Lines of Renormalization Group Fixed Points for Fluid and Crystalline Membranes.

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Abstract. - Unbinding transitions of amphiphilic membranes are studied by a renormalization group (RG) approach. Both for fluid and for crystalline membranes, a whole line of RG fixed points is found. Each line consists of two branches: one branch describes complete unbinding, the other branch represents continuous unbinding transitions. The critical behaviour at the unbinding transition exhibits three scaling regimes *A), B),* and *C).* Regime *A)* is characterized by essential singularities, and regime *B)* by parameter-dependent critical exponents. In regime *C),* a discontinuous transition with rather unusual scaling properties is expected by analogy with wetting transitions.

Amphiphilic membranes play an essential role for the organization of biological systems and for the polymorphism of microemulsions [1]. These membranes exhibit two different types of interactions: i) direct interactions, V_{DI}, between planar membrane segments arising from the intermolecular forces such as [2] van der Waals or electrostatic forces; and ii) fluctuation-induced interactions, V_{FL}, as a result of thermally excited bending modes [3, 4].

From a theoretical point of view, the interplay between V_{DI} and V_{FL} represents a challenging problem, since it involves *many length scales.* Indeed, the smallest wavelength of the membrane fluctuations is set by the size, $a \sim 1 \, \text{nm}$, of the amphiphilic molecules, while their largest wavelength is of the order of the membrane dimension, which is typically $\sim (1 \div 10) \, \mu\text{m}$. These fluctuations renormalize the direct interaction, V_{DI}, in a nontrivial way. The renormalized interaction may be attractive or repulsive at large membrane separation corresponding to a bound or an unbound state of the membranes. These two different states are separated by a phase boundary at which the membranes undergo an unbinding transition [4]. Such a transition which resembles a wetting transition [5] can also occur for a membrane interacting with a solid wall.

The bending modes of a single membrane can be described by a variable, $l(x)$, which measures the local distance of the membrane from a reference plane. The associated bending energy depends on the internal structure of the membrane. For a *fluid* membrane, the bending energy per unit area is given by $\frac{1}{2}x(\nabla^2 l)^2$, where x is the bending rigidity and $\nabla^2 l$ is

the leading term of the mean curvature [6]. This form is valid for wave numbers $q \gtrsim 1/\xi_\mathrm{p}$, where [7] $\xi_\mathrm{p} \simeq a \exp[2\pi\varkappa/T]$ is the persistence length of the membrane and T is the temperature. For wave numbers $q \lesssim 1/\xi_\mathrm{p}$, the membrane looses its orientation and starts to crumble [7]. For the fluid (L_α) phase of lipid bilayers and of biological membranes [8, 9], $\varkappa \simeq (10^{-13} \div 10^{-12})$ erg which implies $\xi_\mathrm{p} \simeq 10^6 \div 10^{66}\, a$ at room temperature. Thus, in these systems, ξ_p is large compared to the length scales which are experimentally accessible.

Lipid bilayers often exhibit crystalline (L_β) phases in which the molecules form a lattice and cannot diffuse freely. Free diffusion is also suppressed in polymerized membranes at sufficiently high cross-linking [10]. Then, the variabile l is coupled to the internal displacement fields [11, 12]. At low T, this leads to an effective rigidity [12]

$$\varkappa_\mathrm{eff}(q) \sim K/q^\eta \qquad \text{for small } q\,. \tag{1}$$

A self-consistent calculation gives $\eta = 1$ [12], while a systematic perturbation expansion yields $\eta \simeq 2/3$ [13]

In this paper, the unbinding of membranes with $\eta \geqslant 0$ will be studied. Two nontrivial scaling regimes for the direct interactions, V_DI, are identified both of which are accessible to experiments. The scaling properties within these regimes are analysed by a functional renormalization group (RG) approach which leads to a simple differential recursion relation [5]. In spite of its simplicity, this RG transformation is shown to predict a wide variety of critical behaviour, since it leads to *whole lines* of RG fixed points both for fluid and for crystalline membranes.

