Renormalization Group Treatment of Entropic Interactions in Lamellar Phases

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Abstract

Functional renormalization group is applied to study entropic interactions in lamellar phases consisting in a stack of tensionless fluid membranes. This approach allows to recover Helfrich's formula for the steric free energy of a layer per unit area $f(d) = c_\infty (k_B T)^2/\kappa d^2$ (where d is the mean distance between layers) and to estimate analytically the universal coefficient c_∞. Our estimate $c_\infty \simeq .0810$ is much closer to the estimates extracted from Monte Carlo simulations $c_\infty \simeq .106$ than to the original estimate of Helfrich $c_\infty = 3\pi^2/128$.

Under suitable conditions the separation between parallel fluid membranes in lyotropic smectics can be as large as hundreds or thousands of Å's. In such highly diluted lamellar phases it is expected that one approaches a "complete unbinding transition" and that the elastic properties of such lyotropic liquid crystals are dominated by entropic effects due to the thermal fluctuations of the surfactant bilayers. In [1] Helfrich suggested to relate the vertical compressibility B to the second derivative of the entropic free energy (per projected area per membrane), $\Delta f(d)$, for a stack of membranes at average distance d by $B = d\partial^2 \Delta f(d)/\partial d^2$. From a simple scaling argument [1] Δf should scale for large d as

$$\Delta f(d) \;=\; c_\infty \, \frac{(k_B T)^2}{\kappa \; d^2} \tag{1}$$

where κ is the bending rigidity of a single membrane. c_∞ is a dimensionless pure number and should therefore be *universal*. Helfrich [1] proposed the

estimate $c_\infty = 3\pi^2/128 = .231319....$ Numerical estimates, based on Monte Carlo simulations, lead to consistent estimates ($c_\infty \simeq 0.106$ [2] and $c_\infty = 0.101 \pm 0.002$ [3]), which differ qualitatively by a factor $1/2$ from the analytical (but heuristic) estimate of [1].

We show here that a derivation of the steric repulsion law (1), as well as an analytical estimate for the value of the universal constant c_∞, can be obtained with the help of the "non linear functional renormalization group" (NFRG)[1]. This approximate RG procedure was first proposed in [5], and has been more recently developed for the study of the unbinding transitions of interfaces [6] and of membranes [7].

We start from the phenomenological Hamiltonian for a stack of M membranes, whose position over the equilibrium plane of the layers with coordinates $x = (x_i,\ i = 1, 2)$ is labelled by $(z_I(x),\ I = 1, M)$ (as in [2, 3]):

$$H(z) \ = \ \sum_{I=1}^{M} \int d^2x \ \left\{ \frac{\kappa}{2} \left(\Delta_x z_I\right)^2 \ + \ V(z_I - z_{I-1}) \ + \ p \ (z_I - z_{I-1}) \right\} \qquad (2)$$

The repulsive potential V between membranes is taken to be

$$V(z) \ = \ \begin{array}{ll} +\infty & \text{if} \ \ z \leq 0 \\ 0 & \text{if} \ \ z > 0 \end{array} \qquad (3)$$

$p > 0$ is the osmotic pressure and forces the membranes to be at a finite average distance $< d > = < z_I - z_{I-1} >$. In (2) z_{-1} is taken to be equal to $-\infty$. In deriving (2) we have neglected higher order terms in derivatives of z; this is justified as long as the longitudinal correlation length $\xi_\parallel$ (which is of the order of $d\sqrt{\kappa/k_BT}$), is small with respects to the persistence length ξ_κ for a free fluid membrane with the same bending rigidity κ [8, 9]. In addition the tension σ of the membranes vanishes identically (it should gives a $\sigma/2 \ \left(\nabla_x z\right)^2$ in (2)) because the liquid crystal is not subjected to any lateral stress. The reason for this effect is discussed for instance in [9].

The principle of the NFRG calculation is the following (see e.g. [10]). We assume a sharp momentum cut-off Λ on the wave-vectors k of the transverse fluctuations z_I of the layers ($|k| < \Lambda$). Then we compute in the one loop approximation (first order in k_BT) the effective potential per unit of area

[1]The result of our estimate has already appeared in [4].

per layer for an infinite stack of parallel layers with constant interlayers separation d, $\Gamma(d;V)$. Using a discrete Fourier transform in the z direction, we get

$$\Gamma(d;V) = V(d) + \frac{k_B T}{2} \int_{-\pi}^{\pi} \frac{dq}{2\pi} \int_{|k|<\Lambda} \frac{d^2k}{(2\pi)^2} \ln\left(\kappa|k|^4 \ + V''(d) 4 \sin^2\left(\frac{q}{2}\right)\right) \quad (4)$$

