PHYSICAL REVIEW E **80**, 020902(R) (2009)

# Critical dynamics in multicomponent lipid membranes

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(Received 30 April 2009; revised manuscript received 24 July 2009; published 18 August 2009)

The formation and dynamics of spatially extended compositional domains in multicomponent lipid membranes both *in vivo* and *in vitro* lie at the heart of many important biological and biophysical phenomena. While the thermodynamic basis for domain formation has been explored extensively in the past, the roles of membrane and exterior fluid hydrodynamics on domain formation kinetics have received less attention. A case in point is the impact of hydrodynamics on the dynamics of compositional heterogeneities in lipid membranes in the vicinity of a critical point. In this Rapid Communication it is argued that the asymptotic dynamic behavior of a lipid membrane system in the vicinity of a critical point is strongly influenced by hydrodynamic interactions. More specifically, a mode-coupling argument is developed which predicts a scaling behavior of lipid transport coefficients near the critical point for both symmetric and asymmetric bilayers immersed in a bulk fluid.

DOI: 10.1103/PhysRevE.80.020902 PACS number(s): 87.16.D−, 64.60.Ht, 87.14.Cc, 87.15.Zg

The presence of compositional heterogeneities and their dynamics in lipid membranes in intact cells ("lipid rafts") [1–3] present intriguing questions from both the physical and biological perspectives. Fundamentally, one would like to understand both how these domains relate to different biological functions and which physical mechanisms living cells can employ to control the membrane "microstructure" *in vivo*, e.g., the spatiotemporal evolution of local lipid compositions and the presence (or absence) of specific membrane proteins. In the latter endeavor, experimental [4–8] and theoretical [9–15] studies of *in vitro* membranes have proven very useful as they have provided a beautiful physical picture of the phase behavior of multicomponent lipid mixtures in the absence of cellular phenomena (such as active lipid transport to and from the membrane). In contrast to equilibrium behavior in synthetic membranes, however, the dynamics of compositional lipid domains is currently not as well understood even though it is by now well established that the raft domains *in vivo* are highly dynamic entities [3].

One often overlooked complication in developing physically based models for membrane domain kinetics resides in the treatment of the surrounding fluid. In particular, fluctuations in the compositional domains will induce flow fields in the surrounding fluid, which will subsequently back react with the membrane. One direct consequence of this back reaction is the nontrivial dependence of the diffusion coefficient $D$ of a membrane inclusion or compositional domain on the domain/inclusion size $r$: both theory [16,17] and experiments [6] have demonstrated that $D \sim \ln(1/r)$ for small $r$ with a crossover to $D \sim 1/r$ at large $r$. Particle-based models for membranes, which appropriately account for hydrodynamic effects arising from the surrounding fluid and provide a detailed microscopic view of collective phenomena associated with lateral lipid diffusion [13,15,18], have difficulties in probing large-scale compositional domain dynamics due to computational restrictions. While rather simple spatially extended continuum models have been proposed recently which address domain formation processes in synthetic bilayers across mesoscopic and macroscopic scales [10,11] and elucidate the potential role of active cellular processes on raftlike domain formation dynamics [19], these models do not explicitly incorporate the role of hydrodynamics on compositional domain formation and dynamics, with the notable exception of Ref. [20] wherein hydrodynamics within the membrane was included while the coupling between the membrane the exterior fluid was neglected. In order to develop a better understanding of compositional raft domain dynamics in lipid membranes, both *in vivo* and *in vitro*, it is thus necessary to quantitatively assess when hydrodynamics may or may not be neglected or when they perhaps completely dominate the dynamics.

