

## Topological instabilities and phase behavior of fluid membranes

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A phenomenological model of membrane phase behavior is presented in which the lamellar ( $L_\alpha$ ) phase is subject to two instabilities, one towards the formation of multiply connected, nearly minimal surfaces, and the other toward the formation of disconnected, nearly spherical surfaces. It is shown that upon dilution, fluctuations generally drive the system through one of these instabilities at a lamellar spacing  $d_c$  less than the persistence length  $\xi_\kappa$  beyond which they destroy its mean rigidity, resulting when  $d_c \ll \xi_\kappa$  in isotropic phases of quasirigid membranes with well-defined preferred topologies.

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Dilute solutions of surfactant can exhibit several phases in which the molecules aggregate as extended bilayer membranes [1]. These include a smectic-A lamellar ( $L_\alpha$ ) phase, an isotropic bicontinuous ( $L_3$ ) or "sponge" phase [2], and, less frequently [3,4], an ( $L_4$ ) phase of disconnected vesicles. An analogous set of phases can occur in microemulsions of oil, water, and surfactant, in which microdomains of oil and water are separated by monolayers of surfactant. It has been the goal of a great deal of theoretical work, including the present paper, to describe the phase behavior of these surfactant systems in terms of the elasticity and fluctuations of their constituent membranes.

The microscopic elasticity of a bilayer fluid membrane is described [5] by a harmonic bending energy

$$H = \int dA \left\{ \frac{1}{2} \kappa C^2 + \bar{\kappa} K \right\} \quad (1)$$

in which  $C = C_1 + C_2$  and  $K = C_1 C_2$  are the membrane's mean and Gaussian curvatures,  $C_1$  and  $C_2$  are the principal curvatures, and  $\kappa$  and  $\bar{\kappa}$  are the mean and Gaussian rigidities. The integral over Gaussian curvature is directly related, by the Gauss-Bonnet theorem  $\int dA K = 4\pi(1-g)$  [6], to the topological genus  $g$ , or number of "handles," of the surface in question. Consequently, the parameter  $\bar{\kappa}$  enters  $H$  only as a chemical potential for the formation of handles, while  $\kappa$  along determines the energies of topology-preserving deformations of the membrane.

The free energy cost of imposing upon a fluctuating membrane a deformation of length scale  $\xi$  can be described theoretically by a scale-dependent renormalized rigidity  $\kappa(\xi)$ . Several calculations of  $\kappa(\xi)$  [6,7] have shown that it is softened by fluctuations, and decreases as

$$\kappa(\xi) = \kappa(a) - \frac{\alpha T}{4\pi} \ln \left( \frac{\xi}{a} \right) \quad (2)$$

with increasing length scale, with a coefficient  $\alpha = 3$ , where  $a$  is a microscopic cutoff length. A similar renormalization has been found [6] for the renormalized Gaussian rigidity  $\bar{\kappa}(\xi)$ , which determines the free energy cost for the formation of handles of size  $\xi$ . The length scale  $\xi_\kappa \sim a e^{4\pi\kappa/\alpha T}$  at which  $\kappa(\xi)$  vanishes, known as the persistence length, is the

correlation length beyond which the orientations of distant points on a membrane of simple topology become uncorrelated.

The thermodynamic consequences of this softening of rigidity has been explored in a series of phenomenological "random surface" models of both the  $L_3$  [2,8] and bicontinuous microemulsion [9–11] phases. The  $L_3$  phase is described in such models as a random self-avoiding surface, which is characterized by a structural length  $\xi$  that determines the typical radii of curvature and distances between membranes. The magnitude of  $\xi$ , like the spacing  $d$  of the  $L_\alpha$  phase, is determined by the amount of surfactant available to form membranes. The softening effects of thermal fluctuations are included by the use of a  $\xi$ -dependent bending energy. For simplicity, these models have often used a single-bending-constant approximation, which uses a scale-dependent mean rigidity  $\kappa(\xi)$  but sets  $\bar{\kappa}(\xi)$  to zero. Within the assumptions of a random topology and of zero Gaussian rigidity, the  $L_3$  phase is found to have a lower free energy than the  $L_\alpha$  phase for concentrations such that  $\xi \geq \xi_\kappa$ , in agreement with the original heuristic argument of de Gennes and Taupin [9] that a random structure should become stable at concentrations for which its typical radii of curvature exceed the orientational correlation length  $\xi_\kappa$  of the membrane.

