Thermal fluctuations and stiffening of symmetric heterogeneous fluid membranes

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(Received 2 September 2014; published 9 January 2015)

We study the effects of thermal fluctuations on symmetric tensionless heterogeneous (two-component) fluid membranes in a simple minimal model. Close to the critical point T_c of the associated miscibility phase transition of the composition and for sufficiently strong curvature-composition interactions, mediated through a composition-dependent bending modulus, thermal fluctuations lead to enhancement of the effective bending modulus. Thus, the membrane conformation fluctuations will be *suppressed* near T_c, in comparison with a pure fluid membrane, for which thermal fluctuations are known to reduce the effective bending modulus at all nonzero temperatures.

DOI: 10.1103/PhysRevE.91.012119 PACS number(s): 64.60.De, 05.70.Jk, 64.60.F−

I. INTRODUCTION

Miscibility phase transitions (MPT) in model heterogeneous membranes are prominent examples of phase transitions in two dimensions; see, e.g., Ref. [1] for a review on phase transitions with a connection to biomembranes. Typical experiments on model lipid bilayers, e.g., artificially prepared lipid bilayers made of lipids and cholesterol, display second-order MPTs from high-temperature homogeneous phases to a low-temperature phase-separated state with coexisting liquid-ordered (L_o) and liquid-disordered (L_d) domains [2–4] with distinctly different densities. For instance, a bilayer membrane composed of three components—dipalmitoylphosphatidylcholine (DPPC), diphytanoylphosphatidylcholine (diPhyPC), and cholesterol—clearly displays this MPT [5]. The universal scaling exponents that characterize MPTs in model lipid bilayers are experimentally found to be close to those of the two-dimensional (2D) Ising model [5,6]. In general, model lipid bilayers are symmetric under inversion, due to the identical nature of the two monolayers, i.e., nothing distinguishes the top from the bottom of the membranes. While MPTs for the composition field are well-studied, the statistical properties of the membrane conformation fluctuations at the MPTs have received much less attention theoretically.

It is well known that at any finite temperature T, a sufficiently large 2D tensionless pure or homogeneous (i.e., made of only one lipid) fluid membrane is *always crumpled* due to thermal fluctuations (see below), i.e., there are no *long-ranged orientational correlations* [7,8]. As a result, such a membrane cannot be in a statistically planar configuration beyond a certain size, determined by T and the microscopic bending rigidity κ_0. Physically this is seen as a consequence of the Mermin-Wagner theorem (MWT) [9]. At the critical temperature T_c of the MPT, the presence of the long-ranged composition fluctuations may introduce long-ranged interactions between different parts of the membrane. These long-ranged interactions should take the system outside the validity of the MWT. This leaves the nature of the effective membrane fluctuations open to various possibilities. Whether or not membrane fluctuations are affected by the MPTs remains a question of both theoretical and experimental significance. In this work, we address this issue by introducing a simple coarse-grained model for a tensionless two-component heterogeneous membrane, symmetric under inversion, useful as a model for lipid bilayers with two identical monolayers. We find that at the physically relevant dimension 2, the behavior of the membrane at or near T_c is essentially controlled by the strength of the interactions between the local curvature and composition (heterogeneity). If the strength of the interaction coupling between composition and membrane conformation fluctuations (introduced here through a composition-dependent bending modulus; see below) exceeds a critical value that depends on the temperature, the effective large-scale bending rigidity diverges with the system size L. As a result, the membrane conformation fluctuations are suppressed near T_c, or, equivalently, the membrane appears stiffer. The variance of the associated local normal fluctuations diverges very slowly with L; on a formal note, this L-dependence is found to be weaker than the L-dependence of the variance of the elastic variable fluctuations in models with continuous symmetries, described by an elastic free energy. On the other hand, if the interaction strength is less than the critical value, the effective scale-dependent bending rigidity vanishes for a sufficiently large length scale at any $T > 0$. Hence the membrane at that scale (or at larger scales) appears crumpled. In this respect, this behavior is qualitatively the same as that for a pure fluid membrane. In addition, we discuss the true area of the membrane and show that the fractal dimension $\tilde{d}$ approaches 2 near T_c if the curvature composition interaction strength exceeds the critical value, in agreement with the behavior of the associated membrane conformation fluctuations. The rest of the paper is organized as follows: In Sec. II, we construct our model for a two-component symmetric heterogeneous membrane. In Sec. III, we briefly discuss the phase transitions in the model within a mean-field approach. Then in Sec. IV, we consider the effects of thermal fluctuations on the membrane conformations at T_c. Next, in Sec. V, we discuss a generalized version of the above-mentioned model and show that the essential qualitative results do not change. Finally, in Sec. VI, we conclude.