To proceed, consider two membrane segments with effective bending rigidities, K_1/q^η and K_2/q^η, which are on average parallel. The effective Hamiltonian for their local separation, $l(\boldsymbol{x})$, or its Fourier transform, $\bar{l}(\boldsymbol{q})$, is taken to be

$$\mathscr{H}\{l\} = \int \frac{\mathrm{d}^2 q}{(2\pi)^2} \frac{1}{2} K q^{4-\eta} |\bar{l}(\boldsymbol{q})|^2 + \int \mathrm{d}^2 x\, V[l(\boldsymbol{x})]\,, \tag{2}$$

with [1] $K = K_1 K_2/(K_1 + K_2)$ and $V(l) \equiv Pl + V_\mathrm{DI}(l)$. The first term in (2) represents the l-dependent part of the bending energies. For fluid membranes, $\eta = 0$ [14], while $2/3 \lesssim \eta \lesssim 1$ for crystalline (or polymerized) membranes. In the following, I will consider the general case with $0 \leqslant \eta < 2$

The second term in (2) represents the free-energy increase resulting from an external pressure, P, and from the direct interaction, V_DI, of the membranes. The direct interaction includes hydration, van der Waals, and electrostatic forces [4, 1]. It has two generic properties: i) it has a hard wall at $l = 0$, *i.e.* $V_\mathrm{DI}(l) = \infty$ for $l < 0$; and ii) it decays to zero for large l, *i.e.* $V_\mathrm{DI}(l = \infty) = 0$

Now, assume that the membranes are in a bound state. Then, their fluctuations are bounded as well, and have an amplitude $\sim \xi_\perp$. Furthermore, their longitudinal extension will be set by another scale, $\xi_\parallel$. Following the scaling arguments developed in ref. [5, 4], the first term in (2) leads to

$$\xi_\perp \sim (T/K)^{1/2} \xi_\parallel^\zeta \qquad \text{with } \zeta = (2 - \eta)/2 \tag{3}$$

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[1] The bending energy per unit area for both membranes is $q^{4-\eta}[K_1|\bar{l}(\boldsymbol{q})|^2 + K_2|\bar{l}_2(\boldsymbol{q})|^2]$. Then, the linear transformation $\bar{l}_1 = \bar{l}_0 - \bar{l}K_2/(K_1 + K_2)$ and $\bar{l}_2 = \bar{l}_0 + \bar{l}K_1/(K_1 + K_2)$ leads to $q^{4-\eta} \cdot [(K_1 + K_2)|\bar{l}_0(q)|^2 + K|\bar{l}(q)|^2]$ with $K = K_1 K_2/(K_1 + K_2)$.

This implies that the bending free energy per unit area scales as

$$V_{\mathrm{FL}}(\xi_\perp) \sim T\,(T/K)^{\tau/2}/\xi_\perp^\tau \qquad \text{with } \tau = 4/(2-\eta)\,. \tag{4}$$

The scaling form (4) can be used in order to *classify* the direct interactions, $V_{\mathrm{DI}}(l)$. Four different scaling regimes must be distinguished [4, 5, 15]. Here I will study those two regimes at $P = 0$ [16] for which the critical behaviour *cannot* be obtained by the superposition ansatz, $V_{\mathrm{DI}} + V_{\mathrm{FL}}$: i) the intermediate-fluctuation (IFL) regime with $|V_{\mathrm{DI}}(l)| \sim V_{\mathrm{FL}}(l)$; and ii) the strong-fluctuation (SFL) regime with $|V_{\mathrm{DI}}(l)| \ll V_{\mathrm{FL}}(l)$ for large l.