An alternative view of the  $L_3$  phase has been put forward by Porte and collaborators [13,14], who describe the  $L_\alpha$ - $L_3$  transition as a topological transition controlled primarily by the value of  $\bar{\kappa}$ . This description is based on several experimental observations:

(i) Comparison of measurements of  $\kappa$  and of phase behavior show that the  $L_\alpha$  phase can, in at least some systems [15], melt at a critical spacing  $d_c$  that is several orders of magnitude smaller than the calculated value of  $\xi_\kappa$ .

(ii) Neutron scattering [13,15–17], transport (electrical conductivity and self-diffusion) [13,16–18], and freeze fracture microscopy [19] results are all consistent with a description of the  $L_3$  phase as a single multiply connected sheet with almost uniformly negative Gaussian curvature.

(iii) The phase boundaries of the  $L_\alpha$  and  $L_3$  phases seem to show a sensitive and systematic dependence on  $\bar{\kappa}$ . In membranes that contain a mixture of surfactant and alcohol cosurfactant, increasing the volume fraction  $\chi$  of cosurfactant is believed to decrease  $-\bar{\kappa}$  (where  $\bar{\kappa} < 0$ ) for bilayers

separated by water but to increase  $-\bar{\kappa}$  for inverted bilayers separated by oil, while decreasing  $\kappa$  in either case. Both types of system typically exhibit a sequence of phases with increasing Gaussian curvature, passing from an  $L_3$  to an  $L_\alpha$  to either an  $L_4$  or micellar phase, as  $\chi$  is varied in the direction of increasing  $-\bar{\kappa}$  [4,13,16,17,20]. This sequence is also observed in single component membranes, where  $\bar{\kappa}$  can be controlled by varying temperature [21] or solvent salinity [15]. Where an  $L_\alpha$  and  $L_3$  phase coexist, the minimum structural length of the  $L_3$  phase typically decreases with decreasing  $-\bar{\kappa}$ , even, as when varying  $\chi$  in an inverted bilayer [17], when this is also the direction of increasing  $\kappa$ .

In what follows, I present an extension of the random surface models, in which one treats more carefully the scale dependence of  $\bar{\kappa}$  and the effect of both  $\kappa$  and  $\bar{\kappa}$  upon the system's topology. Many of the ideas presented here have been arrived at independently by Golubovic [12]. I begin by rewriting bending energy (1) in a symmetric form that emphasizes the limits of stability of a flat surface,

$$H = \frac{1}{2} \int dA \{ \kappa_+ C_+^2 + \kappa_- C_-^2 \}, \quad (3)$$

in which  $C_\pm \equiv C_1 \pm C_2$ ,  $\kappa_+ \equiv \kappa + (1/2)\bar{\kappa}$ , and  $\kappa_- \equiv -(1/2)\bar{\kappa}$ . A flat membrane is energetically stable only when both  $\kappa_+$  and  $\kappa_-$  are positive: When  $\kappa_+ < 0$ , the flat state is unstable towards the formation of many spherical surfaces, for which  $C_1 = C_2$ , with a bending energy of  $E = 8\pi\kappa_+$  per sphere. When  $\kappa_- < 0$  (or  $\bar{\kappa} > 0$ ), it is unstable towards the formation of an infinite minimal surface, defined by the property that  $C_1 = -C_2$  everywhere. Because these instabilities both involve discontinuous changes of topology, rather than continuous deformation, I refer to  $\kappa_\pm$  as the topological rigidities of the membrane. The membrane becomes unstable with respect to *continuous* deformations only under the more stringent condition that  $\kappa < 0$ , where  $\kappa = \kappa_+ + \kappa_-$ .

