lambda)$  should be smaller for larger coupling strength, and  $D_Q(\Lambda)$  is a monotonically decreasing function of  $\Lambda$ . However, its  $\Lambda$ -dependence has two asymptotic regimes, as obtained by equations (20) and (21):  $D_Q(\Lambda)$  decreases only logarithmically with  $\Lambda$  when  $Q \gg \Lambda$ , while it decreases algebraically ( $\sim \Lambda^{-2}$ ) when  $Q \ll \Lambda$ . According to figure 2, the logarithmically dependent regime increases for larger  $Q$  (compare  $Q = 10$  and 100). This is because the length scale of our investigation ( $q^{-1}$ ) is much smaller than the screening length  $\kappa^{-1}$ , and  $D_Q(\Lambda)$  is hardly affected by the coupling effect.

From equation (17) the limiting value of  $D_Q(\Lambda)$  for  $Q \rightarrow 0$  becomes

$$D_0(\Lambda) = \frac{k_B T}{4\pi\eta} \frac{\ln \Lambda}{\Lambda^2 - 1}, \quad (22)$$

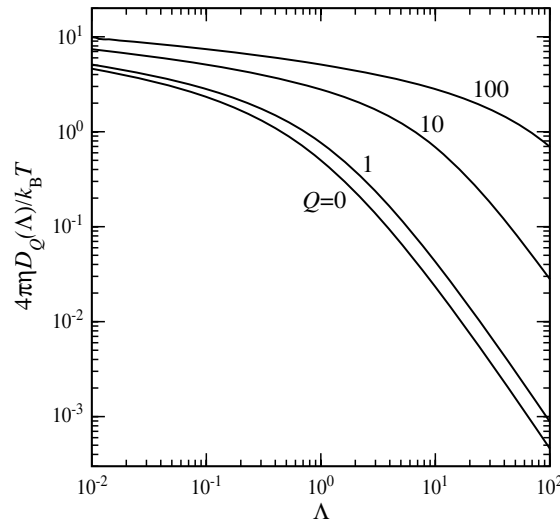
which is called the *cooperative diffusion constant*. This equation is plotted in figure 2 for  $Q = 0$ . We note that equation (22) is not singular at  $\Lambda \rightarrow 1$  and gives a finite value  $D_0(\Lambda) = k_B T / (8\pi\eta)$ . Two further limits of equation (22) are

$$D_0(\Lambda) \approx \frac{k_B T}{4\pi\eta} \ln(1/\Lambda), \quad (23)$$

for  $\Lambda \ll 1$ , and

$$D_0(\Lambda) \approx \frac{k_B T}{4\pi\eta} (1/\Lambda)^2, \quad (24)$$

for  $\Lambda \gg 1$ . These logarithmic and algebraic dependences on  $\Lambda$  are analogous to those given in equations (20) and (21), respectively. It should be noticed that  $\Lambda$  is proportional to the correlation length. The cooperative diffusion constant  $D_0(\Lambda)$  is smaller for larger correlation



**Figure 2.** Scaled effective diffusion coefficient  $D_Q(\Lambda)$  as a function of  $\Lambda = \kappa\xi$  for  $Q = 0, 1, 10$  and  $100$  from bottom to top. The line  $Q = 0$  corresponds to the scaled cooperative diffusion constant given by equation (22).

length  $\xi$ , as it should be. However, its dependence on the correlation length is different from that of the three-dimensional cooperative diffusion constant, which is inversely proportional to the correlation length  $\xi$  [20]. Moreover, equation (24) shows that the membrane viscosity  $\eta$  in the denominator of  $D_0(\Lambda)$  drops out since  $(1/\Lambda)^2 = \eta/\lambda\xi^2$ . The fact that  $D_0(\Lambda)$  is sensitive to the coupling to the environment (represented by  $\lambda$ ) has been pointed out recently for lipid mixtures close to a critical point [22].

It is worthwhile comparing the obtained cooperative diffusion constant with the tracer diffusion constant of a disc-like molecule with radius  $R$  moving in a two-dimensional fluid with momentum decay. Based on the same hydrodynamic equation as in equation (3) (without the thermodynamic force), the tracer diffusion constant was previously obtained as [13, 16]

$$D = \frac{k_B T}{4\pi\eta} \left( \frac{z^2}{4} + \frac{zK_1(z)}{K_0(z)} \right)^{-1}, \quad (25)$$

with  $z = \kappa R$ . In the above,  $K_0(z)$  and  $K_1(z)$  are the modified Bessel functions of the second kind of order zero and one, respectively. In the weak coupling limit,  $\kappa R \ll 1$ , equation (25) becomes

$$D \approx \frac{k_B T}{4\pi\eta} \left( \ln \frac{2}{\kappa R} - \gamma \right), \quad (26)$$

where  $\gamma = 0.5772 \dots$  is Euler's constant. Such a logarithmic dependence is consistent with the result of Saffman and Delbrück [23, 24]. In the strong coupling limit,  $\kappa R \gg 1$ , we have

$$D \approx \frac{k_B T}{4\pi\eta} \left( \frac{2}{\kappa R} \right)^2. \quad (27)$$

Comparing these equations with equations (23) and (24), we can easily confirm that the dependences on the correlation length  $\xi$  and the radius  $R$  are the same. Notice that  $\eta$  in equation (27) drops out, as in equation (24). Indeed the validity of equation (25) was recently confirmed by the measurement of diffusion constant of phase-separated domains in mixed membranes [25].