II. THE MODEL

For simplicity, we ignore the bilayer structure [10] and consider a single membrane consisting of two different components (lipids) A and B. The local inhomogeneity is

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appropriately described by a single composition field $\phi(\mathbf{x})$, which is defined as the local difference between the concentrations n_A and n_B of A and B, respectively: $\phi = n_A - n_B$ [11]; ϕ is the order parameter field for the MPT. How composition fluctuations may affect the membrane conformation fluctuations depends upon the nature of coupling between the local composition and curvature. We consider a bilayer with two identical monolayers; toward that end, we study a symmetric membrane, invariant under inversion and hence with zero spontaneous curvature. In the simplest description for such a membrane, ϕ couples with the local mean curvature through a composition-dependent bending modulus $\kappa(\phi)$. We make a simple choice $\kappa(\phi) = \kappa + 2\bar{\lambda}\phi + 2\lambda\phi^2$ giving a minimal coupling between the composition and curvature (see, e.g., Ref. [11] for a similar model with $\lambda = 0$). The sign of $\bar{\lambda}$ is arbitrary, whereas $\lambda > 0$ strictly. Therefore, depending upon the sign of $\bar{\lambda}$, $\kappa(\phi)$ is either higher or lower in A- or B-rich regions. As a result, domains with smaller $\kappa(\phi)$ have a lower free-energy cost for supporting a given curvature of either sign. Naturally, regions of higher curvature will tend to favor regions of smaller curvature. Given the extensive experimental evidence in support of second-order MPTs in symmetric heterogeneous membranes, we adopt the standard Ginzburg-Landau free-energy functional for binary mixtures [12,13], useful near a critical point. We study a nearly flat membrane, for which membrane conformations are conveniently described by a single-valued height field $h(x,y)$ measured from a perfectly flat base plane in the Monge gauge [8]. Obviously, the choice for the base plane is arbitrary. Hence, the membrane free-energy functional density should be invariant under the tilt: $h \to h + \mathbf{a}\cdot\mathbf{r}$, where $\mathbf{a}$ is an arbitrary constant three-dimensional (3D) vector and $\mathbf{r}$ is a 3D radius vector.

Our model free-energy functional $\mathcal{F}$ for a two-component symmetric tensionless membrane (i.e., zero effective surface tension) [14] to the lowest order in gradients and nonlinearities is given by

$$\begin{aligned}\mathcal{F} &= \int dS\left[\frac{1}{2}\kappa(\phi)H^2 + \frac{r}{2}\phi^2 + \frac{1}{2}(\nabla_\alpha\phi)(\nabla^\alpha\phi) + \frac{u}{4!}\phi^4\right] \\ &= \int dS\left[\frac{1}{2}\kappa H^2 + \frac{r}{2}\phi^2 + \frac{1}{2}(\nabla_\alpha\phi)(\nabla^\alpha\phi)\right. \\ &\quad \left. + \frac{u}{4!}\phi^4 + \lambda\phi^2H^2 + \bar{\lambda}\phi H^2\right], \end{aligned} \tag{1}$$

where H is the mean curvature, ∇_α is the gradient operator, $\nabla^\alpha = g^{\alpha\beta}\nabla_\beta$, $g_{\alpha\beta}$ is the metric on the membrane, and $g_{\alpha\beta}g^{\beta\gamma} = \delta_\alpha^\gamma$ [8]. Further, κ is the (bare) bending modulus, $r \sim T - T_c$, $u > 0$ is a coupling constant, and $\lambda,\bar{\lambda}$ are coupling constants that couple the composition with the mean curvature. Note that the λ term in (1), which is quadratic in ϕ and H, ensures thermodynamic stability with λ chosen to be strictly positive. With ϕ as a number density, taking $\zeta \sim$ microscopic molecular length ($\sim$10 Å), $\phi \sim 1/\zeta^2$, $\lambda \sim K_BT\zeta^4$, $\bar{\lambda} \sim K_BT\zeta^2$. Further, surface element dS is related to the projected surface element $dx\,dy$ in a flat reference plane via $dS = dx\,dy\sqrt{g}$, $g = \det g_{\alpha\beta}$. In the Monge gauge, the metric tensor $g_{\alpha\beta}$ is given by

$$g_{\alpha\beta} = \begin{pmatrix} 1+(\partial_x h)^2 & \partial_x h\partial_y h \\ \partial_x h\partial_y h & 1+(\partial_y h)^2 \end{pmatrix}.$$

In addition, surface element $dS = dx\,dy\sqrt{1+(\nabla h)^2}$ and mean curvature $H = \nabla[\frac{-\nabla h}{\sqrt{1+(\nabla h)^2}}]$ in the Monge gauge. The membrane being symmetric is invariant under $h \to -h$. In the Monge gauge, $dS = \sqrt{1+(\nabla h)^2}dx\,dy \approx [1 + \frac{1}{2}(\nabla h)^2]dx\,dy$, assuming small height fluctuations for a nearly flat membrane. Evidently, if the nonlinear forms of dS and H are included in $\mathcal{F}$ above, additional nonlinear terms will be generated. These are *geometric nonlinearities* due to their origin in the Monge gauge, as opposed to the thermodynamic nonlinearities in $\mathcal{F}$.