For *fluid* membranes with $\tau = 2$, the SFL regime contains i) the usual van der Waals attraction between two membranes which decays as $\sim -1/l^4$, and ii) the van der Waals attraction between a membrane and a solid wall which decays as $\sim -1/l^3$. Two examples for the IFL regime are: i) membranes interacting across a binary solution at its consolute point; then, the critical concentration fluctuations lead to $V_{\mathrm{DI}}(l) \sim -1/l^2$, compare ref. [17]; ii) membranes with adsorbed polymers; for two walls with adsorbed polymers, $V_{\mathrm{DI}}(l) \sim -1/l^2$ has been theoretically predicted [18].

For *crystalline* membranes, the precise value of τ and, therefore, the precise boundary of the SFL regime is presently unknown. For $\tau = 4$, the usual van der Waals attraction $\sim -1/l^4$ between membranes would belong to the IFL regime; for $\tau = 3$, this interaction would still belong to the SFL regime but then, the van der Waals attraction $\sim -1/l^3$ between a membrane and a solid wall would lie in the IFL regime.

The functional renormalization group (RG) developed in ref. [5, 4] can also be applied to the effective Hamiltonian as given by (2). In the infinitesimal rescaling limit with rescaling factor $b \to 1 + \Delta t$, one obtains

$$\partial V/\partial t = 2V + \zeta l\,V' + \frac{1}{2}B\ln[1 + A^2/B)\,V'']\,, \tag{5}$$

where the prime indicates a derivative with respect to l, and the scale factors are given by $B = T/2\pi a^2$ and $A^2 = (T/2\pi K)\,a^{2\zeta}$ (a is the short-distance cut-off). The above differential recursion also applies to wetting transitions in d dimensions, provided $2V$ is replaced by $(d-1)V$ and the scale factors B and A^2 are chosen in an appropriate way [5].

It is convenient to use the dimensionless variables

$$z \equiv \sqrt{2\zeta}l/A \qquad \text{and} \qquad U(z) \equiv 2\zeta V(Az/\sqrt{2\zeta})/B\,. \tag{6}$$

Then, the flow equation becomes

$$\partial U/\partial t = \zeta[\tau U + zU' + \ln(1 + U'')]\,. \tag{7}$$

The fixed points, $U^*(z)$, of finite RG transformation satisfy

$$\tau U^* + z\,\partial U^* + \ln[1 + \partial^2 U^*/\partial z^2] = 0\,. \tag{8}$$

Therefore, the rescaled fixed points depend only on *one* parameter, namely τ. This implies that the unbinding of fluid membranes in $d = 3$ is characterized by the same RG fixed points as wetting in $d = 2$, since $\tau = 2$ in both cases.

The differential equation (8) must be supplemented by appropriate boundary conditions. These are i) the presence of a hard wall at $z = z_w$ with $U^*(z) = \infty$ for $z \leqslant z_w$, and ii) $U^*(z) \approx 0$ for large z. Local analysis of (8) shows that $z_w > 0$ is not possible: any singularity at $z_w > 0$

has the form $U^*(z) \approx (z/z_w - 1) \ln(z - z_w)$ and, thus, does not represent a wall. On the other hand, a wall may be present at $z_w = 0$ with

$$U^*(z) \approx \sigma/z^\tau + \frac{\tau + 2}{\tau} \ln(z) + u_3 + u_4 z^\tau \qquad (\sigma > 0),\qquad (9)$$

for small z with $u_3 = -(\tau + 2)/\tau^2 - \ln[(\tau + 1)\tau\sigma]/\tau$ and $u_4 = (\tau + 2)/2\tau^3(\tau + 1)\sigma$. The leading term in (9) comes from the *linear* terms in (8), and thus involves the arbitrary coefficient σ. However, $1 + \partial^2 U^*/\partial z^2$ must be positive which implies. $\sigma > 0$