Finally, we discuss the physical meaning of the parameter  $\lambda$  in equation (3). When the lipid membrane is placed on a solid substrate, the term  $-\lambda\mathbf{v}$  represents the friction, which is proportional to the local membrane velocity relative to the substrate. Under the presence of a thin liquid layer of thickness  $\ell$  between the membrane and the substrate, the laminar flow gives rise to the friction  $\eta'\mathbf{v}/\ell$ , where  $\eta'$  is the fluid viscosity. This means that  $\lambda = \eta'/h\ell$  for a membrane with thickness  $h$  [13].

When  $\ell$  is infinitely large, the flow of the fluid surrounding the membrane deviates from laminar flow. In this case, Saffman and Delbrück obtained the diffusion constant in the weak coupling limit by taking into account the surrounding flow [23, 24]. By comparing their result and equation (26), we find that  $\lambda = \eta(2\eta'/h\eta)^2$  [17]. In the strong coupling limit, on the other hand, a small deviation was found between equation (25) and the extended result of the Saffman and Delbrück theory [13, 26]. In the strong coupling limit of Saffman and Delbrück theory, the drag force comes not only from the cylinder walls of molecules but also from the parts directly interacting with surrounding liquids [26]. Although the membrane-bound molecules are assumed to be flush with the membrane interface, the results depend on whether the molecules protrude into surrounding liquids or not in the limit [26]. Since equation (25) correlates with the extended results of Saffman and Delbrück theory over a wide range of  $\lambda$  values, our phenomenological approach is justified at least for the molecules that are flush with the membrane interface. It should also be pointed out that the size of diffusing molecules should be larger than that of the lipids composing the membrane in the Saffman and Delbrück theory. Therefore, the applicability of equation (3) to a mixed membrane is rationalized for the laminar flow of surrounding liquids but not justified from the model of Saffman and Delbrück theory.

In summary, we have calculated the decay rate of concentration fluctuations in a binary fluid membrane which is coupled to the adjacent water. In contrast to the case of three-dimensional critical fluids, the effective diffusion coefficient increases logarithmically with wavenumber. Our prediction should be checked experimentally.

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## References

- [1] Lipowsky R and Sackmann E (ed) 1995 *Structure and Dynamics of Membranes* (Amsterdam: Elsevier)
- [2] Simons K and Ikonen E 1997 *Nature* **387** 569
- [3] Brown D A and London E 2000 *J. Biol. Chem.* **275** 17221
- [4] Veatch S L and Keller S L 2002 *Phys. Rev. Lett.* **89** 268101
- [5] Baumgart T, Hess S T and Webb W W 2003 *Nature* **425** 821
- [6] Veatch S L and Keller S L 2005 *Phys. Rev. Lett.* **94** 148101
- [7] Veatch S L and Keller S L 2003 *Biophys. J.* **85** 3074
- [8] Saeki D, Hamada T and Yoshikawa K 2006 *J. Phys. Soc. Japan* **75** 013602
- [9] Yanagisawa M, Imai M, Masui T, Komura S and Ohta T 2007 *Biophys. J.* **92** 115
- [10] Taniguchi T 1996 *Phys. Rev. Lett.* **76** 4444
- [11] Kumar P B S, Gompfer G and Lipowsky R 2001 *Phys. Rev. Lett.* **86** 3911
- [12] Laradji M and Kumar P B S 2004 *Phys. Rev. Lett.* **93** 198105  
Laradji M and Kumar P B S 2004 *J. Chem. Phys.* **123** 224902
- [13] Evans E and Sackmann E 1988 *J. Fluid Mech.* **194** 553
- [14] Merkel R, Sackmann E and Evans E 1989 *J. Physique* **50** 1535
- [15] Suzuki Y Y and Izuyama T 1989 *J. Phys. Soc. Japan* **58** 1104
- [16] Seki K and Komura S 1993 *Phys. Rev. E* **47** 2377
- [17] Komura S and Seki K 1995 *J. Physique II* **5** 5
- [18] Nonomura M and Ohta T 1999 *J. Chem. Phys.* **110** 7516

- [19] Chaikin P M and Lubensky T C 1995 *Principles of Condensed Matter Physics* (New York: Cambridge University Press)
- [20] Kawasaki K 1970 *Ann. Phys.* **61** 1
- [21] Swinney H and Henry D L 1973 *Phys. Rev. A* **8** 2586
- [22] Tserkovnyak Y and Nelson D R 2006 *Proc. Natl Acad. Sci. USA* **103** 15002
- [23] Saffman P G and Delbrück M 1975 *Proc. Natl Acad. Sci.* **72** 3111
- [24] Saffman P G 1976 *J. Fluid Mech.* **73** 593
- [25] Kaizuka Y and Groves J T 2004 *Biophys. J.* **86** 905
- [26] Hughes B D, Pailthorpe B A and White L R 1981 *J. Fluid Mech.* **110** 349