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Fluctuating Membranes With Tilt Order

Abstract

Thermal fluctuations are important for amphiphilic bilayer membranes since typical bending stiffnesses can be a few $k_B T$. The rod-like constituent molecules are generically tilted with respect to the local normal for packing reasons. We study the effects of fluctuations on membranes with nematic order, a simplified idealization with the same novel features as realistic tilt order. We find that nematic membranes lie in the same universality class as hexatic membranes, i.e. the couplings that distinguish nematic from hexatic order are marginally irrelevant. Our calculation also illustrates the advantages of conformal gauge, which brings great conceptual and technical simplifications compared to the more popular Monge gauge.

Disciplines

Physical Sciences and Mathematics | Physics

Fluctuating Membranes With Tilt Order

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Thermal fluctuations are important for amphiphilic bilayer membranes since typical bending stiffnesses can be a few $k_B T$. The rod-like constituent molecules are generically tilted with respect to the local normal for packing reasons. We study the effects of fluctuations on membranes with nematic order, a simplified idealization with the same novel features as realistic tilt order. We find that nematic membranes lie in the same universality class as hexatic membranes, *i.e.* the couplings that distinguish nematic from hexatic order are marginally irrelevant. Our calculation also illustrates the advantages of conformal gauge, which brings great conceptual and technical simplifications compared to the more popular Monge gauge.

The study of fluctuating surfaces in an intriguing statistical mechanics problem with numerous applications. Amphiphilic molecules in water can self-assemble into thin flexible bilayer membranes which provide an experimental realization of random surfaces [1]. As we recall below, a membrane’s internal order determines the nature of its shape fluctuations. Although fluctuating membranes with hexatic bond-orientational order have received a lot of theoretical attention [2][3][4][5], the generic situation is for the rod-like constituent molecules to *tilt* with respect to the local surface normal. We shall see that tilt order differs from hexatic order since tilt allows certain anisotropic couplings. In this letter we consider “nematic” membranes, the simplest membrane model with in-plane orientational order and anisotropy. We find the anisotropy to be marginally irrelevant; these membranes may be considered hexatic at very long length scales.

Our framework is continuum elastic theory since the micron size of membrane structures is much larger than the typical molecular size, a few tens of Ångstroms. If the molecules are allowed to take up their preferred area, then surface tension will be unimportant and bending rigidity will dominate. Typical bending moduli for lipid bilayer membranes are on the order of $10k_B T$ [6], which is low enough for thermal fluctuations to play a role. We study fluid-like membranes in which the molecules are free to diffuse along the membrane in response to shape fluctuations. Therefore shape and orientational order are the only elastic degrees of freedom, and our elastic theory must be coordinate-invariant [7].

The simplest type of in-plane orientational order is hexatic order [8]. Pure hexatic order is readily seen in Langmuir monolayers [9] and thin smectic liquid crystal films [10], but this order has not to our knowledge been confirmed in bilayer membranes. Instead, in the ordered phases the constituent molecules generically tilt for packing reasons. If tilt and hexatic order are present together [11], then in the absence of defects we can for the purposes of elastic theory treat these as locked together. Tilt and hexatic order break the same rotational symmetry, and together lead to just one elastic mode. Since tilt order has less symmetry than hexatic order (see below), the relevant order is tilt when both tilt and hexatic order are present.

One can represent hexatic order on a membrane by a unit tangent vector field [3]. Each vector points from a molecule to one of the molecule’s nearest neighbors and is defined up to $2\pi/6$ rotations. Demanding this symmetry automatically brings along a larger symmetry, *i.e.* global rotations through an arbitrary angle; in this sense hexatic order is isotropic. This is reminiscent of the isotropy of the continuum elastic theory of a

two-dimensional triangular lattice [12], but holds even on curved surfaces, whose principal curvatures could in principle have cared about a global rotation of in-plane order.

Similarly one can represent tilt order as a unit tangent vector field \hat{m} defined by the direction of the projection of the axis of each molecule on the local tangent plane. We consider only the elastic modes so we fix the polar angle between the normal \hat{n} and the molecular axis. Bilayers lack a preferred normal, so to complete our specification of \hat{m} we demand the bilayer symmetry $\hat{n} \rightarrow -\hat{n}$, $\hat{m} \rightarrow -\hat{m}$ [13][14]. In contrast to the hexatic case, the energy for bending a membrane along an axis parallel to the direction of tilt can clearly be different from the energy to bend the membrane the other way. (Mathematically we will see in a moment how this conclusion arises from the reduced symmetry of tilt order.) This tilt-shape coupling has a number of simple consequences; for example, tilt order is responsible for the $P_{\beta'}$ ripple phases [11][15]. Tilt order is also manifested dramatically when the constituent molecules are chiral: the microscopic molecular chirality affects macroscopic membrane shape only in the presence of tilt [16]. Tilt order can play a role in the budding of artificially manufactured vesicles [17] and it may be the typical order in biological membranes [18].