III. PHASE TRANSITION

It is instructive to begin with a mean-field theory (MFT) description in terms of (assumed constant) order parameter $m = \phi$ and mean curvature $C = -\nabla^2 h$. Neglecting the geometric nonlinearities and minimizing $\mathcal{F}$ with respect to m and C, we obtain

$$rm + \frac{u}{3!}m^3 + 2\lambda mC^2 + \bar{\lambda}C^2 = 0, \tag{2}$$

$$\kappa C + 2\lambda Cm^2 + 2\bar{\lambda}mC = 0, \tag{3}$$

yielding $C = 0$ at all T and $rm + \frac{u}{3!}m^3 = 0$, which describes a second-order transition for the order parameter m with $m = 0$ for $r > 0$ ($T > T_c$) and $m^2 = -3!r/u$ for $r < 0$ ($T < T_c$). Solution $C = 0$ is consistent with the inversion symmetry of the model free energy (1), a requirement for any symmetric membrane. Notice that Eq. (3) may be written as

$$\kappa_m C = 0, \tag{4}$$

where $\kappa_m = \kappa + 2\bar{\lambda}m + 2\lambda m^2$ is the effective bending modulus in MFT (we ignore the possibility of $\kappa_m = 0$ for a nearly flat membrane). Clearly, with $\bar{\lambda} > 0$, k_m is reduced in domains with $m < 0$, but enhanced in domains with $m > 0$; $\lambda > 0$ contributes positively to κ_m for both signs of m. This is consistent with the interpretation of the $\bar{\lambda}$ and λ terms in $\mathcal{F}$, as discussed above.

IV. THERMAL FLUCTUATIONS AND BENDING MODULUS NEAR T_c

We begin with the partition function given by

$$\mathcal{Z} = \int Dh\,D\phi\,\exp[-\beta\mathcal{F}], \tag{5}$$

$\beta = 1/k_BT$. Taking the Boltzmann constant $k_B = 1$, T/κ is a dimensionless number in the problem. We wish to explore the possible nearly flat configuration of the membrane, such that the available thermal energy $\sim k_BT \ll$ bending energy, the scale of the latter being set by κ. Hence, we take $T/\kappa \ll 1$ [7,13]. Below we construct a perturbation theory with T/κ as the (small) expansion parameter; we calculate fluctuation corrections to κ to first order in T/κ. It is convenient to truncate the free-energy functional (1) up to $O(T/\kappa)$. We find to the lowest order in T/κ that the free energy $\mathcal{F}$

is given by

$$\mathcal{F} = \int d^2x \Big[\frac{\kappa}{2}(\nabla^2 h)^2 - \frac{\kappa}{4}(\nabla^2 h)^2(\nabla h)^2 - \kappa \nabla^2 h \nabla_\alpha h \nabla_\beta h \nabla_\alpha \nabla_\beta h + \frac{r}{2}\phi^2 + \frac{1}{2} g^{\alpha\beta}(\nabla_\alpha \phi)(\nabla_\beta \phi) + \frac{1}{4}(\nabla\phi)^2(\nabla h)^2 + \lambda \phi^2 (\nabla^2 h)^2 + \bar{\lambda}\phi(\nabla^2 h)^2 \Big], \quad (6)$$

where the nonlinear terms are kept up to $O(T/\kappa)$ [15]. Notice that (1) and hence (6) is invariant under inversion of the membrane, i.e., under $h \to -h$, as it should be for a symmetric membrane. However, the $\bar{\lambda}$ term in (1) or (6), being linear in ϕ, breaks the Ising symmetry. For the special case of $\bar{\lambda} = 0$, the Ising symmetry of ϕ is restored.

Consider a pure (homogeneous) fluid membrane, $\phi = 0$. It is evident from (6) with $\phi = 0$ that if the geometric nonlinearities are included, then κ receives a negative fluctuation correction proportional to $\langle (\nabla h)^2 \rangle$ [7]. In a renormalization-group (RG) language this implies that the effective scale-dependent bending modulus $\kappa(l)$ has no fixed point and flows to a negative value for a large length scale at any finite temperature $T > 0$. This result is interpreted as the impossibility of finding a (statistically) flat membrane beyond a finite scale determined by T and the bare or small-scale bending modulus κ_0 at dimension $d \leqslant 2$ [7]. On the other hand, for a heterogeneous membrane $\phi \neq 0$, and the λ term positively contributes to the scale-dependent $\kappa(l)$ by an amount proportional to $\langle \phi^2 \rangle$. This contribution formally diverges at T_c and hence can potentially lead to stiffening of membranes. Thus, for an inhomogeneous membrane, the competition between the geometric nonlinearity and the nonlinearity of the curvature-composition interaction, mediated by a composition-dependent $\kappa(\phi)$, should determine the membrane fluctuations in the thermodynamic limit.

