

Rigid Chiral Membranes

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Statistical ensembles of flexible two-dimensional fluid membranes arise naturally in the description of many physical systems. Typically one encounters such systems in a regime of low tension but high stiffness against bending, which is just the opposite of the regime described by the Polyakov string. We study a class of couplings between membrane shape and in-plane order which break 3-space parity invariance. Remarkably there is only *one* such allowed coupling (up to boundary terms); this term will be present for any lipid bilayer composed of tilted chiral molecules. We calculate the renormalization-group behavior of this relevant coupling in a simplified model and show how thermal fluctuations effectively reduce it in the infrared.

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Statistical ensembles of random geometrical shapes pervade theoretical physics. Initially one-dimensional curves in space were most thoroughly studied due to their ease of description and the many applications of such ensembles to the conformation and dynamics of polymers, but today ensembles of two-dimensional membranes are at least as important. Physical realizations of such surfaces include lipid bilayers and surfactant films, which spontaneously self-assemble from amphiphilic molecules in solution or at fluid interfaces (for reviews see [1]). More speculative applications as diverse as the 3D Ising model and other 3D phase transitions, cosmic strings, flux tubes in QCD, and models of elementary particles all rest upon the key property that the important physical degrees of freedom in these problems are *shapes* with no preferred choice of coordinates [2]. The condition that the coordinate choice be immaterial greatly constrains the possible forms of the statistical weights in these systems, leading to very few independent couplings and hence physically simple models.

In this Letter we will study a model appropriate for the description of tilted lipid bilayers (e.g., the lamellar L_{β^*} phases of lyotropics or S_C^* phases of smectics), though we think the analysis is potentially interesting in other contexts as well. Bilayers are typically *rigid*, that is their resistance to bending is characterized by a dimensionless quantity $\bar{\kappa}_0 \equiv \kappa_0/k_B T$ (see below) which is greater than 1. Accordingly, we will carry out a perturbative expansion about $\bar{\kappa}_0 \rightarrow \infty$, the high-stiffness, low-temperature limit. Bilayers with free boundary conditions also typically adjust themselves to zero effective surface tension [3], and we will also work in this limit [4].

We can summarize our logic as follows (further details will appear elsewhere). At length scales far longer than the size of the constituent molecules a continuum description becomes appropriate. In our nearly flat regime the important degrees of freedom are the elastic (or “Goldstone”) modes corresponding to transverse undulations as well as director fluctuations if in-plane order develops. We will find only a few allowed couplings of these modes, as we expect in any elastic system at very long wave-

lengths: The system forgets most of the details about its constituents. Somewhat surprisingly, however, the membranes with in-plane order *can* remember at long scales whether or not their constituent molecules are *chiral*, even though this chirality is often a rather subtle property of the amphiphiles. Gross chiral behavior has long been seen in monolayers, where one gets pinwheel domains [5,6] as well as in flexible membranes which can form helical ribbons [7] whose sense depends on the constituent molecules’ handedness [8]. We will see that this memory follows from the existence of an allowed parity-violating term in the free energy, which couples in-plane order to shape [9]. When in-plane order is thermally destroyed the theory admits no such terms; the system cannot express the chirality of its constituents even if present. Indeed, experimentally a loss of gross chiral structure does seem to accompany the chain-melting transition, and chiral structures do not form at all above this temperature [8,10].

Thermal fluctuations are often important in membranes at room temperature where $\bar{\kappa}_0 \lesssim 40$ is not too close to infinity. It is well known that without in-plane order such fluctuations induce a logarithmically scale-dependent softening of the effective stiffness $\bar{\kappa}$ in the infrared [11]. What we will show is that the unique bulk chiral coupling, if present, similarly suffers a logarithmic renormalization. The renormalization-group behavior of this chiral term seems not to have been studied before. While some of our analysis will reproduce others’ results, we hope that our unified treatment of allowed free energy terms will clarify some of the important symmetries; we have also tried to clear up several subtleties in the fluctuation problem [12]. Finally, the renormalization of chirality will affect the average shapes taken on by membranes, since the chiral coupling helps determine those shapes. We will derive an anomalous scaling relation for the radius of helical ribbons as chirality varies which departs from the mean-field formula [13] and may be experimentally testable [14].