A particularly interesting and experimentally relevant regime is that close to a second order phase transition at $T = T_c$, where an initially homogeneous lipid system becomes compositionally patterned. While the static critical behavior of the membrane system is expected to belong to the two-dimensional (2D) Ising model universality class (UC) [7,8,21], the critical dynamics has been explored experimentally only very recently [7]. Specifically, Ref. [7] studied the scaling behavior of the concentration diffusion coefficient $D_c$ and reported a result that was consistent with theoretical models which only include simple diffusive transport of the lipids along the membrane, akin to the model in Ref. [19] in the absence of cellular lipid trafficking. Theoretically, on the other hand, the coupling of lipids to both the membrane and exterior fluid flow fields is expected to influence the dynamic critical behavior and in particular affect $D_c$ [21]—indeed, a recent theoretical analysis by Tserkovnyak and Nelson demonstrated that the diffusivity of a membrane protein is strongly affected by critical fluctuations in a lipid membrane suspended over a solid substrate [22]. In this Rapid Communication it is argued that the asymptotic behavior of the concentration diffusion coefficient $D_c$ in lipid bilayers approaching a critical point is strongly influenced by hydrodynamic interactions above a characteristic hydrodynamic length scale $l_H$, which depends on the viscosities of the membrane and the exterior fluid, respectively. Specifically, it is argued that asymptotically close to the critical point and in lipid membranes immersed in a bulk fluid $D_c^{-1} \sim \xi$, where $\xi$ denotes the static correlation length.

The analysis may be developed as follows. The membrane, which occupies the region $-h \le z \le 0$, $-\infty < x < \infty$, and $-\infty < y < \infty$, is assumed to be surrounded by a simple Newtonian fluid with viscosity $\eta$. Furthermore, the velocity field $\mathbf{u}(\mathbf{r},t)$ of the exterior fluid satisfies [17]

$$\eta \nabla^2 \mathbf{u} - \nabla p = 0 \quad \text{and} \quad \nabla \cdot \mathbf{u} = 0. \tag{1}$$

Along the membrane-fluid interfaces we assume a no-slip boundary condition, which implies that $\mathbf{u}(x,y,0)=\mathbf{u}(x,y,-h)=\mathbf{u}_M(x,y)$, where $\mathbf{u}_M$ denotes the local velocity field within the membrane. Furthermore, the exterior fluid applies shear traction forces along the two membrane-fluid interfaces of magnitudes $\mathbf{F}_1$ and $\mathbf{F}_2$, respectively, which appear as body forces within the membrane.

For simplicity, we assume that the membrane comprises of two lipid types, A and B, with local concentrations $c_A$ and $c_B$, respectively. Now, within the membrane, the dimensionless concentration field $\psi \equiv [c_A(\mathbf{x},t) - c_B(\mathbf{x},t)]/\bar{c}$, where $\bar{c}$ denotes the critical composition, evolves according to

$$\frac{\partial \psi}{\partial t} + \nabla \cdot (\psi \mathbf{u}_M) = \lambda_0 \nabla^2 \mu_\psi + \epsilon, \tag{2}$$

where $\mu_\psi \equiv \delta F/\delta \psi$ denotes the chemical potential with the free energy given by [21]

$$F = \int dxdy \left[ \frac{1}{2}(\nabla \psi)^2 + \frac{1}{2} r_0 \psi^2 + u_0 \psi^4 + \Phi(x,y,t)\psi \right], \tag{3}$$

where $\mathbf{u}_M$ denotes the local velocity field within the membrane, $\Phi = -\mathbf{F} \cdot \mathbf{x}$ denotes an external field, and the stochastic noise term satisfies $\langle \epsilon \rangle = 0$ and $\langle \epsilon(\mathbf{x},t)\epsilon(\mathbf{x}',t')\rangle = -2\lambda_0 \nabla^2 \delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$. Here, we have assumed that the bilayer is symmetric with respect to lipid compositions and that the compositions are coupled in the two leaflets such that they become spatially synchronized as has been observed in experiments [5]; asymmetric bilayers will be discussed briefly at the end of this Rapid Communication.