We employ the perturbative Wilson RG procedure [13,16] to the lowest order in $O(T/\kappa)$ to circumvent the difficulty in perturbative expansions near T_c due to the large fluctuations. In addition, we obtain our results within a harmonic approximation for ϕ, i.e., we set $u = 0$ for simplicity. We eliminate fields $h(\mathbf{q})$ and $\phi(\mathbf{q})$ with wave vector $\Lambda/b < q < \Lambda$, $b > 1$, where Λ is an upper cutoff for the wave vector, by integrating over them perturbatively up to one-loop order and then rescaling wave vectors according to $\mathbf{q}' = b\mathbf{q}$ (or rescaling real-space coordinate $\mathbf{x}$ according to $\mathbf{x}' = \mathbf{x}/b$). Since we are interested in finding the renormalization of κ due to the geometric nonlinearities and the curvature-heterogeneity couplings, it is convenient to let $h(\mathbf{x})$ rescale as $h(\mathbf{x}') = h(\mathbf{x})/b$ with $\mathbf{x}' = \mathbf{x}/b$. Further, rescale ϕ by $\phi(\mathbf{x}') = b^{-\epsilon/2}\phi(\mathbf{x})$, $\epsilon = 2 - d$. Under this rescaling, $\kappa' = b^{-\epsilon}\kappa$, $\lambda' = b^0\lambda$, $\bar{\lambda}' = b^{-\epsilon/2}\bar{\lambda}$.

One-loop corrections to κ may be straightforwardly extracted from (6) by contracting fields in the nonlinear terms leaving two external $\nabla^2 h$ legs [17]. We follow the procedure as outlined above and find perturbatively

$$b^\epsilon \kappa' - \kappa = -\kappa \frac{3}{2}\langle (\nabla h)^2 \rangle_\Lambda + 2\lambda \langle \phi^2 \rangle_\Lambda - \bar{\lambda}^2 \int_{\Lambda'}^{\Lambda} \frac{d^2q}{(2\pi)^2} \langle |\phi(\mathbf{q})|^2 q^4 |h(\mathbf{q})|^2 \rangle, \quad (7)$$

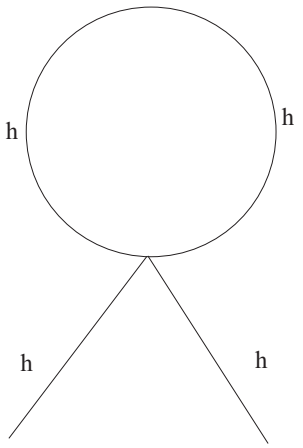

FIG. 1. One-loop Feynman diagram contributing to the fluctuation correction to κ. This originates from the geometric nonlinearity; see Eq. (8).

where $\Lambda/\Lambda' = b$ and

$$\langle (\nabla h)^2 \rangle_\Lambda = \int_{\Lambda/b}^{\Lambda} \frac{d^2q}{(2\pi)^2} \frac{Tq^2}{\kappa q^4} = T \frac{\ln b}{2\pi\kappa}, \quad (8)$$

$$\langle \phi^2 \rangle_\Lambda = \int_{\Lambda/b}^{\Lambda} \frac{d^2q}{(2\pi)^2} \frac{T}{q^2} = \frac{T \ln b}{2\pi}. \quad (9)$$

The two relevant one-loop Feynman diagrams corresponding to the corrections (8) and (9) are shown in Figs. 1 and 2, respectively.

By using Gaussian decomposition and $\langle |h(\mathbf{q})|^2 \rangle = T/\kappa q^4$, we note from Eq. (7) that the contribution to κ' from the $\bar{\lambda}$ term in (1) or (6) is linear in T/κ, whereas the remaining contributions are $O(T/\kappa)^0$. Thus, we neglect this $O(T/\kappa)$ contribution from the $\bar{\lambda}$-nonlinear term below.

Now, let $b = e^{dl} \simeq 1 + dl$. Then,

$$\frac{d\kappa}{dl} = \kappa \Big[-\epsilon - \frac{3T_c}{4\pi\kappa} + \frac{2\lambda T_c}{2\pi\kappa} \Big]. \quad (10)$$

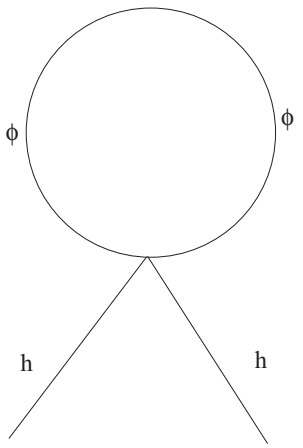

FIG. 2. One-loop Feynman diagram contributing to the fluctuation correction to κ. This is due to the composition fluctuations and exists only for inhomogeneous membranes; see Eq. (9).