The nature of membrane shape fluctuations depends on the degree of internal order. We characterize these fluctuations by using the Wilson renormalization group to compute the long-distance behavior of the elastic couplings. For example, the bending rigidity of a fluid membrane becomes ineffective beyond a persistence length ξ_P , leading to a crumpled phase at any nonzero temperature [19].¹ Internal order tends to stiffen a membrane. For stiff enough elastic constants, the bending rigidity for a hexatic membrane reaches a fixed point at very long length scales, possibly leading to a “crinkled” phase with quasi-long-range order in the normals [3]. Finally, non-self avoiding tethered membranes (crystalline membranes with an infinite core energy for dislocations) undergo a crumpling transition and have a flat phase at low temperature [20]. It is natural to extend these analyses to the case of tilt order, since as we remarked it is experimentally the most relevant regime with orientational order. To keep our formulas compact we will impose a “nematic,” or 2-atic symmetry to get the simplest model with anisotropy; the same technique applies to the more realistic case with no discrete symmetry. We will then ask, does the anisotropy lead to new fixed points and new physics, or is the anisotropy marginally irrelevant as in flat thin liquid crystal films [2]?

¹ We ignore self-avoidance since we will ultimately study a fixed point at large stiffness.

To answer these questions with precise calculations, we must choose coordinates. The most popular choice for such calculations is Monge gauge, in which the surface is parametrized by its height above some flat reference surface [19][14]. It turns out that Monge gauge is not very convenient for tilted membranes; conformal gauge, as used for example by Polyakov [21], is much better-suited. Conformal gauge has the technical advantages of leading to compact expressions and thus less algebra than Monge gauge, as well as the conceptual advantage of making explicit a useful separation between extrinsic and intrinsic geometry.

We begin by describing the geometrical constructions necessary to write down the tilt free energy. Even with the benefits of conformal gauge, the formulas can get a bit long. Thus, we make some inessential but simplifying assumptions. Next, we briefly summarize the issues involved in conformal gauge calculations. Finally, we present our result and discuss its implications.

Following the notation of [21], we parametrize our surface by $\vec{x}(\xi)$, where \vec{x} is a three dimensional vector and (ξ^1, ξ^2) are the (arbitrary) two dimensional coordinates. From $\vec{x}(\xi)$ we construct the metric tensor $g_{ab} = \partial_a \vec{x} \cdot \partial_b \vec{x}$ and the second fundamental form $K_{ab} = \hat{n} \cdot \partial_a \partial_b \vec{x}$, where $\partial_a = \partial / \partial \xi^a$ and \hat{n} is the local normal. We denote the covariant derivative associated with g_{ab} by ∇ , the inverse of g_{ab} by g^{ab} , and the determinant of g_{ab} by g . It is useful to introduce a local set of orthonormal frames \hat{e}_α ; we denote by e_α^a the change-of-basis matrix that converts orthonormal frame indices α, β, \dots to coordinate indices a, b, \dots . Thus for example $\nabla_a m^\alpha = \partial_a m^\alpha + \epsilon^\alpha_\beta \Omega_a m^\beta$ where Ω_a is the spin connection [3].

The elastic free energy for tilted bilayer membranes must have coordinate invariance, Euclidean invariance, and the discrete bilayer symmetry discussed above. Writing only the bulk terms quadratic in the curvature or derivatives of \hat{m} , *i.e.* only the marginal terms, we find $F_{\text{tilt}} = F_1 + F_2$ where

$$\begin{aligned}
F_1 &= \frac{1}{2} \int d^2 \xi \sqrt{g} [k_1 (\nabla \cdot \hat{m})^2 + k_2 (\nabla \times \hat{m})^2 + \kappa (K_a^a)^2 \\
&\quad + \kappa_1 \hat{m} \cdot K \cdot K \cdot \hat{m} + \kappa_2 (\hat{m} \cdot K \cdot \hat{m})^2] \\
F_2 &= \frac{1}{2} \int d^2 \xi \sqrt{g} [\beta_1 (\hat{m} \cdot K \cdot \hat{m}) (\nabla \cdot \hat{m}) + \beta_2 (K_a^a) (\nabla \cdot \hat{m}) \\
&\quad + \beta_3 K^a_b \nabla_a m^b + \beta_4 m^c K_{ca} m^b \nabla_b m^a]
\end{aligned} \tag{1}$$