We begin by considering in greater detail the elastic modes of our system. A membrane made of molecules

which slip around each other in some average 2D locus with no in-plane order at all can be described just by specifying a mathematical 2-surface $\mathbf{x}(\xi)$ in 3-space. We can choose coordinates ξ^i , $i=1,2$, for our surface, but we must remember that this choice is arbitrary. The choice of a flat reference surface breaks one transverse translation symmetry, giving one "undulation" mode. We can visualize this mode as height fluctuations from a horizontal plane. At high temperatures this is the only soft mode we expect. At lower temperatures in-plane order can develop, leading to additional soft modes. While experiments do not seem to see full crystalline in-plane order in (hydrated, unpolymerized) membranes [9], and such an order is disfavored on theoretical grounds [15], still orientational quasi-long-range order can survive, giving rise to an *angular* elastic mode. We will consider only temperatures well within the ordered phase. Hence we can visualize the corresponding order parameter as a unit vector field $\mathbf{m}(\xi)$ *tangent* to our 2-surface.

For hexatic in-plane order \mathbf{m} is defined only up to rotations by $2\pi/6$. When the constituent molecules are tilted from the surface normal they break rotation invariance completely, so \mathbf{m} has no periodic identifications, but a new subtlety arises instead. To define $\mathbf{m}(\xi)$ given a configuration of molecules, we must project the molecule axis down to the midplane of the membrane. Since the two sides of the bilayers are equivalent, the overall sign of $\mathbf{m}(\xi)$ is not fixed until we *choose* one side, or equivalently, one of the two normal vectors $\hat{\mathbf{n}}(\xi)$; with this choice we may use the convention where the projection of the molecule on the outward-facing layer defines \mathbf{m} . Since there is no preferred choice of normal, each term of our free energy must be unchanged under the substitutions

$$\mathbf{m} \rightarrow -\mathbf{m}, \hat{\mathbf{n}} \rightarrow -\hat{\mathbf{n}} \quad [16].$$

Our surface gets a few standard tensor fields. It inherits a metric $g_{ij} = \partial_i \mathbf{x} \cdot \partial_j \mathbf{x}$, a corresponding covariant derivative ∇ , and a volume form $d^2 \xi \sqrt{g}$. Once we choose a normal $\hat{\mathbf{n}}$, we also get a second fundamental form $K_{ij} \equiv \hat{\mathbf{n}} \cdot \nabla_i \partial_j \mathbf{x}$ and an alternating tensor

$$\varepsilon_{ij} \equiv \varepsilon_{ija} \hat{\mathbf{n}}^a = \sqrt{g} \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}_{ij}.$$

Here $\varepsilon_{ija} \equiv \partial_i x^b \partial_j x^c \varepsilon_{bca}$ where ε_{bca} is the usual alternating symbol, $\varepsilon_{123} = +1$. Note that ε_{ij} and K_{ij} change sign under the change of normal $\hat{\mathbf{n}} \rightarrow -\hat{\mathbf{n}}$, $\mathbf{m} \rightarrow -\mathbf{m}$ discussed above.