Notice that in the flow Eq. (10), we have set $T = T_c$, since we are at the critical temperature of the MPT. At the RG fixed point (FP), $d\kappa/dl = 0$, which yields

$$-\epsilon - \frac{3T_c}{4\pi} + \frac{2\lambda_c T_c}{2\pi} = 0, \tag{11}$$

which defines a critical λ_c:

$$\lambda_c = \frac{\pi\epsilon}{T_c} + \frac{3}{4}, \tag{12}$$

for this unstable RG FP. Thus for $\lambda < \lambda_c$,

$$\frac{d\kappa}{dl} < 0, \tag{13}$$

implying that under successive applications of the RG procedure, scale-dependent $\kappa(l)$ reduces linearly with l, eventually becoming zero at a particular scale that depends upon κ and λ [18]; see also Ref. [7]. Thus even for a heterogeneous fluid membrane, for a weak coupling $\lambda < \lambda_c$, the membrane still crumples at any T for a sufficiently large size. The discrete recursion relation for κ at 2D is given as

$$\kappa = \kappa_0 - \frac{3T_c}{4\pi}\ln b + \frac{\lambda T_c}{\pi}\ln b. \tag{14}$$

Noting that $\ln b = \ln(\Lambda/q) = -\ln(qa_0)$, where $a_0 \sim \Lambda^{-1}$ is a microscopic length, we obtain a q-dependent $\kappa(q)$,

$$\kappa(q) = \kappa_0 + \frac{3T_c}{4\pi}\ln(qa_0) - \frac{\lambda T_c}{\pi}\ln(qa_0). \tag{15}$$

Similar to Ref. [7], Eq. (15) allows us to define a de Gennes–Taupin persistence length $\xi \sim 1/q$, such that $\kappa(\xi) = 0$. This yields

$$\xi = a_0 \exp\left(\frac{4\pi\kappa_0}{T_c(3-4\lambda)}\right). \tag{16}$$

Clearly, as $\lambda \to \lambda_{c-} = 3/4$ at two dimensions, the membrane crumples less and less, and $\xi \to \infty$.

In contrast, for $\lambda > \lambda_c$,

$$\frac{d\kappa}{dl} > 0 \tag{17}$$

generically, and hence κ grows under successive applications of the RG transformations. Therefore, the membrane at larger scale should appear *stiffer* than it is at smaller scales. Clearly, as dimensionality rises, i.e., as ϵ reduces and becomes negative through zero, λ_c reduces. Formally, therefore, at higher dimensions, weaker curvature-heterogeneity interactions through $\kappa(\phi)$ are enough for $\kappa(l)$ to grow under RG transformations. To know whether or not a heterogeneous fluid membrane with $\lambda > \lambda_c$ crumples in the thermodynamic limit, we need to find $\kappa(q)$ in the limit $q \to 0$ with $\lambda > \lambda_c$. Define $\Delta\lambda = \lambda - \lambda_c > 0$. Then,

$$\kappa(q) = -\frac{\Delta\lambda T_c}{\pi}\ln(qa_0) + \kappa_0, \tag{18}$$

which diverges in the thermodynamic limit $q \to 0$.

The statistical flatness of a membrane may be ascertained by the variance of the fluctuation of the local normal $\mathbf{n} = (-\nabla h, 1)$. We define fluctuation in the normal, $\delta\mathbf{n} = -\nabla h$, as the deviation from the normal for a perfectly flat surface. Now calculate $\Delta_0 = \langle(\delta\mathbf{n})^2\rangle$. If Δ_0 at a given temperature for a membrane (pure or inhomogeneous) is independent of the system size L, Δ_0 remains finite even in the thermodynamic limit $L \to \infty$. This implies that the membrane at that temperature possesses a long-ranged orientational order in the thermodynamic limit, or the membrane is statistically flat [8,13]. Otherwise the membrane is crumpled. We find at two dimensions,

$$\langle(\delta\mathbf{n})^2\rangle = \int \frac{d^2q}{(2\pi)^2}\langle|\delta\mathbf{n}_\mathbf{q}|^2\rangle \sim \int_L \frac{d^2q}{(2\pi)^2}\frac{T_c}{\kappa(q)q^2}, \tag{19}$$

where a subscript L refers to a lower momentum cutoff $\sim 1/L$. With $\kappa(q) \sim -\frac{T_c}{\pi}\Delta\lambda\ln(qa_0)$, clearly, within the validity of our calculations ($T_c/\kappa \ll 1$),

$$\Delta_0 \sim -\frac{1}{\Delta\lambda}\int \frac{dq}{q\ln(qa_0)} \sim -\frac{1}{\Delta\lambda}\ln\ln\left(\frac{a_0}{L}\right) \tag{20}$$

in the critical region ($T = T_c$), regardless of the specific value of T_c, so long as $\lambda > \lambda_c$. Thus, Δ_0 still diverges for $L \to \infty$, albeit very slowly, and it is finite for any finite L. In fact, the system size L required to have a given Δ_0 is given by $L/a_0 \sim \exp(\exp[\Delta_0\Delta\lambda])$. Thus, L rises very rapidly with $\Delta\lambda$. Compare this with a pure fluid membrane, for which $\kappa(q)$ vanishes for a low enough q. Thus for a pure fluid membrane Δ_0 should diverge even for a finite system size ($\sim\xi$). Notice that since $\kappa(q)$ here remains finite (in fact growing) even down to $q \to 0$, ξ is formally infinitely large. As a result, for a finite membrane of any size, $\Delta_0 \neq 0$, and hence a nonzero orientational correlation should be present. The very weak divergence of Δ_0 with L means its experimental detection should be rather difficult; crumpling may be observed only in a very large membrane. Notice that the $\ln\ln(L/a_0)$ dependence on L in Eq. (20) is even *weaker* than the well-known $\ln L$ dependence of the variance of orientation fluctuations in 2D classical spin systems having continuous symmetries that display QLRO [13].

