Some properties of membranes in nematic solvents

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The fluctuation spectrum of membranes in nematic solvents is altered by the boundary condition imposed on the bulk nematic director by the curved membrane. We discuss some properties of single and multi-membrane systems in nematic solvents, primarily based on the Berreman-de Gennes model. We show that: membranes in nematic solvents are more rigid and less rough than in their isotropic counterparts; have a different Helfrich steric stabilization energy, proportional to $d^{-3}$, and hence a different compression modulus in the lamellar state; and can exhibit phase separation via unbinding during a quench into the nematic state. We also discuss the preparation and possible experimental effects of nematic-mediated surfactant membrane system.

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Short Title: Membranes in nematic solvents
There are many instances in physics where the configuration of a field on a boundary is influenced by the fluctuations of a conjugate field in the adjacent volume. Examples of this are the Casimir effect, where the confinement of the electromagnetic field between conducting plates results in a net attraction \[1\]: related effects in soft condensed matter include a fluctuation-enhanced interaction between inclusions on a membrane \[2\] and the the interaction between surfaces dipped in a structured fluid due to the change in fluctuation spectrum of the fluid \[2,4\]. While these are entropic in origin, there is another class of fluctuation enhancements due to the energy of deforming the bulk field coupled to the boundary field by some anchoring condition. Examples here include: interaction between membrane inclusions due to the strain induced in the membrane \[3\]; the non-analytic contribution to the wetting contact line elasticity due to deformations of the adjacent fluid-air interface \[6\]; and the example which we explore here, a non-analytic contribution to the free energy of a surface in contact with a liquid crystalline solvent \[7,8\].

In this article we explore some of the properties of fluid membranes in contact with nematic liquid crystalline solvents, discussing both entropic and energetic effects. By ‘membrane’ we envision surfactant bilayers arranged in the archetypical structures found in surfactant systems: in this work we focus on lamellar phases. It is well known that surfactants induce varying degrees and strengths of boundary conditions on the nematic director \[1\]. Here we consider the simple natural case when the mesogenic molecules are strongly anchored by the hydrophobic tails of surfactant in the direction along the membrane normal.

Our starting point is the well-known Berreman-de Gennes model \[7,8\], which was introduced to describe the anchoring energy of a liquid crystal on grooved substrates. Here we consider the ‘grooved substrate’ to be thermal undulations of a bilayer surface, and hence consider some of the consequences of the deformation of the nematic director field, induced by these undulations, Fig.1. The principle is this: the equilibrium thermodynamics of a membrane–liquid-crystalline-solvent system includes as fluctuating variables both the solvent and the membrane. If we are interested in properties of the membrane we can ‘integrate out’ the solvent degrees of freedom to find an effective theory for the membrane thermodynamics. To perform such an integration and speak of a renormalized theory of membranes requires a separation of timescales. That is, if we are interested in dynamical properties of the membrane, we must ensure that the solvent fluctuations (in this case, the establishment of a deformed director field in response to a surface undulation) are much faster than the characteristic membrane decay time. However, if we are interested in equilibrium effects, such as fluctuation spectra as would be measured in experiments lasting ‘long’ times, then this procedure is valid. It is with these kinds of experiments in mind that we proceed. Let us first recall the fundamental ideas of the Berreman-de Gennes model.

**Nematic Energy**—Consider a surface with a modulation of wavevector \(q_\perp\), in contact with a nematic solvent, and assume strong homeotropic boundary conditions, \(\delta \hat{n}(r_\perp, z = 0) = -\nabla_\perp u(r_\perp)\), where \(\delta \hat{n}\) is a variation of the nematic director and \(u(r_\perp)\) is a surface displacement along its equilibrium normal \(\hat{z}\); the dimensions in the membrane plane are denoted by \(r_\perp\). The bulk nematic solvent minimizes the Frank elastic energy \(F_E = (1/2)K_F \int d^3\gamma (\nabla \hat{n})^2\), \[3\], in which we make the one constant approximation for the Frank constants \(K_F\). The solution is
\[ \delta \mathbf{n}(r, z) = \int_{q_\perp} iq_\perp u(q_\perp) e^{-iq_\perp \cdot r - |q_\perp| z}, \quad (1) \]