In (1), we contract indices with g^{ab} , and $\nabla \times \hat{m} = \epsilon^a_b \nabla_a m^b$, where ϵ_{ab} is the covariant antisymmetric tensor.² Note that F_1 reduces to the hexatic membrane free energy when $\kappa_1 = \kappa_2 = 0$ and $k_1 = k_2 = k_A$, since even on a curved surface $(\nabla \cdot \hat{m})^2 + (\nabla \times \hat{m})^2 = \nabla_a m^b \nabla^a m_b$. Following Nelson and Pelcovits [2], we find it convenient to take $k_2 > k_1$ without loss of generality and rewrite the terms involving only \hat{m} and the metric as $k_2 \nabla_a m^b \nabla^a m_b + \bar{k} (\nabla \cdot \hat{m})^2$, where $\bar{k} = k_2 - k_1$.

As we alluded to above, for large stiffnesses κ, k_A , hexatic membranes are governed by a line of fixed points in the $\kappa^{-1} - k_A^{-1}$ plane [3]. Our first simplification for tilt is to study the stability of this line against anisotropy by working to first order in the anisotropic couplings. We further simplify our job by dropping the terms of F_2 in (1). This truncation is mathematically consistent because F_1 is a complete list of terms with the ‘‘nematic’’ $\hat{m} \rightarrow -\hat{m}$ symmetry. We do not know of an experimental system with this symmetry. One can imagine a membrane made of rod-like molecules that lie parallel to the local tangent plane, or perhaps more realistically a membrane with stiff rod-like molecules aligned along the local normal but with a rectangular cross-section. In any case, this free energy is the simplest membrane model with in-plane order and anisotropy; we expect it to be qualitatively similar to the case of real tilt.

We study the long-distance behavior of our model with the Wilson momentum-shell renormalization group [22]. The first step of this procedure is to decompose the fields into slowly varying and rapidly varying parts $\vec{x} = \vec{x}_0 + \vec{x}_1, \hat{m} = \vec{m}_0 + \vec{m}_1$, where \vec{x}_0, \vec{m}_0 have Fourier modes with wavevector k satisfying $|k| < \Lambda/b$, and \vec{x}_1, \vec{m}_1 have Fourier modes in a shell $\Lambda/b < |k| < \Lambda$. $\Lambda = 2\pi/a$ is the wavevector cutoff corresponding to the short-distance cutoff a , and b is a number slightly greater than one. The next step is to coarse-grain the system by tracing over the fast modes in the partition function. We write the partition function for the membrane as a path integral over the shape and orientational degrees of freedom [1]. Since membranes are stiff but not completely rigid, we work to first order in T/κ .³ This approximation amounts to integrating out the fast modes in the Gaussian approximation. Finally, we rescale the distances to restore the cutoff to its original value and obtain the recursion relations for the effective couplings.

² In (1) we have corrected a redundancy in [14]; we note here that the α_1 and α_2 terms of [14] differ by a total derivative.

³ Since we work in the stiff regime, we ignore defects in the in-plane order. These will not qualitatively change our result (but see [5]).

To carry out these calculations, we must choose coordinates. The metric $g_{ab} = \partial_a \vec{x} \cdot \partial_b \vec{x}$ at each point of a two dimensional surface is a symmetric 2×2 matrix and thus has three independent degrees of freedom. Two of these are removed by the two independent coordinate degrees of freedom, leaving one physical degree of freedom. In fact, we can always find local coordinates in which the metric is a spatially dependent conformal factor times the trivial metric [7]:

$$g_{ab}(\xi) = \rho(\xi) \delta_{ab}. \quad (2)$$

Conformal gauge is the two dimensional analog of arc-length parameterization of a curve in space.

The tensor fields associated with a surface have a compact form in conformal gauge. For example, the mean curvature is $\hat{n} \cdot \nabla^2 \vec{x} = \rho^{-1} \hat{n} \cdot \partial^2 \vec{x}$, the change of basis matrix is $e_a^\alpha = \sqrt{\rho} \delta_{a\alpha}$, and the spin connection is $\Omega_a = \frac{1}{2} \epsilon_{ab} \partial_b \log \rho$ [7].