Noting that (12) yields an unstable FP for $d\kappa/dl = 0$, our results above may be interpreted as a phase transition in the membrane between a crumpled phase (finite ξ, $\Delta\lambda < 0$) and a stiff phase (diverging ξ, $\Delta\lambda > 0$) for a given $T = T_c$, with λ appearing as the control parameter. We take $\overline{O} = [\ln(\xi/a_0)]^{-1} = -T_c\Delta\lambda/(\pi\kappa_0)$ with $\lambda < \lambda_c$ and $\overline{O} = 0$ with $\lambda \geqslant \lambda_c$ as the

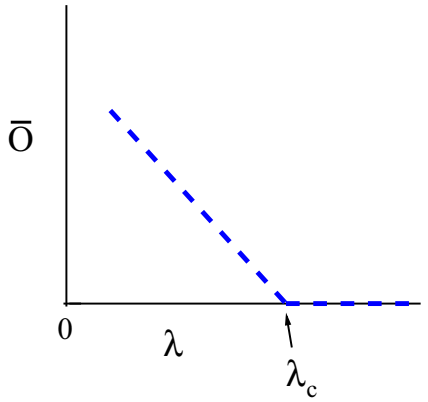

FIG. 3. (Color online) Schematic variation of order parameter $\overline{O}$ as a function of λ; $\overline{O} \propto (\lambda - \lambda_c)$ for $\lambda \leqslant \lambda_c$, $\overline{O} = 0$ for $\lambda\lambda_c$ as shown by the blue broken line; see the text.

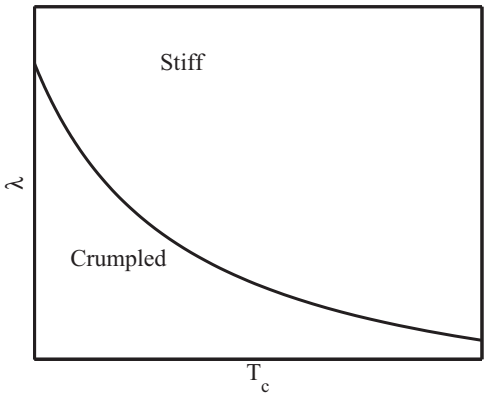

FIG. 4. Schematic phase diagram in the λ-T_c plane for $\epsilon > 0$. The continuous line represents Eq. (12).

order parameter; thus $\overline{O}$ in the crumpled phase rises smoothly from zero as λ is reduced from λ_c. Thus, with λ as the control parameter, the "order parameter exponent" is 1. In the stiff phase, $\overline{O}$ is naturally zero. We plot $\overline{O}$ versus λ in Fig. 3. Figure 4 shows a schematic phase diagram in the λ-T_c plane.

Notice that the membrane fluctuations are unaffected by the $\bar{\lambda}$ term to the leading order in T/κ. This suggests that in the long-wavelength limit, the Ising symmetry is restored. So far we have discussed the membrane fluctuations at the MPT, i.e., at $T = T_c$. Away from T_c, contribution of the λ term to renormalized κ is small; renormalized κ is dominated by the contributions from the geometric nonlinearities. Therefore, at $T \neq T_c$, the large-scale properties of a heterogeneous fluid membrane described by (1) are identical to a pure fluid membrane. However, as one approaches T_c from either side, the contribution from the λ term becomes significant. For a membrane of finite linear size L, $\langle(\nabla h)^2\rangle$ scales as $\ln L$, whereas $\langle\phi^2\rangle \sim \ln \frac{L}{L\sqrt{r}+1}$ at $T > T_c$, i.e., $r > 0$. Clearly, the latter contribution rises as $r \to 0$, and hence renormalized κ, which now depends on L, rises in magnitude. Therefore, any measurements of κ as a function of T should detect this rise as T_c is approached. Similar behavior for $\kappa(L)$ follows for $T < T_c$ as well. A schematic plot of $\kappa(L)$ versus r is shown in Fig. 5. The general nature of this plot remains unchanged regardless of whether $\lambda > \lambda_c$ or not.