The tails of $U^*(z)$ can be determined by linearizing (8). One finds two independent solutions, U_1^* and U_2^*, with

$$U_1^*(z) = \rho/z^\tau[1 + O(1/z^2)] \qquad (10)$$

and

$$U_2^*(z) = \bar\rho z^{\tau - 1} \exp[-z^2/2][1 + O(1/z^2)], \qquad (11)$$

for large z with two independent coefficients, ρ and $\bar\rho$. This indicates that all solutions to (8) decay to zero for large z irrespective of the value for σ in (9). This is indeed confirmed by numerical integration of (8): each value of σ yields a unique solution, $U^*(z|\sigma)$, with its tail characterized by $\rho = \rho(\sigma)$. The function $\rho(\sigma)$ is displayed in fig. 1 for $\tau = 2$.
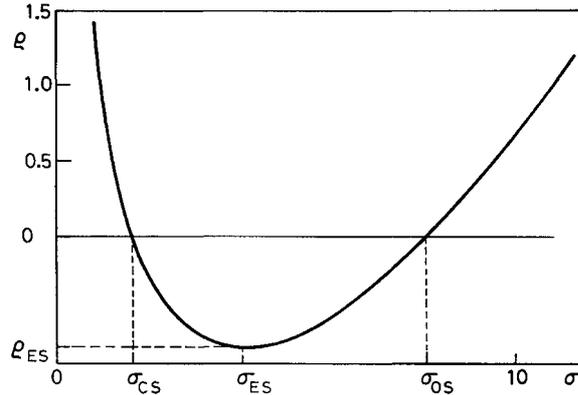
Fig. 1. - The function $\rho(\sigma)$ for $\tau = 2$. The parameter σ determines the wall of the fixed points, see eq. (9); the parameter ρ determines their tails, see eq. (10).

Thus, for each τ, the fixed-point equation (8) together with the boundary condition (9) leads to a *whole line* of RG fixed points, $U^*(z|\sigma)$, parameterized by σ. This line consists of several parts as indicated in fig. 1. For $\sigma > \sigma_{OS}$, the fixed points $U^*(z|\sigma)$ are purely repulsive and decay as ρ/z^τ for large z. For $\sigma = \sigma_{OS}$, U^* is still repulsive but decays as $\sim \exp[-z^2/2]$ for large z since $\rho(\sigma_{OS}) = 0$, see fig. 2a). For $\sigma_{CS} < \sigma < \sigma_{OS}$, the fixed points have an attractive part, since $\rho(\sigma) < 0$ with a power law tail $\sim -|\rho|/z^\tau$. For $\sigma = \sigma_{CS}$, the tail of the attractive part is short-ranged and behaves as $\sim -\exp[z^2/2]$, see fig. 2a). For $\sigma < \sigma_{CS}$ the fixed points are repulsive for large z, but attractive for small z, see fig. 2b).

In order to understand the physical meaning of these fixed points, one must analyse the nature of their eigenperturbations, $f_\lambda(z)$. These perturbations are governed by