Now, the membrane flow field $\mathbf{u}_M(\mathbf{x},t)$ satisfies [17,23]

$$\frac{\partial \mathbf{u}_M}{\partial t} = \eta_M \nabla^2 \mathbf{u}_M - \nabla p_M - \psi \nabla \mu_\psi - (\mathbf{F}_1 + \mathbf{F}_2)/h + \mathbf{f}(\mathbf{x},t) \tag{4}$$

and

$$\nabla \cdot \mathbf{u}_M = 0, \tag{5}$$

where $\eta_M$ denotes the membrane viscosity and where the fluctuating term $\mathbf{f}$ satisfies $\langle f_\alpha \rangle = 0$ and $\langle f_\alpha(\mathbf{x},t) f_\beta(\mathbf{x}',t')\rangle = -2\eta_M \nabla^2 \delta(\mathbf{x}-\mathbf{x}')\delta(t-t')\delta_{\alpha,\beta}$. The effective membrane pressure $p_M$ is employed to enforce the incompressibility condition, while the term $\psi \nabla \mu_\psi$ accounts for the impact of compositional variations and external field $\mathbf{F}$ in the membrane stress tensor. Note that we have assumed that the velocity fields of the two leaflets are spatially synchronized [24].

Upon switching on the external field, a net concentration current density $\mathbf{j}^\psi$ is established such that $\mathbf{j}^\psi = \lambda \mathbf{F}$, which defines the transport coefficient $\lambda$ for the concentration difference. It is straightforward to show that the concentration diffusion constant $D_c = \lambda/\chi_\psi$, where the order-parameter susceptibility $\chi_\psi \equiv \partial \psi/\partial \mu_\psi \sim \xi^{7/4}$ for the 2D Ising model; here, $\xi$ denotes the correlation length with asymptotic scaling behavior $\xi \sim (T-T_c)^{-1}$. Furthermore, it is precisely $D_c$ which has been measured in Ref. [7]. Note that in the absence of advective transport, $\lambda = \lambda_0$ even as the critical point is approached [21], which implies that $D_c \sim \xi^{-7/4}$. The goal of the mode-coupling analysis below is to account for the effects of membrane and exterior fluid hydrodynamics on $\lambda$.

To this end, following Ref. [25], we introduce a dimensionless variable $\delta$ through $\delta \equiv 2\xi\eta/h\eta_M$. Here, $\delta$ denotes the correlation length normalized by a hydrodynamic length $l_H = h\eta_M/(2\eta)$, which plays an important role in the subsequent analysis. We note that upon employing typical values $h\eta_M \approx 10^{-10}$ Pa s m (see, e.g., Ref. [26]) and $\eta \approx 10^{-3}$ Pa s for water at room temperature, one obtains $l_H \simeq 50$ nm for a lipid bilayer. Now, consider a region of the membrane of area $\sim \xi^2$. The typical value of $\psi$ in this region $(\bar{\psi})$ can be obtained by combining the spatial integral of the two-point correlation function $\int d\mathbf{R} G(\mathbf{R}) = \int d\mathbf{R} \langle \psi(\mathbf{r},t)\psi(\mathbf{r}+\mathbf{R},t)\rangle \approx \bar{\psi}^2 \xi^2$ with the static susceptibility sum rule $\int d\mathbf{R} G(\mathbf{R}) = k_B T \chi_\psi$:

$$\bar{\psi}^2 \approx k_B T \chi_\psi / \xi^2. \tag{6}$$

In the presence of a field $\mathbf{F}$, the net applied force on the region is

$$\mathbf{f}_{app} = -\int_0^\xi dx \int_0^\xi dy \psi \nabla \mu_\psi \approx \xi^2 \bar{\psi} \mathbf{F}. \tag{7}$$