To describe the system's phase behavior, I use an analogous description of the free energy. The free energy of a fluctuating membrane of fixed topology, with a characteristic structural length  $\xi$  and equilibrium curvatures  $C_+$  and  $C_-$  can be written in terms of a renormalized bending energy [6]

$$H_R \approx \frac{1}{2} \int dA \{ \kappa_+(\xi) C_+^2 + \kappa_-(\xi) C_-^2 \}, \quad (4)$$

in which  $\kappa_+(\xi) \equiv \kappa(\xi) + (1/2)\bar{\kappa}(\xi)$  and  $\kappa_-(\xi) \equiv -(1/2)\bar{\kappa}(\xi)$ . The rigidities  $\kappa_\pm(\xi)$  both decrease with  $\xi$  as

$$\kappa_\pm(\xi) = \kappa_\pm - \frac{\alpha_\pm T}{4\pi} \ln\left(\frac{\xi}{a}\right), \quad (5)$$

with coefficients  $\alpha_+ = \frac{4}{3}$  and  $\alpha_- = \frac{5}{3}$  equivalent to the coefficients  $\alpha = 3$  and  $\bar{\alpha} = -\frac{10}{3}$  [6] for  $\kappa$  and  $\bar{\kappa}$ . By analogy with the definition of the persistence length  $\xi_\kappa$  as the length at which  $\kappa(\xi)$  vanishes, I define  $\xi_\pm \sim ae^{4\pi\kappa_\pm/(\alpha_\pm T)}$ , as the lengths at which these topological rigidities vanish.

For mechanically stable membranes, for which the bare  $\kappa_\pm > 0$ , one expects that upon dilution the  $L_\alpha$  phase will melt at a critical spacing  $d_c \approx \min(\xi_-, \xi_+, \xi_\kappa)$  corresponding roughly to the first value of  $d$  for which one of the rigidities

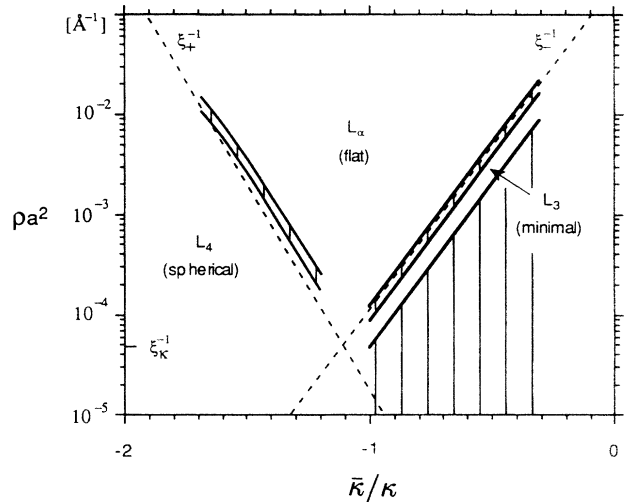


FIG. 1. Phase diagram for varying  $\bar{\kappa}/\kappa$  at fixed  $\kappa = 2T$  as a function of  $\rho a^2$  (i.e., inverse lamellar spacing). The solid lines show boundaries of idealized  $L_3$  and  $L_4$  phases within their expected regions of validity, with vertical tie lines to show phase coexistence between the  $L_\alpha$  and  $L_3$  or  $L_4$  phases, and between the  $L_3$  and a dilute phase of micelles or single surfactant molecules. The dashed lines show  $\xi_\pm^{-1}$ .

$\kappa_-(d)$ ,  $\kappa_+(d)$ , or  $\kappa(d)$  vanishes, indicating an instability towards the formation of, respectively, either nearly minimal, nearly spherical, or random surfaces. The rigidities are evaluated at length scale  $\xi \sim d$  because the characteristic lengths of the competing  $L_\alpha$  and disordered phases are expected to be similar near the melting transition. Using the calculated scale dependence of  $\xi_\kappa$  and  $\xi_\pm$ , it is found that

$$\xi_\kappa = \xi_+^{4/9} \xi_-^{5/9} \quad (6)$$

indicating that  $\xi_\kappa$  is always intermediate between the two topological persistence lengths. The instability associated with  $\kappa(\xi)$  is thus *always* preempted by one of the topological instabilities. This conclusion also follows immediately from the observation that, because  $\kappa(\xi) \equiv \kappa_+(\xi) + \kappa_-(\xi)$ ,  $\kappa(\xi)$  will always remain positive at a length  $\xi$  for which one of topological rigidities vanishes while the other remains positive. A crossover between the regimes  $\xi_- < \xi_\kappa < \xi_+$  and  $\xi_+ < \xi_\kappa < \xi_-$  can be affected by varying the ratio of bare parameters (Fig. 1) and occurs at the point

$$\bar{\kappa}/\kappa = -\frac{10}{9}, \quad \kappa_-/\kappa_+ = \frac{5}{4} \quad (7)$$

for which  $\xi_+ \sim \xi_- \sim \xi_\kappa$ .