$$\zeta\left[\tau + z\frac{\partial}{\partial z} + \left(1 + \frac{\partial^2 U^*}{\partial z^2}\right)^{-1}\frac{\partial^2}{\partial z^2}\right]f_\lambda(z) = \lambda f_\lambda(z), \qquad (12)$$
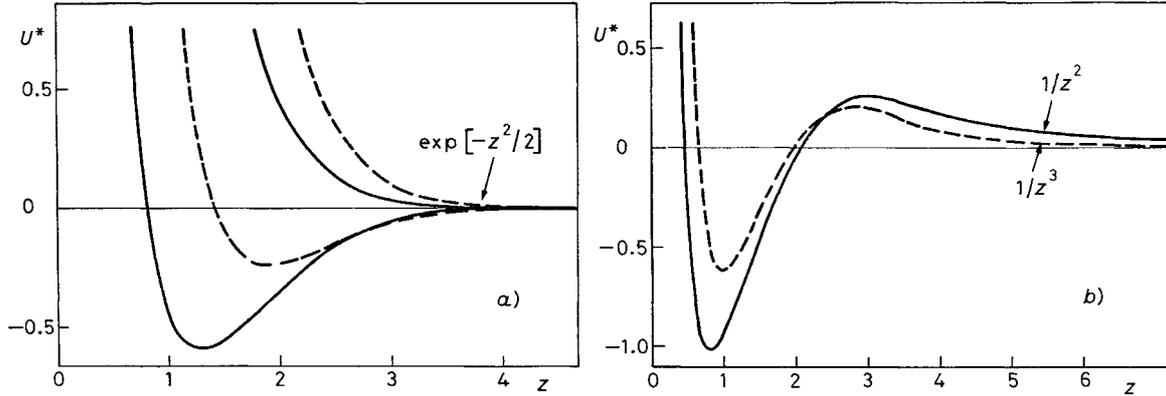
Fig. 2. - a) The fixed points $U^*(z|\sigma_{CS})$ have a minimum, the fixed points $U^*(z|\sigma_{OS})$ are purely repulsive. The full and dashed curves are for $\tau = 2$ and $\tau = 3$, respectively, b) Fixed points $U^*(z|\sigma)$ with $\rho = 3|\rho_{ES}|$ and $\sigma < \sigma_{ES}$ for $\tau = 2$ (full curve) and $\tau = 3$ (dashed curve).

which follows from (8) by linearization. This linear problem can be analysed in detail [19]. For each τ, the RG flow is found to change its character at $\sigma = \sigma_{ES}$: i) for $\sigma > \sigma_{ES}$, there is *no* relevant perturbation with $\lambda > 0$; ii) for $\sigma < \sigma_{ES}$, there is *exactly one* such perturbation with $\lambda \equiv \lambda_1 > 0$

Inspection of (12) shows that λ_1/ζ depends only on τ and, via U^*, on ρ: $\lambda_1/\zeta = \Lambda_1(\tau, \rho)$. The function Λ_1 as determined from (12) behaves as $\Lambda_1(\tau, \rho) \sim (1 - \rho/\rho_{ES})^{1/2}$ for small $\rho - \rho_{ES}$, increases monotonically with ρ, and has the limiting behaviour $\Lambda_1(\tau, \rho) \approx \tau$ for large ρ. The case $\tau = 2$ includes wetting in $d = 2$ for which Λ_1 can be determined exactly. Using the results of ref. [15], one finds

$$\Lambda_1(2, \rho) = \begin{cases} (1 - \rho/\rho_{ES})^{1/2} & \text{for} \quad \rho_{ES} \leqslant \rho \leqslant 3|\rho_{ES}|, \\ 2 & \text{for} \quad 3|\rho_{ES}| \leqslant \rho. \end{cases} \tag{13}$$

From the above analysis, one can identify three subregimes A), B), and C) within the IFL regime. The SFL regime is then viewed as a special case in subregime B). As mentioned, these regimes contain all direct interactions, $V_{DI}(l) \sim U_{DI}(z)$, which decay at least as ρ/z^τ for large z.

Subregime A). - For $\rho < \rho_{ES}$, one has no fixed point and the minimum of U_{DI} becomes deeper and deeper under the RG which implies a *bound* state of the membranes. As $\rho \to \rho_{ES}$ from below, the membranes can undergo a continuous unbinding transition characterized by a vanishing scaling index $\lambda_1 = 0$. This scaling index determines the critical exponent, $\nu_{||}$, for the divergence of the correlation length $\zeta_{||}$ [5,4]: $\nu_{||} = 1/\lambda_1$. Thus, at $\rho = \rho_{ES}$ $\nu_{||} = \infty$ corresponding to an essential singularity of $\xi_{||}$ [19].