The domain accelerates until the viscous drag from the membrane fluid and the exterior fluid provides a counteracting force. The drag force can be obtained in closed form by neglecting thermal fluctuations in Eq. (4) and assuming that the domain translates as a rigid body [27], in which case it can be mathematically treated in exactly the same way as a cylindrical membrane inclusion [16,17,25] with the result

$$\mathbf{f}_{visc} = -\frac{4\pi\eta_M h}{\ln(2\delta^{-1}) - \gamma} \mathbf{u}_M \quad \text{when} \quad \delta \ll 1, \tag{8}$$

where $\gamma = 0.577215$ denotes the Euler constant and

$$\mathbf{f}_{visc} = -8\eta_M h \delta \mathbf{u}_M \quad \text{when} \quad \delta \gg 1. \tag{9}$$

A very useful analytical expression for the diffusion coefficient of a cylindrical membrane inclusion, which has been shown to provide an accurate approximation of the exact result from Ref. [17] for all $\delta$, has been provided by Petrov and Schwille [25], in terms of which the expression for $\mathbf{f}_{visc}$ becomes

$$\mathbf{f}_{visc} = -\frac{4\pi\eta_M h}{\Delta(\delta)} \mathbf{u}_M, \tag{10}$$

where

$$\Delta(\delta) = \frac{\ln(2\delta^{-1}) - \gamma + 4\delta/\pi - \frac{1}{2}\delta^2 \ln(2\delta^{-1})}{1 - \frac{1}{3}\delta^3 \ln(2\delta^{-1}) + c_1\delta^{b_1}/(1 + c_2\delta^{b_2})}. \tag{11}$$

Here, $c_1=0.73761$, $b_1=2.74819$, $c_2=0.52119$, and $b_2=0.61465$ [25].

Now, the region of fluid will accelerate until the viscous force balances the external one:

$$\mathbf{f}_{app} = -\mathbf{f}_{visc} \leftrightarrow \mathbf{f}_{app} = \frac{4\pi\eta_M h}{\Delta(\delta)}\mathbf{u}_M. \tag{12}$$

Finally, the net current density is given by

$$\mathbf{j}^{\psi} = \bar{\psi}\mathbf{u}_M = \frac{\Delta(\delta)}{4\pi\eta_M h}\bar{\psi}^2\xi^2\mathbf{F} \tag{13}$$

or

$$\mathbf{j}^{\psi} = \frac{\Delta(\delta)}{4\pi\eta_M h}k_B T\chi_{\psi}\mathbf{F} = \lambda\mathbf{F}. \tag{14}$$

Thus, the transport coefficient λ, renormalized by hydrodynamic interactions, in this simple mode-coupling approach is given by

$$\lambda = \lambda_0 + \frac{\Delta(\delta)}{4\pi\eta_M h}Bk_B T\chi_{\psi}, \tag{15}$$

where we have included a dimensionless constant $B \sim \mathcal{O}(1)$ which cannot be obtained from the present analysis. Finally, the expression for λ above implies that

$$D_c = \frac{\lambda}{\chi_{\psi}} = \frac{A\lambda_0}{\xi^{7/4}} + \frac{\Delta(\delta)}{4\pi\eta_M h}Bk_B T. \tag{16}$$

Let us now briefly discuss the implications of Eq. (16), which is the central result of this Rapid Communication. When $\delta=\xi/l_H \ll 1$, the correlation length is below the characteristic hydrodynamic length scale, and a logarithmic correction to $D_c$ emerges. On the other hand, in the asymptotic case $\xi/l_H \gg 1$, the logarithmic correction is replaced by a more slowly varying term in $D_c$, which dominates asymptotically,

$$D_c \propto \frac{k_B T}{\eta\xi} \quad \text{when} \quad \xi \to \infty, \tag{17}$$