We now calculate the effective membrane area $S = \langle\sqrt{1+(\nabla h)^2}\rangle S_0$ and compare it with the base area S_0 (the projected area on the Monge gauge reference plane) by defining the fractal dimension $\tilde{d}$ of the membrane via the relation $S \propto S_0^{\tilde{d}/2}$. Hence, for $\tilde{d} = 2$, $S \propto S_0$, corresponding to a membrane that is on average flat, while for $\tilde{d} > 2$, S rises faster than S_0 and hence a more corrugated membrane at larger scales is obtained. We find at T_c, to the lowest order in h fluctuations [7],

$$\frac{\tilde{d}}{2} = \frac{d\ln S}{d\ln S_0} \simeq 1 + \frac{1}{2}\frac{d}{d\ln S_0}\langle(\nabla h)^2\rangle = 1 + \frac{1}{8\pi}\frac{T_c}{\kappa}. \tag{21}$$

Thus, if $\lambda < \lambda_c$, renormalized κ gets smaller at larger scales, and hence $\tilde{d} > 2$, implying a highly corrugated membrane at large scales. In contrast, for $\lambda > \lambda_c$, the effective scale-dependent κ diverges in the long wavelength limit. Thus, $\tilde{d} \to 2$, and hence a nearly flat membrane is obtained. These conclusions are consistent with the behavior of $\langle(\delta\mathbf{n})^2\rangle$ for $\lambda < \lambda_c$ and $\lambda > \lambda_c$, respectively. We close this section with a technical comment. The phase transition elucidated above takes place at a fixed temperature T_c, with λ appearing as the control parameter. Thus, experimentally, various heterogeneous membranes described by (1) will exhibit different characteristic membrane fluctuations at T_c, depending upon whether $\lambda > \lambda_c$ or not. This is different from the usual crumpling transition of membranes [8], where the membrane goes from the high-temperature crumpled phase to the low-temperature flat phase through a critical temperature of the crumpling transition; the temperature is the control parameter of the usual crumpling transition, unlike in the present case.

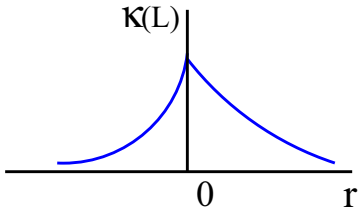

FIG. 5. (Color online) Schematic plot of renormalized κ for a system of size L as a function of $r = T - T_c$; a rise in κ as $T \to T_c$ due to a nonzero λ is shown.

V. GENERALIZED MODEL

Our results above depend crucially on the positivity of the λ term in (1). We now generalize (1) by considering an arbitrary form of interaction $\tilde{f}$ between the local curvature and inhomogeneity, maintaining only the tilt invariance and invariance under inversion of h. We briefly discuss its effects on the membrane fluctuations near T_c. Expanding $\tilde{f}$ in powers of ϕ and $(\nabla^2 h)^2$, we write the generalized free-energy functional $\mathcal{F}_g$ as

$$\mathcal{F}_g = \int dS \left\{ \frac{\kappa}{2}(\nabla^2 h)^2 + \frac{r}{2}\phi^2 + \frac{1}{2}g^{\alpha\beta}(\nabla_\alpha\phi)(\nabla_\beta\phi) + \frac{u}{4!}\phi^4 + \sum_{m,n} A_{m,n}\phi^m(\nabla^2 h)^{2n} \right\}. \tag{22}$$

Thus, comparing (22) with (1) above, we find

$$A_{1,1} = \bar{\lambda}, \quad A_{2,1} = \lambda. \tag{23}$$

All the other A's are zero in (1). It is clear that contributions of all terms in $\tilde{f}$ with $n > 1$ to renormalized κ are higher order in T/κ. Thus, in a perturbation theory first order in T/κ, all such terms may be ignored here. The remaining terms in $\tilde{f}$ are all quadratic in $\nabla^2 h$:

$$\tilde{f} = \sum_m A_{m,1}\phi^m(\nabla^2 h)^2. \tag{24}$$

For reasons of thermodynamic stability, the series on the right side of (24) must be truncated at an even $m = m_{\max}$, with

$A_{m_{\max},1} > 0$. Notice that under spatial rescaling, $\mathbf{x}' = \mathbf{x}/b$, $A'_{2m} = b^{(m-1)(d-2)}$, implying that for $m > 1$, all of A_{2m} decreases under the successive application of rescaling. At $d < 2$, following the calculation outlined above, it is straightforward to obtain the flow equation for κ for the generalized model. We obtain at $d = 2 - \epsilon$ to the leading order in T/κ

$$\frac{d\kappa}{dl} = -\epsilon - \frac{3T_c}{4\pi} + 2A_{2,1}\langle\phi^2\rangle, \tag{25}$$

up to the one-loop order. Equation (25) is identical in form to its counterpart (10) above. However, unlike the previous case, A_2 (in fact all of A_{2m} with $m < m_{\max}$) has one-loop fluctuation corrections at $O(T/\kappa)^0$. For $d \leqslant 2$ and at $T = T_c$, fluctuations corrections to A_{2m} diverge with L and will dominate over the corresponding bare parameters. Since A_{2m} with $m = m_{\max}$ is necessarily positive, it is expected that all the fluctuation-corrected (i.e., renormalized or effective) A_{2m} should be positive. Then Eq. (25), with $A_{2,1}$ being interpreted as the renormalized quantity (hence positive), yields similar behavior for $\kappa(q)$ from (1). In the generalized model A_{2m} with $m = m_{\max}$ appears as the tuning parameter [19].