This stability analysis suggests a division of the phase diagram into three regimes, based on the value of an elastic parameter  $\Delta \equiv \bar{\kappa} + (10/9)\kappa$ :

(I) For  $\Delta \geq T$  [or  $\bar{\kappa} \geq -(10/9)\kappa$ ], where  $\xi_- \ll \xi_\kappa \ll \xi_+$ , the  $L_\alpha$  phase is expected to melt at a spacing  $d \sim \xi_-$  into an  $L_3$  phase of multiply connected surfaces with small mean curvature  $C_+$ . A mean rigidity of  $\kappa(\xi_-) \approx (9/10)\Delta$  remains along this limit of stability and increases with increasing  $\Delta$ , thus tending to suppress conformations for which

$C_+ \neq 0$  and to make a description of the resulting phase as one of minimal surfaces increasingly accurate as  $\Delta$  is increased.

(II) For  $\Delta \leq -T$ , where  $\xi_+ \ll \xi_\kappa \ll \xi_-$ , the  $L_\alpha$  phase is expected melt upon dilution at a spacing  $d \sim \xi_+$  into an  $L_4$  phase of primarily spherical surfaces. A mean rigidity of  $\kappa(\xi_+) \approx (9/8)|\Delta|$  remains when  $\xi \sim \xi_+$ , which, as  $|\Delta|$  is increased, becomes increasingly effective in suppressing the appearance of nonspherical surfaces.

(III) For  $|\Delta| \ll T$ , where  $\xi_- \sim \xi_+ \sim \xi_\kappa$ , the  $L_\alpha$  phase is expected to melt at a spacing  $d \sim \xi_\kappa$  for which all of the renormalized rigidities are of order  $T$ . It is only within this crossover region that the physical assumption of a random surface and the corresponding prediction [8–11] of a structural length of order  $\xi_\kappa$  are correct.

These differences in geometrical structure are naturally parametrized by a quantity  $\psi \equiv 2\langle K \rangle / \langle C_1^2 + C_2^2 \rangle$  that measures the degree of correlation between the principal curvatures  $C_1$  and  $C_2$ , and that approaches  $\psi = \pm 1$  for  $|\Delta| \gtrsim T$ . A rough idea of the width of the crossover region, or the range of  $\Delta$  over which  $\psi$  crosses between  $\pm 1$ , can be obtained by considering the energies of various defects that are expected to appear in the idealized minimal and spherical structures as  $\Delta \rightarrow 0$ , such as the bending energy  $E = 4\pi^2\kappa(\xi_+) \sim -44.4\Delta$  of a toroid of size  $\xi_+$  within the  $L_4$  phase or the energy  $E = 8\pi\kappa_+(\xi_-) \sim 22.6\Delta$  of a disconnected sphere within the  $L_4$  phase. The relatively large [i.e.,  $O(10)$ ] prefactors found in such defect energies suggest the possibility of a rather narrow crossover region, of width significantly less than  $T$ , for which the division into three regimes would remain valid even for the rather small rigidities  $\kappa = (1-3)T$  typical of experiments.

As pointed by Cates *et al.* [8], the  $L_3$  and  $L_4$  phases can also be distinguished by considerations of symmetry: The properties of a bicontinuous phase are statistically symmetric with respect to a global relabelling of “inside” and “outside” volumes of solvent, while those of a vesicle phase are not. The existence of this hidden symmetry implies that there must exist a phase transition separating the “asymmetric”  $L_4$  phase from a “symmetric”  $L_3$  phase, where the  $L_3$  phase may continuously connect regions of random and nearly minimal surfaces. A separate transition between an  $L_3$  phase of disordered but nearly minimal surfaces and a true minimal crystal may occur for  $\bar{\kappa} \approx 0$ .