Then we look for a change of the Hamiltonian (2), $H \to H_S$, which do not change the effective potential when the rescaling

$$x \ \to \ Sx \quad , \quad z \ \to \ S^{\zeta} z \quad (5)$$

is performed. The assumption that there is no renormalization of κ implies that $\zeta = 1$ and the renormalization group tranformation must act only on the potential V in (2), $V \to V_S$, in such a way that

$$\Gamma(d;V) \ = \ S^{-2} \ \Gamma\left(\frac{d}{S}; V_S\right) \quad (6)$$

The differential recursion equation for V_S is obtained by differentiating (6) with respect to S (infinitesimal rescaling) and by keeping only the terms linear in $k_B T$ (one loop approximation). After some standard calculations we obtain the recursion equation

$$S\frac{\partial V}{\partial S} = 2V(d) + d \ V'(d) + \frac{k_B T}{4\pi} \Lambda^4 \int_{-\pi}^{\pi} \frac{dq}{2\pi} \ln\left(1 + \frac{4}{\kappa\Lambda^4} \sin^2(q/2) V''(d)\right) \quad (7)$$

It is more convenient to work with dimensionless variables

$$V(d) \ = \ \Lambda^2 \ \frac{k_B T}{8\pi} \ W(x) \quad ; \quad d \ = \ \sqrt{\frac{k_B T}{4\pi \ \kappa}} \ \frac{x}{\Lambda} \quad (8)$$

and to write the recursion equation (7) for the derivative $U(x) = W'(x)$ of the potential, since the q integration can be done explicitely. We obtain

$$S\frac{\partial U}{\partial S} \ = \ 3 \ U \ + \ x \ U' \ + \ 2 \ \frac{U''}{U'} \ \left(1 - \frac{1}{\sqrt{1 + 2U'}}\right) \quad (9)$$

This recursion equation is very similar to the one written in [10, 11] for the study of unbinding phenomena for a *single* membrane, and they coincide in the linearized regime (U small). It follows in particular that the results obtained from the approximate recursion relation (9) should become exact near the upper critical dimension $D \to D_{\rm uc} = 4$ (D being the dimension of "hyper membranes" forming a stack in $D+1$ dimensional hyperspace) [10].

Starting from the original potential given by (3) as initial condition for $S = 1$, one expects that as $S \to \infty$ V_S flows to a IR stable fixed point potential V_0^*. This potential is a solution of the fixed point equation (r.h.s. of (7)$= 0$), it must be purely repulsive ($U(x) < 0$ for every $x > 0$), decrease exponentially as $x \to \infty$, and from (9) it must behave for $x \to 0$ as

$$W_0^*(x) \ \sim \ A \ x^{-2} \tag{10}$$

where A is some constant. This IR stable fixed point should describe the unbound phase, and therefore it contains non trivial information about the properties of the lamellar phase at very large interlayer spacing d. Indeed, using (6), the full effective potential density $\Gamma(d)$ must scale for large interlayer spacing ($S \to \infty$) as

$$\begin{aligned} \Gamma(Sd; V) \ &= \ S^{-2} \ \Gamma(d; V_S) \\ &\sim \ S^{-2} \ \Gamma(d; V_0^*) \ = \ S^{-2} \Lambda^2 \frac{k_B T}{8\pi} A x^{-2} \\ &= \ \frac{A}{32\pi^2} \frac{(k_B T)^2}{\kappa} \frac{1}{(Sd)^2} \end{aligned} \tag{11}$$

Thus we have obtained Helfrich's law (1), with the explicit estimate for c_∞, $c_\infty = A/(32\pi^2)$.

We have solved numerically the fixed point equation (r.h.s. of (9)$= 0$) by the technics of [10]. We have indeed found a IR stable repulsive fixed point potential. From the value found for A we estimate

$$c_\infty \ = \ 0.081009 \tag{12}$$

This estimate is smaller by $\sim 20\%$ than the Monte Carlo estimates of [2, 3]. However in our approach we have no problems in extrapolating from a finite

system consisting in a finite number of layers to the infinite case. We have applied the same technics to estimate the strength of steric interactions in 1+1 lamellar phases with tension ($\sigma > 0$), where exact results are available (for instance via the free fermion representation). The estimates from the NFRG differ from the exact results by typically 30%. Therefore one may consider that the agreement between our result and the Monte Carlo simulations is "reasonable".

The applications of the approach described here are twofold:

Theoretical Our renormalization group approach is much less heuristic than the one of [1]. In particular, as quoted above, it becomes exact if the dimensionality D of the layers goes to 4. As for unbinding transitions, it would be extremely interesting to find a theoretical scheme which extend the validity of this RG approach beyond one loop. From the recursion equation (9) one obtains also an attractive fixed point V_1^* with one IR unstable direction, which describes the critical unbinding transition in lamellar phases. More generally, for D close to 4, the general structure of the RG flow and of the fixed points is completely similar to the rich structure found for unbinding transitions [10], with an infinite series of fixed points describing multicritical unbinding transitions. In particular, it is worthwhile to stress that the critical exponents should be the same for pure unbinding and for unbinding in lamellar phases (at least as $D \to 4$).

Experimental Our estimate may be considered as favouring the theoretical estimate $c_\infty \sim 0.1$ with respects to the one of Helfrich [1]. Of course the discrepancy between this estimate and the experimental ones, obtained from measurements of the vertical compressibility B (via scattering experiments) in highly diluted lamellar phases [12, 13] by using the harmonic theory of [14, 15], remains to be understood. Our RG approach should also be applicable to the understanding of crumpling effects in lamellar phases [8, 16], which are important in systems with low rigidity.

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