Let us enumerate all allowed free-energy terms. We assume all nonlocal interactions are absent or screened to a length scale shorter than the scale of interest. Then we simply seek all local terms in $\mathbf{x}(\xi)$ and $\mathbf{m}(\xi)$ with the above symmetries, relevant or marginal in the low-temperature or high-stiffness expansion. We will not seek to derive such terms from some three-dimensional liquid-crystal free energy, but instead simply construct them directly from the ingredients listed above. The relevance of an operator about the weakly fluctuating $\bar{\kappa}_0 \rightarrow \infty$ fixed point depends on its naive engineering dimension. The only short-distance cutoff in the problem is the characteristic size Λ^{-1} of the constituents. Λ^{-1} is a 3-space length, not a parameter-space length as in ordinary 2D quantum field theory, so when counting the dimensions of operators we should consider only their behavior under rescaling \mathbf{x} (not ξ^a) [17]. $\hat{\mathbf{n}}$ and \mathbf{m} are unit vectors and hence dimensionless.

We can now list all the independent relevant and marginal terms allowed in the free energy:

$$H_1 = \frac{\kappa_0}{2} \int d^2 \xi \sqrt{g} [(K^i_i)^2 + \gamma_1 (\nabla_i m^j)(\nabla^i m_j) + \gamma_2 (\mathbf{m} \cdot \nabla m^i)(\mathbf{m} \cdot \nabla m_i)], \quad (1)$$

$$H_2 = \frac{\kappa_0}{2} \int d^2 \xi \sqrt{g} [a_1 \mathbf{m} \cdot K \cdot K \cdot \mathbf{m} + a_2 (\mathbf{m} \cdot K \cdot \mathbf{m})(K^i_i) + a_3 (\mathbf{m} \cdot K \cdot \mathbf{m})^2 + \beta_1 (\mathbf{m} \cdot K \cdot \mathbf{m})(\nabla \cdot \mathbf{m}) + \beta_2 (K^i_i) \nabla \cdot \mathbf{m} + \beta_3 K^i_j \nabla_i m^j + \beta_4 m^i K_{il} m^j \nabla_j m^l], \quad (2)$$

$$H_* = \frac{c_0^*}{2} \int d^2 \xi \sqrt{g} m^i \varepsilon_i^j K_{jl} m^l. \quad (3)$$

In these formulas we raise and lower i, j indices using g_{ij} . Most of these terms have already been discussed by Helfrich and Prost [13]; see also [18]. H_1 is the usual Canham-Helfrich elastic energy [19] with zero tension [3], plus the covariant form of an X - Y model energy. H_2 contains various anisotropies in the bending energy due to the tilt of the constituents. $\gamma_i, \alpha_i, \beta_i$ are dimensionless numbers we take to be ~ 1 , while κ_0 is an energy scale we take much larger than the temperature T (we set Boltzmann's constant = 1). Every term of $H_{1,2}$ (every non-chiral term) is marginal. H_* is the only allowed bulk chiral term; it is relevant. As mentioned earlier, there are *no* available relevant or marginal bulk chiral terms involving only shape (no tilt).

In the enumeration (1)–(3) we have dropped all total

derivatives, including for instance the Gaussian curvature and the covariant form of the $\nabla \times \mathbf{m}$ term [20]. Such terms are important for systems with boundaries (e.g., ribbons) or defects (e.g., rippled phases), but they will not affect our calculation of the renormalization of c_0^* . (Near defects one should also allow \mathbf{m} to vary in magnitude [21].) What is remarkable about (1)–(3) is that while there are a number of allowed couplings, still the list is rather short, especially compared to a 2D scalar at its trivial fixed point, for which infinitely many marginal terms exist.

We will consider a limit in which $\bar{\kappa}_0 \equiv \kappa_0/T$ is large but c_0^*/T is very small compared to the cutoff. This is reasonable since typically chirality is a very minor feature

of the long-chain amphiphiles. Thus, we will carry all our calculations out to lowest nontrivial order in the loop-counting parameter $(\bar{\kappa}_0)^{-1}$ and the chiral coupling c_0^* .