For purposes of illustration we first consider the *fluid* membrane (no in-plane order) in conformal gauge, as discussed by Polyakov [21]. Since ρ depends on \vec{x} , the fluid membrane free energy is a complicated nonlinear functional of \vec{x} and not in the most convenient form to carry out the renormalization group procedure. To treat ρ as an independent field, we introduce a delta function constraint and then enforce the constraint by introducing some Lagrange multiplier fields [21]:

$$\begin{aligned} Z &= \int [d\vec{x}] \exp \left[-\frac{\kappa}{2T} \int d^2 \xi \rho^{-1} (\partial^2 \vec{x})^2 \right] \\ &= \int [d\vec{x}] [d\rho] \delta[\partial_a \vec{x} \cdot \partial_b \vec{x} - \rho \delta_{ab}] \exp \left[-\frac{\kappa}{2T} \int d^2 \xi \rho^{-1} (\partial^2 \vec{x})^2 \right] \\ &= \int [d\vec{x}] [d\rho] [d\lambda^{ab}] e^{-F} \end{aligned} \quad (3)$$

where

$$F = \frac{\kappa}{2T} \int d^2 \xi [\rho^{-1} (\partial^2 \vec{x})^2 + i\lambda^{ab} (\partial_a \vec{x} \cdot \partial_b \vec{x} - \rho \delta_{ab})]. \quad (4)$$

To complete the specification of the partition function, we must define the functional measure. There are various factors that arise from coordinate invariance [23]. We simply note that the geometrical measure factors will not enter our calculations to $O(T/\kappa)$, and the Liouville counterterm will not enter the recursion relations for the bending stiffnesses. A further advantage of conformal gauge over Monge gauge is that these conformal gauge measure factors have been well-studied [23], making conformal gauge the better choice for calculations beyond $O(T/\kappa)$.

The Wilson renormalization group procedure requires the most general low order expression consistent with all the symmetries; (4) is not obviously of this form. The Lagrange multiplier field enters in a very specific way. More precisely, since the renormalization group is an iterative procedure, we must be sure that after eliminating short wavelength modes the long-wavelength effective free energy differs from the original only by the values of the couplings. If *e.g.* a λ^2 term were generated, then the delta function constraint would be softened to a Gaussian and we could not define the recursion relations. Such terms do in fact appear, but always suppressed by powers of the short-distance cutoff.

The extra fields λ^{ab} and ρ were supposedly introduced to simplify the calculation; it is natural to ask why the extra fields do not lead to more complexity. The continued sharpness of the delta function during coarse-graining leads to a technical simplification. To see this, we recall Polyakov's trick to diagonalize the fluctuation part of the free energy functional [21]. He decomposes the rapidly varying part λ_1^{ab} of the Lagrange multiplier field λ^{ab} into a traceless and transverse trace part taking

$$\lambda_1^{ab} = \partial_a f_{1b} + \partial_b f_{1a} - \delta_{ab} \partial_c f_{1c} + \left(\delta_{ab} - \frac{\partial_a \partial_b}{\partial^2} \right) \zeta_1. \quad (5)$$

Using this decomposition, we expand the free energy to quadratic order in the fast fields and split its quadratic part F^{II} into a free part and a part to be treated as a perturbation $F^{II} = F_A + F_B$,

$$F_A = \frac{\kappa}{2T} \int d^2 \xi [\rho_0^{-1} (\partial^2 \vec{x}_1)^2 - 2i (\partial^2 f_{1a}) (\partial_a \vec{x}_0) \cdot \vec{x}_1 - i \zeta_1 \rho_1], \quad (6)$$

$$F_B = \frac{\kappa}{2T} \int d^2 \xi [i \lambda_0^{ab} (\partial_a \vec{x}_1) \cdot (\partial_b \vec{x}_1) - 2 \rho_1 \rho_0^{-2} (\partial^2 \vec{x}_0) \cdot (\partial^2 \vec{x}_1) + \rho_1^2 \rho_0^{-3} (\partial^2 \vec{x}_0)^2 - 2i \lambda_1^{ab} (\partial_a \partial_b \vec{x}_0) \cdot \vec{x}_1].$$

In the diagrammatic expansion of the effective free energy, F_A determines the propagators and F_B determines the vertices. Since there is no ζ_1^2 term in (6), the ρ_1 propagator is zero: $\langle \rho_1 \rho_1 \rangle = 0$. Many diagrams that otherwise would have contributed are thus zero. Also, the bending free energy for a fluid membrane is independent of derivatives of ρ , so we can treat ρ_0 as a constant. Thus the conformal factor ρ does not play much of a role in the calculations.