Subregime B). - For $\rho_{ES} < \rho < 0$, the RG is governed by two fixed points: i) one of these fixed points has $\sigma > \sigma_{ES}$ and no relevant perturbation. It describes the completely unbounded state of the membranes; ii) the second fixed point has $\sigma < \sigma_{ES}$ and one relevant perturbation with scaling index $\lambda_1 > 0$. It represents a continuous unbinding transition with $\nu_{||} = 1/\lambda_1$ which depends on ζ, τ and ρ.

The case $\rho = 0$ corresponds to the SFL regime: it contains all direct interactions, $U_{DI}(z)$, which decay faster than $1/z^\tau$ for large z. The RG flow within this regime is again governed by two fixed points as has been found previously from functional renormalization with rescaling factor $b > 1$ [5, 4]. For $b = 2$, the numerical values $\lambda_1/\zeta = 0.98$ and $\lambda_1/\zeta = 1.00 \pm 0.03$ were

found for wetting in $d = 2$ [5] and for fluid membranes in $d = 3$[4]. These values for $\tau = 2$ compare very well with presumably exact value $\lambda_1/\zeta = 1$ which follows from (13) for $\rho = 0$ Thus, the SFL regime for *fluid* membranes is characterized by $\nu_\| = 1$

When the RG with $b = 2$ is applied to membranes with $\tau = 3$ ($\zeta = 2/3$) and $\tau = 4$ ($\zeta = 1/2$), one obtains $\lambda_1/\zeta = 1.25$ and $\lambda_1/\zeta = 1.46$, respectively. It follows that the SFL regime for *crystalline* membranes is characterized by a universal value for $\nu_\|$ with $1.20 \lesssim \nu_\| \lesssim 1.37$

It is important to notice that only two fixed points are found for the SFL regime. This implies the absence of tricritical behaviour and, therefore, the *absence of discontinuous unbinding transitions* within the SFL regime: the fluctuations are so strong that they overcome any potential barrier of the direct interaction.

For $\rho > 0$, the RG flow is still governed by two fixed points. Within the RG approach used here, all unbinding transitions with $\rho > 0$ are predicted to have the same character as the continuous transition for $\rho = 0$. However, it follows by analogy with wetting in $d = 2$[15] that this holds only up to a boundary value, $\rho = c(\tau)|\rho_{ES}|$ with $c(2) = 3$

Subregime C). - For $\rho > c(\tau)|\rho_{ES}|$, the unbinding transitions are expected to acquire very unusual properties. Indeed, for wetting in $d = 2$, the transition is discontinuous in that the interfacial energy exhibits a jump, but the correlation length $\xi_\|$ and the moments $\langle (l - \langle l \rangle)^n \rangle$, for sufficiently large n, diverge in a continuous fashion (where $\nu_\| = 1$ independent of ρ) [15, 20]. This reflects the strongly non-Gaussian character of the surface fluctuations: at the unbinding transition, the distribution, $P_*(l)$, which gives the probability to find a certain value for l, has a power law tail $\sim l^{1-2\mu}$ for large l with $2\mu = (1 - \rho/\rho_{ES})^{1/2}$ as follows from the results of ref. [15]. Thus, the distribution $P_*(l)$ is *scale-invariant*. Similar behaviour with $\nu_\| = 1/\zeta\tau = 1/2$ should also occur for fluid and crystalline membranes in $d = 3$ For fluid membranes with $\tau = 2$, I expect that the tail of $P_*(l)$ exhibits the same power law as for wetting in $d = 2$ [21].

In summary, the interplay between fluctuations and interactions of membranes leads to unbinding transitions which can exhibit a wide variety of critical behaviour. These critical phenomena should be accessible to experiments on the cohesion of vesicles and on the swelling of lamellar phases. Particularly interesting behaviour should be observable for the unbinding of fluid membranes which are separated by a (near) critical solution close to its consolute point. Finally, one might speculate about the biological relevance of these phenomena. Indeed, near an unbinding transition, a *small* change in the physical parameters leads to a *large* change in the state of the membranes. Therefore, these transitions represent a possible mechanism by which biological cells could alter their separation in a very efficient way.

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