We will calculate the effects of thermal fluctuations in saddle-point (one-loop) approximation. Before we can do the required integrals, however, we must fix certain coordinate-choice redundancies in our description of a surface. Our problem is that we have five variables $\mathbf{x}(\xi)$, $\mathbf{m}(\xi)$ in (1)–(3), while as we have seen only two are truly independent. We must first write \mathbf{x} in terms of one independent field $u(\xi)$. For our purposes, the easiest choice is “Monge gauge,” $\mathbf{x}(\xi) = (\xi^1, \xi^2, u(\xi))$, since nearly flat surfaces can be expanded easily in powers of u . Next we may write \mathbf{m} as $\mathbf{m}(\xi) = \mathbf{e}_1(\xi)\cos\theta(\xi) + \mathbf{e}_2(\xi)\sin\theta(\xi)$, where $\{\mathbf{e}_1, \mathbf{e}_2\}$ are a field of orthonormal tangent vectors to the surface. \mathbf{e}_α depend on $\mathbf{x}(\xi)$, but \mathbf{x} does not fully determine them: We must fix an $O(2)$ gauge freedom. Monge gauge has the pleasant feature that we may choose $\mathbf{e}_\alpha = e_\alpha^i \partial_i \mathbf{x}$, where $e_\alpha^i = \delta_\alpha^i - \frac{1}{2} \partial_i u \partial_\alpha u + O(u^3)$. This expression is not covariant because Monge gauge is not. From now on we will raise and lower indices using the flat metric δ_{ij} ; all g_{ij} factors will be shown explicitly. Similarly, we convert index type using δ_α^i ; all e_α^i factors will be shown explicitly. The fact that we can choose a frame with no $O(u)$ terms will make Monge gauge very convenient.

We now have all the necessary ingredients. When viewing the system on a scale $L \gg \Lambda^{-1}$ we may forget

$$H_* = \frac{c_0^*}{2} \int d^2\xi [u_{12} + \theta(u_{22} - u_{11}) + \theta u_\gamma (u_{11} u_{1\gamma} - u_{22} u_{2\gamma}) - \frac{1}{2} u_\gamma (u_{11} u_{2\gamma} + u_{22} u_{1\gamma}) - 2\theta^2 u_{12}] + \dots, \quad (4)$$

where $u_\gamma \equiv \partial_\gamma u \equiv \partial u / \partial \xi^\gamma$, etc., and the ellipsis denotes terms with at least five fields or at least three θ 's. While H_{eff} is a complicated power series in $\bar{u}, \bar{\theta}$, we can unambiguously determine c_{eff}^* by expanding H_{eff} to first order in \bar{u} and zeroth order in $\bar{\theta}$, since the first term breaks parity and does not appear in any of the total derivative terms dropped in (3). While u_{12} is a total derivative, so that this term seems to vanish, we can still compute it by giving the coupling c_0^* a small fictitious spatial dependence in intermediate stages of the calculation.

We now quote the results of the calculation. Letting $D \equiv (T/4\pi\kappa_0) \ln b^{-1}$, $H_{*,\text{eff}}$ looks like H_* with c_0^* replaced by $c_0^* (1 - 4D/\gamma_1)$. Hence we find that the effect of fluctuations may be summarized by omitting them but replacing c_0^* by c_{eff}^* , where

$$\frac{dc_{\text{eff}}^*(b^{-1})}{d \ln b^{-1}} = \frac{-c_{\text{eff}}^* T}{\pi\kappa_0\gamma_1}. \quad (5)$$

The constants κ_0, γ_1 were defined in (1). Strictly speaking they too should be allowed to run, but as mentioned they will arrive at the fixed line [15] $(\kappa\gamma_1)_{\text{eff}} = 4\kappa_{\text{eff}}$. Solving (5) we find

$$c_{\text{eff}}^*(b^{-1}) = (b^{-1})^{-T/4\pi\kappa_{\text{eff}}} c_0^* < c_0^*, \quad (6)$$

the promised thermal softening.