such that $D_c^{-1}$ diverges exactly the same way as the correlation length $\xi$ as the critical point is approached. Furthermore, we note that although the static critical behavior belongs to the 2D Ising model UC, the dynamics appears to belong to the so-called three-dimensional (3D) "model H" UC in the classification scheme of Hohenberg and Halperin [21]. However, important differences exist. In particular, in the 3D model H case, a more rigorous renormalization group treatment of the model equations indicate that the bare viscosity $\eta$ also becomes renormalized to $\bar{\eta}$ such that $\bar{\eta} \sim \xi^{x_\eta}$ where $x_\eta \approx 0.155(0.065)$ in 2D (3D) [21], leading to a small correction to the above simple mode-coupling result: $D_c \sim \xi^{-1-x_\eta}$. In the membrane case, however, the exterior fluid remains noncritical, and thus $\eta$ is expected to remain finite at the membrane critical point. Hence, we conjecture that Eq. (17) is the exact asymptotic scaling law for the concentration diffusion coefficient $D_c$ in membranes immersed in a bulk fluid. This conclusion remains valid even if the renormalized membrane viscosity $\bar{\eta}_M$ displays a weak divergence, as long as $\bar{\eta}_M/\xi \to 0$ as $\xi \to \infty$ such that $\xi/\bar{l}_H = 2\eta\xi/(h\bar{\eta}_M) \gg 1$. Also note that if the bilayer is suspended above a rigid substrate at a distance $H$ away from the membrane, asymptotically the viscous drag force on the domain is given by $\mathbf{f}_{visc} \sim \eta\xi^2/H\mathbf{u}_M$ [28], which implies that $D_c \sim H\xi^{-2}$, in agreement with Ref. [22].

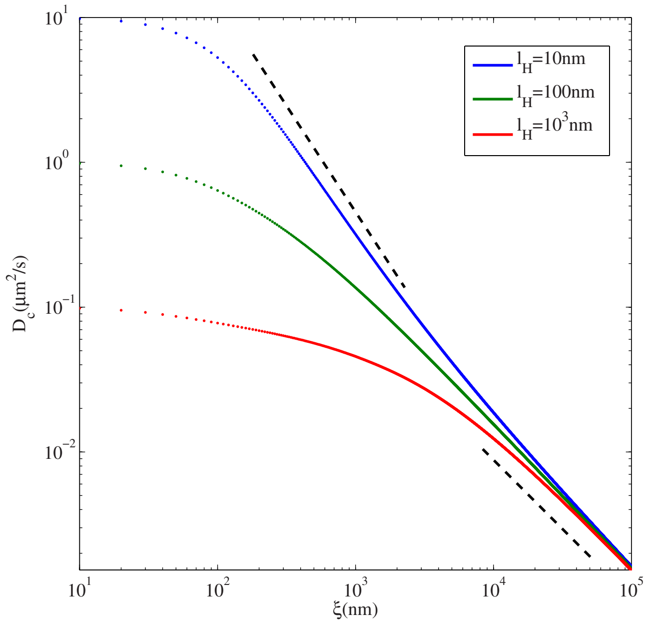

FIG. 1. (Color online) Concentration diffusion coefficient $D_c$ as a function of the static correlation length $\xi$ for three representative values of the hydrodynamic length $l_H$ from Eq. (16): $l_H=10$ nm (upper line), $l_H=100$ nm (middle line), and $l_H=10^3$ nm (bottom line). The two dashed lines on the top and bottom have slopes $-7/4$ and $-1$, respectively.