where \( \int_{q_\perp} \equiv \int d^2q_\perp/(2\pi)^2 \) with the limits between an upper cutoff \( 2\pi/a \) and a lower cutoff \( 2\pi/L_\perp \), with \( a \) being a microscopic dimension and \( L_\perp \) the membrane size. Substituting into the Frank energy and integrating over the dimension \( z \) normal to the interface, we find

\[ F_F = \frac{1}{2} K_F \int_{q_\perp} |q_\perp|^3 (1 - e^{-|q_\perp| L_z})|u(q_\perp)|^2 \approx \frac{1}{2} K_F \int_{q_\perp} \frac{q_\perp^4 L_z}{1 + |q_\perp| L_z}|u(q_\perp)|^2, \quad (2) \]

where \( L_z \) is a large distance cutoff which we take below to be \( L_z = \infty \) for an isolated membrane or \( L_z = d \) for a stack of membranes spaced by \( d \). Eq. (3) interpolates between the \( \sim |q_\perp|^3 \) regime for an isolated membrane and the \( \sim |q_\perp|^4 L_z \) long wavelength behavior in a finite system. The second expression in (2) is an alternative approximation which handles properly the large and small \( q \) limits, and is much easier for calculations. We shall mostly use this form of Eq. (2) in this work, since we are primarily concerned with qualitative results. To this must be added the Helfrich energy of the fluctuating membrane,

\[ F_H = \frac{1}{2} \kappa \int d^2r_\perp (\nabla^2 u)^2 + \int d^2r_\perp \bar{\kappa} G, \quad (3) \]

where the Gaussian curvature \( G \) integrates to zero for lamellar systems without topological defects and plays no further role here.

The unusual non-analytic form of Eq. (2) arises from the same considerations as the linear-\(|q|\) elasticity of the air-fluid-solid triple line, where energy is stored in the deformation of the air-fluid surface [6]. From this energy one finds many respects in which membranes in nematic solvents differ from their isotropic counterparts. This behavior could most easily be seen by preparing mixtures of surfactant and thermotropic liquid crystal, with the latter playing the role of an oil (possibly with water or a cosurfactant, as is common in conventional surfactant systems, to select from the zoo of possible phases), and cycling through the solvent’s isotropic-nematic transition temperature. We proceed by briefly describing some of these properties in order of complexity. We only consider the case of membranes without surface tension, where this new term is most important. In the Appendix we present the contribution due to the Casimir effect [4], which is an entropic effect in the correlated fluid mediating the membrane, and show that the main effects of it are a renormalization of the area per surfactant head group, and the bending modulus \( \kappa \).

**Single-membrane properties**—For these properties we take \( L_z = \infty \). The first obvious new effect is a qualitative change in the surface fluctuations. Bilayers are typically rough due to thermal fluctuations, and the combination of a two-dimensional surface fluctuating in three dimensions yields divergent height fluctuations. In a nematic solvent, however, this changes. For example, fluctuations of the surface normal are given by

\[ \langle (\delta \mathbf{n}(r) - \delta \mathbf{n}(0))^2 \rangle = 2k_BT \int_{q_\perp} \frac{1}{K_F |q_\perp| + \kappa q_\perp^2} \simeq k_BT \frac{\log \left[ K_F + 2\pi\kappa/a \right]}{\log \left[ K_F + 2\pi\kappa/r \right]}, \quad (4) \]

where \( a \) is a microscopic cutoff. If we define the correlation length \( \xi_0 \) as that distance along the membrane for which fluctuations in the normal vector \( \mathbf{n} \) orientation are of order 1 [10], we find
\[
\frac{\xi_0}{a} = \frac{e^{2\pi\kappa/(k_B T)}}{1 - \frac{K_F a}{2\pi\kappa} (e^{2\pi\kappa/(k_B T)} - 1)} \]