Equation (6) should not be construed as a temperature

about irrelevant couplings; moreover all the effects of fluctuations on scales between Λ^{-1} and $(b\Lambda)^{-1} \gtrsim \Lambda^{-1}$ may be summarized by readjusting the values of our couplings, since we took care to include all allowed terms and our cutoff respects the symmetries. Our strategy is to expand $u = \bar{u} + h$, $\theta = \bar{\theta} + \zeta$, where $\bar{u}, \bar{\theta}$ have only long-wavelength components, while h, ζ have only wave numbers greater than $b\Lambda$ [22]. We expand the free energy H in h, ζ about $\bar{u}, \bar{\theta}$, find the quadratic terms in h, ζ , and integrate the fast modes of h, ζ in Gaussian approximation to get the one-loop effective “action” $H_{\text{eff}}[\bar{u}, \bar{\theta}]$ (see, e.g., [23]) [24]. Finally we read off the renormalized chiral coupling c_{eff}^* .

To keep our formulas manageable we will truncate our model (1)–(3), retaining only the first two terms of H_1 and of course H_* (for more details see [12]). We expect that the isotropic terms retained will give a good qualitative guide to the effects of fluctuations on chirality. The terms we have omitted will not be generated to the order we are working, since they lack an extra $O(2)$ symmetry of the first two terms; nor can the chiral term induce them to first order in c_0^* .

We want to pick off from H_{eff} the renormalized coefficient of $\frac{1}{2} \int \bar{g}^{1/2} \bar{m}^i \varepsilon_i^j \bar{K}_{jl} \bar{m}^l$. Since this is the only parity-violating term, we pick terms in the expansion of the logarithm which have an odd power of c_0^* . Since we work to lowest order in c_0^* , this means we keep exactly one power, i.e., we simply renormalize the operator H_* . Using Monge gauge we find that

dependence of c_{eff}^* , since in general the bare couplings will have some unknown T dependence. However, we can draw an interesting qualitative conclusion. The diameter R of helical ribbons (and possibly tubules as well) is controlled by a competition between chirality and stiffness. Oversimplifying somewhat by omitting the $\nabla \times \mathbf{m}$ boundary term (its effect is similar to that of H_* [13]), one finds in mean-field theory that [13] $R \propto \kappa_0/c_0^*$. Since c_0^* can be much smaller than the cutoff Λ , R can be very large; indeed experimentally R can be $\sim 0.5 \mu\text{m}$ [7]. Hence thermal fluctuations can significantly modify the mean-field result; we can approximately account for their effects [12] by writing $R \propto \kappa_{\text{eff}}/c_{\text{eff}}^*(\Lambda R)$. Thus (6) says that varying c_0^* for fixed κ_0, γ_1, T we have that the cylinder radius scales not as $R \propto (c_0^*)^{-1}$ but as

$$R \propto (c_0^*)^{-(1+T/4\pi\kappa_{\text{eff}})}. \quad (7)$$

The nice feature of (7) is that it may be possible experimentally to control c_0^* , without changing significantly the other bare parameters, simply by diluting the chiral amphiphiles with similar but achiral analogs [25]. Thus (7) is potentially a rather clean test of renormalization effects in rigid chiral membranes. Whether the range of dilutions admitting helices or tubules will be great enough, and $T/4\pi\kappa_{\text{eff}}$ can be made large enough, to test the scal-

ing law (7) remains to be seen. Even if not, (6) may still be applicable to rippled phases [13].

We have seen how the constituents of a membrane can express their chiral nature at long scales through the development of in-plane tilt order, and how thermal fluctuations can reduce the effective value of the bulk chiral coupling constant and in turn affect the shapes of self-assembled structures. Near the trivial, low-temperature, fixed point the chiral term is relevant, but we have seen how thermal fluctuations reduce its effective dimension. This raises the possibility of a *critical* chiral membrane when this term becomes marginal. Unfortunately this will not happen at weak coupling, so we can say little about this intriguing possibility.

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