The results of an approximate calculation of the  $\bar{\kappa}$  dependence of the membrane phase diagram, in which the  $L_3$  and  $L_4$  phases have been treated as idealized structures of minimal and spherical surfaces, is shown in Fig. 1. The approach taken here is most appropriate for comparatively large  $\Delta$ , and is a natural complement to the random surface approximation appropriate for  $\Delta$  near zero. I consider a system with a number density  $\rho$  of surfactant molecules and a membrane area of  $a^2$  per molecule, giving a lamellar spacing of  $d = 1/(\rho a^2)$  in the  $L_\alpha$  phase and a structural length  $\xi \sim d$  in the competing  $L_3$  phase. For  $\Delta \gtrsim T$ , an  $L_3$  phase is found to be stable over a narrow range of concentrations  $\rho \sim 1/(\xi_- a^2)$  corresponding to a structural length  $\xi \sim \xi_-$  and to coexist with a phase of nearly pure solvent at lower concentrations, in agreement with the narrow range of stability seen in experiments [4,15–18,21]. For  $\Delta \leq T$ , an  $L_4$  phase is

found to be stable for concentrations below an upper limit of stability  $\rho \sim 1/(\xi_+ a^2)$  close to that predicted above, and remains stable upon dilution down to  $\rho = 0$ . The upper phase boundaries of the idealized  $L_3$  and  $L_4$  phases are seen to converge near the point  $\bar{\kappa}/\kappa \approx -10/9$ , giving an extrapolated phase boundary  $\rho \sim 1/(\xi_\kappa a^2)$  at this point of order that predicted by random surface models.

Details of this calculation are as follows: The free energy of the  $L_\alpha$  phase is approximated by the steric confinement free energy by Helfrich, given by

$$f_\alpha = C_\alpha a^6 \rho^3 T^2 / \kappa, \quad (8)$$

where  $C_\alpha$  is a numerical prefactor, for which I use Helfrich's estimate of  $C_\alpha = 3\pi^2/128$ .

For  $\Delta \gtrsim T$ , the free energy of the  $L_3$  phase is approximated by the renormalized bending energy  $H_R \approx \bar{\kappa}(\xi) \int dA K$  of a minimal surface. Assuming a structural length  $\xi \propto 1/(\rho a^2)$ , this gives a free energy density of the form

$$f_3 = C_3 a^6 \rho^3 T \ln(\rho/\rho_-), \quad (9)$$

where  $C_3$  is a dimensionless constant, and  $\rho_- \sim 1/(a^2 \xi_-)$  is the concentration corresponding to a structural length  $\xi = \xi_-$ . This functional form has been proposed previously [15,14] on the grounds of a scaling argument that assumes only that the structure in question dilates affinely under changes in  $\rho$ . The assumption of a minimal surface thus enters primarily through the identification of  $\xi_-$ , rather than  $\xi_\kappa$ , as the structural length for which  $H_R$  changes sign. For concreteness,  $\kappa_-(\xi)$  is evaluated in Eq. (5) by using a characteristic length  $\xi$  defined as the inverse rms curvature,  $\xi^{-2} \equiv (1/2)\langle C_1^2 + C_2^2 \rangle$ , consistent with the use of the radius on a spherical surface, and using a cutoff equal to the molecular length  $a$ . The remaining geometrical coefficients are set to values  $C_3 = 0.48$  and  $\rho_- = 0.74/(\xi_- a^2)$  that are within 10% of those appropriate for any of the simpler cubic minimal surfaces [22]. Free energy density (9) is concave at small  $\rho$ , and as a result is unstable for  $\rho < \rho_- e^{-1/2}$  towards coexistence with a phase of pure solvent.