For \(K_F = 0\) the membrane in the isotropic non-correlated solvent is crumpled at distances larger than \(\xi_0\). For \(K_F \neq 0\), in a nematic solvent, \(\xi_0\) increases rapidly and reaches the system size (\(\infty\)) for \(K_F a/(2\pi\kappa) = (e^{2\pi\kappa/(k_B T)} - 1)^{-1}\). Hence, for \(K_F a/\kappa \gg 1\), in the regime that should be identified with a ‘strong nematic solvent’, the membrane would not be crumpled at all. A typical estimate of the surfactant bilayer bending rigidity is \(\kappa \sim 5 \cdot 10^{-20} \text{ J}\), only slightly larger than the thermal energy at room temperature. Taking a characteristic value for the Frank constant, \(K_F \sim 10^{-11} \text{ J/m}\) and the molecular size \(a \sim 10 \text{ Å}\), one obtains an estimate of order unity and, therefore, both crumpled and flat regimes are accessible for a membrane in a nematic solvent.

Also of interest is the related quantity, the membrane roughness, given by the mean-square height fluctuations:

\[
\langle u(r)^2 \rangle = k_B T \int_{q_\perp} \frac{1}{\kappa q_\perp^4 + K_F |q_\perp|^3} = \frac{k_B T}{2\pi K_F^2} \left\{ K_F L_\perp + \kappa \log \left[ \frac{2\pi\kappa + a K_F}{2\pi\kappa + L_\perp K_F} \right] \right\},
\]

where \(L_\perp\) is the transverse membrane dimension, coming from the lower cutoff in \(q\)-space. As \(K_F \to 0\) we recover, after expansion in powers of \(L_\perp K_F\), the result for a conventional membrane, \(\langle u^2 \rangle \sim L_\perp^{2\zeta_S}\), with a roughness exponent \(\zeta_S = 1\). In a strong nematic solvent we have \(\zeta_S = 1/2\) and, as expected, the membrane is not as rough.

Renormalization of bending modulus—Since the membrane in the nematic solvent is stiffer, we expect the renormalization of \(\kappa\) due to thermal fluctuations [11] to be much reduced. There are two new sources of renormalization for membranes in nematic solvents: entropic, due to the Casimir effect, which we briefly discuss in the appendix; and energetic, due to the Berreman-de Gennes energy. Following the simple procedure outlined by Helfrich [11], we find

\[
\kappa_R = \kappa - \frac{k_B T}{4\pi} \left[ \log \left( \frac{\kappa q_{\max} + K_F}{\kappa q_{\min} + K_F} \right) - \frac{3}{32} \log \left( \frac{L_\perp}{a} \right) \right],
\]

where the first correction is from the Helfrich renormalization and the second term is produced by the Casimir effect in the correlated solvent. In the limit \(K_F L_\perp/\kappa \ll 1\) (isotropic solvent) the Helfrich effect returns to the usual \(\log L_\perp/a\) reduction of the bending rigidity [11,12]. In the nematic solvent with \(K_F \neq 0\) it is replaced by the constant factor \(\log [1 + (\kappa/K_F a)]\), so that the renormalization \(\kappa_R - \kappa\) can be large or small depending on the ‘strength’ of nematic solvent. In addition, there is an increase of \(\kappa\) due to the Casimir effect, also logarithmically divergent with the system size. This result supports the intuitive expectation for the membrane to become more rigid due to the anchoring with the nematic solvent.

Lamellar Phase: Helfrich Interaction—In a lamellar state we take \(L_z = d\) as the cutoff in Eq. (2), since the range of the solvent extends only up to neighboring membranes. We define a ‘strong’ nematic solvent in this context as one for which \(K_F d \gg \kappa\) [which is, in fact, a much weaker condition than \(K_F a \gg \kappa\) in Eq. (3)]. To estimate this we again take \(\kappa \sim 10^{-20} \text{ J}, K_F \sim 10^{-11} \text{ J/m}\), and lamellar spacings ranging from 10 – 1000 Å, yielding \(K_F d/\kappa \sim 1 - 100\). Since
the moduli $\kappa$ can be changed by adding co-surfactant and Frank constants depend on the nematic order parameter, it is quite easy to span the whole range from weak to strong nematic solvents.