VI. SUMMARY AND OUTLOOK

We have thus proposed a simple coarse-grained model for symmetric heterogeneous tensionless membranes in terms of the local mean curvature and a suitably defined composition field ϕ in the Landau-Ginzburg approach to study the nature of membrane conformation fluctuations near the critical point of the MPT. Our model should be useful for studying symmetric bilayers made of identical monolayers with strong intermonolayer interactions. Within the range of validity of a systematic perturbative expansion in $T/\kappa \ll 1$ (with $T = T_c$), we show that the lowest-order nonlinear term that obeys the Ising symmetry for ϕ leads to an enhancement of renormalized or effective κ, provided its strength λ exceeds a critical value. In that case, renormalized κ diverges in the thermodynamic limit, albeit very slowly. Nonetheless Δ_0, the variance of the local normal fluctuations, grows with system size L, although very weakly, $\Delta_0 \sim -\ln\ln(a_0/L)$. This is weaker than the $\ln L$-dependence of the variance of elastic variable fluctuations in classical models with continuous symmetries, described by elastic free energies. This aspect makes it a theoretically intriguing result. Thus, the membrane should appear crumpled in the formal thermodynamic limit, which in practice may be observed at a very large scale due to the very weak L dependence of Δ_0; experimental studies on finite-size membranes may not be able to observe this. We have argued that our results may be interpreted as a phase transition between a crumpled phase (finite ξ) and a stiff phase (diverging ξ). With λ as a control parameter for a given T_c, the transition is second order characterized by an order parameter $\overline{O} = [\ln(\xi/a_0)]^{-1}$. A generalized model by us predicts qualitatively similar results. With $T_c \sim 300$ K and bare $\kappa \sim 10^{-12}$ erg [20] for a lipid bilayer, $T_c/\kappa \sim 0.05$ is small. Thus, our results should hold for MPT in a typical model heterogeneous bilayer. By tuning λ, experiments (e.g., flicker experiments) on MPTs in a two-component symmetric model lipid bilayer should reveal the suppression of membrane conformation fluctuations (or membrane height fluctuations) for $\lambda > \lambda_c(T_c)$ as T_c is approached. Hence, our results offer an experimental route to investigate the curvature-composition interactions in an inhomogeneous membrane.

Our results depend sensitively on λ. Since $\lambda \sim \zeta^4 k_B T$, performing experiments on model heterogeneous membranes with different sizes of the constituent lipid molecules (and hence varying ζ) should be a promising route to test our results experimentally. Our results highlight the significant differences in the fluctuations of asymmetric [21] and symmetric inhomogeneous membranes near T_c.

We close our discussions with some technical comments. While contributions to renormalized κ from the $\overline{\lambda}$ term are neglected for small $O(T/\kappa)$, for T_c/κ not very small, these may be retained. Notice that this contribution reduces κ; see also Ref. [11]. We have treated ϕ only up to the quadratic order and have set $u = 0$. The effects of a nonzero u may be accounted for by considering ϕ as the *renormalized composition field* with a renormalized critical temperature and a nonzero anomalous dimension. To the lowest order in u, the only effect would be to shift T_c, and our basic results should stay unchanged. The universal properties of ϕ fluctuations are described by the Ising universality class by the ϕ^4 term. It is well known that [1] a small amount of impurities added to an otherwise pure system keeps the critical behavior unchanged, affecting only the value of the critical temperature. In contrast, higher concentration of impurities are known to alter the nature of the transition qualitatively. Noting that in our model free energy (1) the effective critical temperature $\tilde{T}_c = T_c - 2\lambda\phi^2 - 2\overline{\lambda}\phi$ is composition-dependent, it would be interesting to explore possible connections between our results here and the discussions in Ref. [1]. While constructing the form of $\mathcal{F}$ as given in (1), we have coupled the composition with the mean curvature H, but we have ignored the Gaussian curvature G. For a pure fluid membrane with a fixed topology, G does not play any role in the statistical mechanics of the system, due to the Gauss-Bonnet theorem [13]. For a heterogeneous membrane, it is possible to introduce terms coupling ϕ and G. However, they do not contribute to the renormalization of κ to first order in T/κ, and hence they are neglected. We have ignored the technical issue of choosing the correct measure in $\mathcal{Z}$ [22]. These are not expected to affect our low-order perturbative results. Our model may be extended for multicomponent (more than two components) heterogeneous membranes in a straightforward way by introducing additional composition variables and coupling all of them to the mean curvature in ways similar to (6). We expect that the general features of our results should hold there. Lastly, the dynamical behavior of a heterogeneous membrane modeled by (1) near T_c would be interesting to study theoretically [see, e.g., Ref. [23] for a study of the dynamics of a heterogeneous membrane with a curvature-composition interaction from (1)].

ACKNOWLEDGMENTS

We thank J. K. Bhattacharjee for his valuable comments and suggestions. A.B. wishes to thank the Max-Planck-Gesellschaft (Germany) and the Department of Science and Technology/Indo-German Science and Technology Centre (India) for partial financial support through the Partner Group programme (2009).

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