The generic behavior of $D_c$ as a function of $\xi$ is displayed in Fig. 1 for three representative values for the hydrodynamic length $l_H$: $l_H=10$ nm, $l_H=100$ nm, and $l_H=10^3$ nm, corresponding to a variation in $\eta_M$ over two orders of magnitude. Motivated by Ref. [7], the data were generated for each $l_H$ by evaluating $D_c^{-1}=D_0^{-1}+(A\lambda_0/\xi^{7/4}+\Delta D_0)^{-1}$, where $D_0 \approx k_B T/(4\pi\eta_M h)$ denotes the lipid self-diffusion coefficient. The parameter values $A\lambda_0=(5.5\times10^{-24}\ \mathrm{m}^{7/4}, 5.3\times10^{-25}\ \mathrm{m}^{7/4}, 5.3\times10^{-26}\ \mathrm{m}^{7/4})$ and $D_0=(10\ \mu\mathrm{m}^2/\mathrm{s}, 1\ \mu\mathrm{m}^2/\mathrm{s}, 0.1\ \mu\mathrm{m}^2/\mathrm{s})$ for $l_H=10$ nm, $l_H=100$ nm, and $l_H=10^3$ nm, respectively, were employed. The parameters were chosen such that $D_c \approx D_0$ when $\xi \simeq 10$ nm. There are three important features in the plot. First, a microscopic $l_H$ leads to a regime where $D_c \sim \xi^{-7/4}$ dominates before the asymptotic behavior sets in at large $\xi$. Second, as $l_H$ increases, the regime where $D_c \sim \xi^{-7/4}$ can be observed becomes more narrow and is eventually replaced with a less rapidly decreasing behavior, courtesy of the logarithmic term in Eq. (16), before the asymptotic behavior $D_c \sim \xi^{-1}$ again sets in. Third (and perhaps most importantly), the data suggest that the asymptotic scaling behavior should be most readily observed in systems with intermediate values of $l_H$, given the experimental challenges in exploring systems with correlation lengths $\xi \gtrsim 10$ $\mu$m. For example, the crossover to the asymptotic behavior occurs when $\xi \gtrsim 1$ $\mu$m for $l_H = 100$ nm and thus should be experimentally accessible, while for $l_H = 10$ nm and $10^3$ nm, the asymptotic behavior is found only when $\xi \gtrsim 10$ $\mu$m. These observations are consistent with the results in Ref. [7], wherein transient $D_c \sim \xi^{-7/4}$ behavior was reported for $\xi \lesssim 4$ $\mu$m. It is interesting to note that these results imply that membrane and exterior fluid hydrodynamics play a secondary role in the formation kinetics of submicrometer size compositional domains in both *in vivo* and *in vitro* membranes.

The extension of this approach to asymmetric bilayers is straightforward. For concreteness, consider the case where the two leaflets possess different critical temperatures such that $T_c^{\mathrm{I}} \gg T_c^{\mathrm{II}}$. As the temperature is lowered toward $T_c^{\mathrm{I}}$, leaflet "I" will undergo critical fluctuations and exhibits similar critical dynamics as in the symmetric case. In this case, leaflet "II" simply contributes to the viscous drag force on the compositional domains in leaflet I; it is also possible that the coupling between the lipid compositions in the two leaflets may also induce some spatial inhomogeneities in leaflet II. As the temperature is reduced further toward $T_c^{\mathrm{II}}$, leaflet I undergoes spinodal decomposition, while leaflet II remains in a mixed state with some spatial inhomogeneities in the lipid composition. Recently, it has been argued that in this case the characteristic length scale in leaflet I grows as $L(t) \sim t^{1/2}$ asymptotically [29,30]. Finally, we speculate that as $T \to T_c^{\mathrm{II}}$, the fluctuating composition field in leaflet II will display similar critical dynamics as in the symmetric case.

In conclusion, in this Rapid Communication we have argued that the coupling between compositional fluctuations near a critical point in lipid bilayers and membrane/exterior fluid hydrodynamics leads to scaling behavior in lipid transport coefficients. In particular, we predict that the composition diffusion coefficient obeys a universal scaling law $D_c \sim \xi^{-1}$ in immiscible lipid membranes immersed in a bulk fluid as the critical point is approached.

*Note added.* Similar results for the scaling behavior of the concentration diffusion coefficient have been obtained by Inaura and Fujitani [31] by using a different approach.

This work was in part supported by an NSF-DMR Grant No. DMR-0449184. Useful discussions with Jason Fleischer are gratefully acknowledged.

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