In stacked lamellar phases there are two well-known interactions which stabilize the lamellar phase: electrostatic stabilization and steric interaction. The electrostatic effect \cite{13} yields an interaction energy per unit area of $F/A \sim \frac{\kappa B T}{(L_B d)}$, where $L_B$ is the Bjerrum length and $d$ is the membrane separation. We shall not consider this interaction, exploring instead the more interesting statistical effects of electrostatically screened membranes \cite{14}. In an isotropic solvent these effects lead to the steric stabilization \cite{15}, giving $F/A \sim \frac{(\kappa B T)^2}{(\kappa d^2)}$. This steric interaction arises due to the divergence of the height fluctuations of a single membrane, $\langle |u|^2 \rangle \sim \frac{L}{\zeta_{S\perp}}$; membranes in a stack experience collisions with a characteristic length between collisions governed by $\zeta_S$. Since $\zeta_S$ is positive we expect a steric interaction in the nematic solvent as well, but with a different character than for the standard Helfrich interaction.

Rather than going through a calculation similar to Helfrich’s, we content ourselves here with a scaling-type analysis to obtain the $d$-dependence of the steric stabilization of membranes in a nematic solvent. We first calculate the height fluctuations, using the equipartition theorem for the membrane Hamiltonian, given by Eqs. (2) with $L_z = d$ and (3), and integrating over wave vectors $q_{\perp}$

$$\langle |u(r)|^2 \rangle \approx \frac{k_B T}{4\pi \kappa q_0^2} \frac{1 + [K_\rho d/\kappa](1 + 2q_0 d)}{(1 + [K_\rho d/\kappa])^2},$$  

(8)

where $q_0 = 2\pi/L_{\perp}$ is the low-$q$ cutoff in the membrane plane (we have ignored subdominant terms logarithmic in $q_0$).

Now, the membrane will be sterically stabilized when the height fluctuations are of order the layer spacing, $\langle |u|^2 \rangle \simeq d^2$. This determines $q_0$. For a weak nematic solvent, $K_\rho d \ll \kappa$, we recover the Helfrich result $q_0 d \approx \sqrt{\kappa B T/\kappa}$. For a strong nematic solvent we find

$$(q_0 d)^2 = \frac{k_B T}{4\pi K_\rho d} (1 + 2q_0 d); \quad K_\rho d \gg \kappa.$$  

(9)

Since, typically, $\kappa > k_B T$ we have $K_\rho d \gg k_B T$, in which case $q_0 \approx (k_B T/4\pi K_\rho)^{1/2} d^{-3/2}$. This defines a new length $\xi = q_0^{-1}$, the in-plane correlation length for height fluctuations, or mean distance between collisions. Now we may compute the interaction energy. Crudely, the pressure $P$ due to undulations may be calculated as that of a gas of sterically interacting ‘discs’ of dimension $\xi$. This yields $P \sim k_B T/(d \xi^2)$. Using our result for $\xi$, and realizing that the free energy per unit area is then given by $F/A = Pd$, we arrive at

$$\frac{F}{A} = \frac{(k_B T)^2}{4\pi K_\rho d^3},$$  

(10)

which should be compared with the $d^{-2}$ and $d^{-1}$ behavior of, respectively, the standard Helfrich and electrostatic stabilizations.

The conclusion is that, for electrostatically screened membranes in a nematic solvent, much closer lamellar packing can be achieved, which again confirms the intuitive expectation for more rigid and flat membranes. The resulting
“smectic” lamellar phase will have a different layer compression modulus of the corresponding Landau-Peierls elastic energy

\[ F_{sm} = \frac{1}{2} \int d^3q \left[ Bq_z^2 + (K + K_F)q_\perp^4 \right] |u(q)|^2, \]  

(11)

where \( K = \kappa/d \) is the layer bending modulus in the absence of nematic solvent. In writing this we have ignored the \(|q_\perp|d\) term in the denominator of Eq. (2), since smectic elasticity is concerned with wavelengths much larger than the smectic spacing, \(|q_\perp|d \ll 1\). The compression modulus is \( \tilde{B} = B - C_2^*\chi \), where \( B \) is the bare compression modulus, and \( \tilde{B} \) includes the renormalization due to the coupling between solvent composition and layer spacing \([16]\). \( \tilde{B} \) is given essentially by the pressure of the gas of colliding membranes, which scales as \( \tilde{B} \sim 1/d^\rho \), where \( \rho = 2, 3, 4 \) for electrostatic, standard Helfrich, and nematic-solvent membranes Eq.(10). The compression modulus may be measured by, for example, small angle scattering \([14]\).