For  $\Delta \leq -T$ , the  $L_4$  phase is described as a polydisperse suspension of spherical vesicles. The free energy is approximated by the ideal gas free energy

$$f_4 = \sum_{n \geq n_c} \rho_n \{ T \ln(\rho_n a^3) - T + F_n \}, \quad (10)$$

of such a suspension, as in Ref. [23], in which  $\rho_n$  is the number density for vesicles of  $n$  molecules and area  $A_n \equiv 4\pi R_n^2 = a^2 n$ ,  $n_c$  is a minimum vesicle size (chosen to give  $R_{n_c} = 50 \text{ \AA}$  in Fig. 1), and  $F_n \approx 8\pi\kappa_+(R_n) + (5/2)T \ln(n)$  is the internal free energy of a vesicle of constrained area and center-of-mass position, as calculated in [23]. Equilibrium values of  $\rho_n$  and  $f_4$  are obtained by minimizing (10) subject to a constraint on the total surfactant density  $\rho \equiv \sum_n n \rho(n)$ , and, in order to crudely take into account the effects of packing constraints at high concentrations, an additional constraint that the volume fraction  $\phi = \sum_n (4\pi R_n^3/3) \rho(n)$  enclosed by the vesicles not exceed some value  $\phi^*$  of order unity ( $\phi^* = 0.3$  in Fig. 1). These

constraints are imposed by introducing as Lagrange multipliers a chemical potential  $\mu$  and an inside-outside pressure difference  $p$ , where  $p=0$  for  $\phi < \phi^*$ . The average vesicle radius  $\langle R \rangle \propto \phi/\rho$  is found to increase with  $\rho$  as  $\langle R \rangle \propto \rho^{3/5}$  while  $\phi < \phi^*$ , and reaches a maximum value  $\langle R \rangle \sim \xi_+$  when  $\phi = \phi^*$  at a concentration  $\rho^* \approx 1/(\xi_+ a^2)$ , beyond which packing constraints become important and induce a first order transition to the  $L_\alpha$  phase.

The region near  $\bar{\kappa}/\kappa = -10/9$ , representing regime (III), has been left blank in the figure because the simple approximations used to calculate phase boundaries in the surrounding regions are not expected to be valid here. The arguments given above suggest, however, that this region should contain a random bicontinuous structure, much like that postulated by earlier random-surface models, that is continuously connected to the nearly minimal phase, but that is separated from the vesiclelike phase by a thermodynamic transition.

The above results are based upon the use of a perturbatively calculated renormalized elastic energy  $H_R$  that is actually valid [6] only for surfaces of characteristic structural length  $\xi \ll \xi_\kappa$ , for which  $\kappa(\xi) \geq T$ . It is thus important to note that within the quasirigid regimes (I) and (II),  $\xi \ll \xi_\kappa$  not only at the  $L_\alpha$ - $L_3$  and  $L_\alpha$ - $L_4$  transitions but at lower concentrations as well, due to the narrow range of stability of the  $L_3$  phase and the decrease of  $\langle R \rangle$  with decreasing  $\rho$  in the  $L_4$  phase, both of which act to prevent the relevant structural length from increasing further with increasing dilution. The

use of a perturbative treatment of undulations is thus valid throughout these regions.

The above analysis explores the effect of membrane elasticity upon the stability and topological structure of the disordered phases of an ensemble of membranes. By dropping the assumption of a randomly connected surface made some in earlier models, it is shown that the appearance of a bicontinuous phase in a system described by Hamiltonian (1) is primarily controlled by the Gaussian rigidity  $\bar{\kappa}$ , as argued by other authors on the basis of experimental evidence, and predicts a structure that ranges from random to nearly minimal depending upon the value of  $\bar{\kappa}/\kappa$ . The predicted phase diagram, in which an  $L_\alpha$  phase lies between disordered phases of different topology and inside-outside symmetry, is strikingly similar to those reported in Refs. [4,20], though in other systems a micellar or other nonbilayer phase is often found in place of the predicted vesicle phase. The prediction of an  $L_3$  phase with a structural length  $\xi \sim \xi_-$  that varies logarithmically with  $\bar{\kappa}$ , with a narrow range stability as a function of concentration, is consistent with the phase behavior obtained for simple binary solutions of water and nonionic surfactant [21]. Experiments to examine the effects of various control parameters upon both  $\bar{\kappa}$  and the structural length of either an  $L_3$  or bicontinuous microemulsion phase would provide the best way of further testing these ideas.

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