Turning to our model, we introduce another Lagrange multiplier μ to enforce $\hat{m}^2 = 1$. Introducing μ is more convenient than parametrizing \hat{m} as $m^\alpha = (\cos \theta, \sin \theta)^\alpha$, since all the

$$\begin{aligned}
& + \frac{1}{2T} \left[\kappa_2 - \frac{T\kappa_2}{8\pi\kappa} \log b \right] \int d^2\xi \rho_0^{-1} m_0^\alpha m_0^\beta m_0^\gamma m_0^\delta (\partial_\alpha \partial_\beta \vec{x}_0) \cdot (\partial_\gamma \partial_\delta \vec{x}_0) \\
& + \frac{1}{2T} \int d^2\xi i\lambda_0^{ab} \left[\partial_a \vec{x}_0 \cdot \partial_b \vec{x}_0 - \rho_0 \left(\delta_{ab} + \frac{T}{4\pi\kappa} (-\delta_{ab} + \delta_{ab} \frac{2\kappa_1 + \kappa_2}{8\kappa} - m_{0a} m_{0b} \frac{\kappa_1 + \kappa_2}{2\kappa}) \right) \log b \right] \\
& + \frac{k_2}{2T} \int d^2\xi (\partial_\alpha m_0^\beta)^2 \left(1 + \frac{\bar{k}T}{2\pi k_2} \log b \right) + \frac{\bar{k}}{2T} \int d^2\xi (\partial_\alpha m_0^\alpha)^2 \left(1 - \frac{T}{\pi k_2} \log b \right). \quad (11)
\end{aligned}$$

In deriving (11), we have used $\partial_a \vec{x}_0 \cdot \partial_b \vec{x}_0 = \rho_0 \delta_{ab} + O(T/\kappa)$ and $\bar{m}_0^2 = 1 + O(T/\kappa)$.

The purpose of quoting the long expression (11) is to point out an unexpected term: the term with $\lambda^{ab} m_{0a} m_{0b}$ did not appear in the original free energy. This term spoils the conformal gauge condition $\partial_a \vec{x}_0 \cdot \partial_b \vec{x}_0 = \rho_0 \delta_{ab}$ for the slow fields, but it does not spoil the renormalization group calculation. We simply integrate over λ_0^{ab} to get a modified delta function relating $\partial_a \vec{x}_0 \cdot \partial_b \vec{x}_0$, $\rho_0 \delta_{ab}$, and $m_{0a} m_{0b}$. Using the delta function to integrate over ρ_0 , we obtain a free energy of the same form as (1) but with modified coefficients. Substituting the hexatic fixed line relation $k_2 = 4\kappa$, we find the linearized recursion relations $d\vec{g}/d\log b = M\vec{g}$, with

$$M = \frac{1}{4\pi\beta\kappa} \begin{pmatrix} -\frac{3}{\beta\kappa} & \frac{12}{\beta\kappa} & \frac{5}{8} & \frac{17}{2} & \frac{-13}{16} \\ & & -2 & 1 & 1 \\ & & -1 & -2 & -2 \\ & & -\frac{1}{2} & -\frac{7}{2} & \frac{13}{2} \\ & & & & -\frac{3}{2} \end{pmatrix}, \quad (12)$$

$\vec{g} = ((\beta\kappa)^{-1}, (\beta k_2)^{-1}, \beta\bar{k}, \beta\kappa_1, \beta\kappa_2)$, and $\beta = 1/T$. The matrix M has eigenvalues $-3/(4\pi\beta^2\kappa^2)$ and 0 corresponding to the hexatic membrane, and three more negative eigenvalues, $(-9 \pm \sqrt{41})/(16\pi\beta\kappa)$, $-3/(8\pi\beta\kappa)$, corresponding to the anisotropic couplings.

The hexatic fixed line is therefore stable to the anisotropic terms obeying the $\hat{m} \rightarrow -\hat{m}$ symmetry. Beyond a length scale exponential in the stiffness κ/T we expect nematic membranes to behave as hexatic membranes. This is in accord with our intuition that since there is no true long-range in-plane order in this two dimensional system, at very long length scales anisotropy should be irrelevant. The exponential dependence of these length scales on the stiffness means in practice anisotropic membranes will differ from their hexatic counterparts. We remark that Peliti and Prost have also argued that anisotropic membranes should lie in the same universality class as hexatic membranes [13]. However our calculation shows that the actual way in which this happens is different from the screening mechanism they proposed.

We have seen that conformal gauge is well-suited for determining the long-distance properties of fluctuating membranes with in-plane order. We can easily include the terms

we ignored to extend our results to more realistic tilted membranes. Our technique is useful for other applications. One example is the critical exponents of a crinkled tilted membrane; we expect these exponents to differ from the hexatic case since the anisotropy is only marginally irrelevant (*cf.* [3]).

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