More useful information can be extracted from the line shape of the diffusion scattering peak, following from the Landau-Peierls energy \([11]\). As in ordinary smectics, the structure factor behaves as, for example, \( S(0, q_z) \sim (q_z - q_d)^{-2+\eta} \) with the usual Caille exponent \([17]\):

\[ \eta = q_\perp^2 \frac{k_BT}{8\pi \left[ \tilde{B}(K + K_F) \right]^{1/2}}. \]  

(12)

Because \( K_F \gg K \) in strong nematic solvents, \( \eta \) should strongly decrease when the solvent undergoes a nematic transition, leading to a more rapid decay of the structure factor, as noted above. Notice also that the exponent depends on \( \tilde{B} \), which changes its qualitative dependence on \( d \) as one moves into the strong solvent regime. By measuring \( \eta \) and the penetration depth \( \lambda = \sqrt{B/(K + K_F)} \) (which may be extracted by the corrections to the low-\( q \) behavior of \( S(0, q_z) \) \([18]\)) one may determine both \( B \) and \( K + K_F \) by systematic dilution and temperature experiments.

**Unbinding Transition**—Upon lowering the temperature into the nematic phase, the steric repulsion energy \( F/A \) at fixed \( d \) drops by a factor \( f_N/f_I \sim \kappa/(dK_F) \ll 1 \). This dramatic decrease should affect the unbinding of layers \([19,20]\). Within a Flory-type theory, Milner and Roux showed that one can write the free energy per volume \( f \) of a stack of bilayers as \([19]\):

\[ f = \frac{F}{dA} = c \frac{(k_BT)^2}{\kappa \delta^3} \phi^3 - \chi k_BT \phi^2, \]  

(13)

where \( c \) is a numerical constant, \( \delta \) is the bilayer thickness, and \( \chi \) accounts for contributions to the second virial coefficient from other than steric (i.e. typically van der Waals) interactions. Here \( \phi \simeq \delta/d \) is the surfactant volume fraction, and the \( \phi^3 \) term follows from the isotropic-solvent steric interaction. When the solvent undergoes a transition into the nematic phase the \( \phi^3 \) term should be replaced by

\[ c' \frac{k_BT}{K_F \delta^4} \phi^4, \]  

(14)
where $c'$ is another numerical constant. Since $K_F d$ can be much larger than $\kappa$, the result is a smaller repulsion and a smaller preferred interlayer spacing.

At fixed $\phi$ the characteristics of the unbinding transition are: at low $\chi$ a single bound phase exists, while $\chi_c = 3ck_B T \phi/(\kappa \delta^3)$ marks the spinodal line at which the system phase separates into bound ($\phi \neq 0$) and unbound ($\phi = 0$) phases.

For nematic solvents the spinodal line is given by

$$\chi_c = \frac{6c' k_B T}{K_F \delta^2 \phi^2}, \quad (15)$$

which allows for the possibility of, for example, phase separation by quenching the solvent into a nematic state. The resulting dynamics would be very complicated, due to the simultaneous nematic coarsening and phase separation.

**Conclusions**—In summary, a membrane in a nematic solvent should be much stiffer than in an isotropic solvent, leading to its different scaling behavior. Layered systems are in this stiff regime when $K_F d/\kappa \gg 1$, which should be experimentally realizable. The stiffening can be seen in several quantities, such as the correlation length $\xi_0$ for surface normal fluctuations; the roughness exponent $\zeta_S$; and intermembrane interactions, in which the standard Helfrich interaction changes its dependence on the intermembrane spacing $d$. In addition to the effect of the bulk nematic elastic energy, a membrane is also affected by the entropic Casimir-like effect of fluctuations of the coupled director field. We have not yet considered the case of smectic solvents, but it is straightforward to show, by arguments very similar to those used in deriving Eq. (2), that there is a simple renormalization of the membrane elasticity modulus, $\kappa_R = \kappa + 2\sqrt{\kappa_s B_s d}$, where the subscript $s$ refers to the solvent smectic elastic constants; and the Helfrich interaction is the same as in the isotropic case, with $\kappa$ replaced by $\kappa_R$.

Interesting effects are expected in the presence of an external magnetic field $H$, which provides a “mass” for the nematic director fluctuations. While the effect of magnetic field on a membrane is, in principle, the same as for an isotropic solvent, the anisotropy of the diamagnetic susceptibility of a bilayer membrane should be negligibly small compared to that of a bulk nematic liquid crystal. It is straightforward to show that the application of a magnetic field $H$ along the layer normal yields a term in the free energy Eq. (2) (for a single membrane in an infinite system) proportional to $q^2 \sqrt{q^2 \xi^2_\text{m} + \xi^2_\text{m}}$, where $\xi_m = (K_F/(\chi_a H^2))^{1/2}$ is the standard magnetic coherence length. This term further reduces the height fluctuations of the membrane to $\langle u^2 \rangle \sim \log L_\perp /a$, which leads to a very weak steric Helfrich repulsion $F/A \sim 1/d^4$, and suppresses the Landau-Peierls instability in favor of Bragg peaks at the smectic wavevector.

It seems fairly straightforward to perform experimental checks on the described system, by mixing a thermotropic nematic with a small concentration of surfactant, choosing its hydrophobic part to be closely related to mesogenic molecules. Addition of a small amount of water would further stabilize the bilayer membrane structure. All our arguments suggest that it would be very difficult to create curved micellar structures in the nematic solvent (spherical micelles, for example, would have to create a topological defect in the nematic field around them, due to the radial
director anchoring). Instead, we expect the formation of rather flat bilayers even at very low concentrations and dense lamellar and sponge phases with more surfactant/water, with the morphology driven by the elastic energy effects in the mediating nematic solvent.

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Appendix: Casimir Effect—In addition to the energy stored in the director field there is an entropic contribution to the membrane free energy due to the analog of the Casimir effect, calculated for liquid crystals by Ajdari, et al. [3] and Li and Kardar [4]. From the results of Li and Kardar, the entropic contribution to the free energy per unit area of a membrane fluctuating above a flat surface a distance $d$ away is

$$\frac{F}{k_B T A} = -\frac{a_1}{d^2} \left[ 1 + \frac{3A}{2d^2} \int_{q\perp} |u(q\perp)|^2 \right]$$

$$+ \frac{A}{64\pi a^2} \left[ 1 + 4\pi C_1(1) \left( \frac{q}{d} \right)^2 \right] \int_{q\perp} q_{\perp}^2 |u(q\perp)|^2$$

$$+ \frac{3}{128\pi} \left[ \log \left( \frac{L_{\perp}}{a} \right) + 4\pi C_1(2) \right] \int_{q\perp} q_{\perp}^4 |u(q\perp)|^2,$$

where $a_1 = 0.04792$, and $C_1(\zeta)$ is given by Eq. (2.19) of Ref. [4b] and, generally, are very small. To obtain this we have expanded Li and Kardar’s results, which hold for an arbitrary surface, in a gradient expansion in the membrane fluctuation $u(\mathbf{r}_{\perp})$. The term multiplied by $a_1$ is essentially the classic Casimir attraction between the plates, and contributes a small renormalization to the existing attractive interactions, which are typically van der Waals [22]. The $q_{\perp}^4 |u(q_{\perp})|^2$ term renormalizes the surface tension and, since a surfactant system in solution adjusts its area per head group $\Sigma$ to retain equilibrium and satisfy vanishing surface tension [10], leads only to a slight decrease in $\Sigma$. The last term, written in the single-membrane limit $d \to \infty$, renormalizes the membrane bending rigidity $\kappa$ by a relevant logarithmic term (see Eq. (7), where we neglected the small correction $C_1(2) \sim 10^{-3}$), which should be compared with the value $[k_B T/4\pi] \log(L_{\perp}/a)$ found for the renormalization due to thermal fluctuations of membranes in isotropic solvents [11].

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FIG. 1  Director field $\mathbf{n}$ near a fluctuating membrane, which imposes homeotropic anchoring
