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Dynamics of crumpling of fluid-like amphiphilic membranes

D Chowdhury, J K Bhattacharjee and A Bhattacharya

Department of Physics, Indian Institute of Technology, Kanpur 208016, India

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Abstract. Fluid-like amphiphilic membranes, in the absence of self-avoidance, are known to be crumpled in equilibrium at all non-vanishing temperatures. We propose a time-dependent Ginzburg-Landau approach to investigate the kinetics of 'disordering' of such a membrane at a non-zero temperature starting from an artificially prepared flat conformation. We carry out a dynamic renormalization group analysis by exploiting some similarities between the crumpling process and the phenomenon of phase separation in binary alloys. The similarities and differences between this crumpling process and the phenomenon of roughening of crystal surfaces heated above the roughening temperature are also pointed out.

Amphiphilic membranes consist of amphiphilic molecules. Each of these amphiphilic molecules consist of a hydrophilic head and a hydrophobic tail; the hydrophilic head likes to be in contact with water whereas the hydrophobic tail tends to minimize contact with water (Tanford 1973, Israelachvili 1985). The amphiphilic membranes may consist of a single monolayer of the amphiphilic molecules (e.g. that at the interface between oil and water in a microemulsion) or a bilayer (e.g. the plasma membrane of a red blood cell). The out-of-plane thermal fluctuations of these membranes (see Petrov and Bivas 1984, Schneider and Webb 1987, Nelson *et al* 1988, Helfrich 1990, Mitov *et al* 1992 for reviews) and the effects of these fluctuations on the equilibrium phases of amphiphile-containing complex fluids (see Gelbert *et al* 1992 for reviews) have received attention over the last few years. One of the interesting phenomena is that of crumpling of these membranes (Helfrich 1985, Peliti and Leibler 1985). Depending on the nature of the in-plane ordering of the membrane, the membrane may crumple at a finite temperature or at the zero temperature (see Lipowsky 1991 for a recent review). In fact, it is now well established that the fluid-like amphiphilic membranes, in the absence of self-avoidance, are crumpled in equilibrium at all non-vanishing temperatures. To our knowledge, the dynamical evolution of the conformations of the amphiphilic membranes far from equilibrium has not been addressed so far in the literature. In this paper we investigate the kinetics of (dis)ordering of fluid-like amphiphilic membranes starting from an initial conformation where the membrane is prepared in a flat phase. We exploit some striking similarities between this phenomenon and the phenomenon of phase separation in binary alloys and carry out a dynamic renormalization group analysis. Finally, we also point out the crucial differences between this phenomenon and that of roughening of crystal surfaces heated to temperatures above the roughening temperature.

We describe the undulations by a single-valued height function $Z(x_1, x_2)$ in the standard solid-on-solid approximation (i.e. we neglect all overhangs). We define the

unit vector

$$\mathbf{n}(x_1, x_2) = (-\partial Z/\partial x_1, -\partial Z/\partial x_2, 1) \quad (1)$$

along the normal to the membrane. The bending energy can be expressed in terms of these normal vectors as a Heisenberg-like Hamiltonian (Nelson et al 1988).

$$F_b = -\tilde{K} \sum \mathbf{n}_i \cdot \mathbf{n}_j \quad (2)$$

where \tilde{K} is a microscopic bending rigidity. From the equilibrium statistical mechanical treatment of the out-of-plane thermal fluctuations of the membranes one finds (de Gennes and Taupin 1982)

$$\langle \mathbf{n}(x) \cdot \mathbf{n}(\mathbf{0}) \rangle \propto \exp(-|x|/\xi_p) \quad (3a)$$

with the 'persistence length'

$$\xi_p \approx a \exp(4\pi K/3k_B T) \quad (3b)$$

where k_B is the Boltzmann constant, T is the temperature and a is a characteristic microscopic length scale. Therefore, fluid-like membranes are crumpled at all non-vanishing temperatures (see Helfrich 1990, Lipowsky 1991 for reviews). One convenient way of characterizing the fluctuations of a membrane is through the orientational correlation function $G(r)$, which is defined as

$$G(r) = \langle [\nabla Z(\mathbf{r}) - \nabla Z(\mathbf{0})]^2 \rangle.$$

In the limit $r \rightarrow \infty$, one obtains

$$G(r) \sim \begin{cases} r^{2-d} & d < 2 \\ \ln r & d = 2 \\ \text{constant} & d > 2. \end{cases}$$

For distances $r > \xi_p$, the sos description is inconsistent. However, at sufficiently low temperatures, ξ_p can be larger than even the system size and, therefore, our sos description will remain valid as long as we confine our attention to length scales smaller than the persistence length. A comparison with the ordering in binary alloys may be useful. At any finite temperature kinetics of the ordering process is monitored to extract the growth law as long as the equilibration process continues. Similarly, we are interested in the process of equilibration of a membrane and one should monitor this equilibration process when the system has not reached the equilibrium state; in equilibrium the size of the crumpled regions is of the order of the persistence length. In other words, we shall monitor the size $R(t)$ of the growing crumpled regions as long as $R(t) \ll \xi_p$.

The Langevin equation describing the relaxation of a conserved order parameter ψ is given in the Fourier (momentum) space by (Bray 1989, 1990)

$$(1/\lambda k^2) (\partial \psi_k / \partial t) = -(\partial H / \partial \psi_{-k}) + \xi_k(t) \quad (4)$$

with

$$\langle \xi_k(t_1) \xi_{-k}(t_2) \rangle = (2T/\lambda k^2) \delta(t_1 - t_2) \quad (5)$$

where λ is a phenomenological rate constant, H is the effective Hamilton (coarse-grained free energy functional), and

$$\psi_k(t) = V^{-1/2} \int d^d r \psi(r, t) \exp(i k \cdot r). \quad (6)$$

Assuming that the cost of the out-of-plane fluctuations of the fluid-like amphiphilic membranes comes entirely from the bending stiffness of the membrane, we obtain the effective Hamiltonian (coarse-grained free energy functional)

$$H = (K/2) \int d^2 x |\nabla^2 Z|^2 \quad (7)$$

where K is the modulus of bending elasticity and the integration is to be carried out over the true area of the membrane. Since we are interested in the crumpling of the membrane the appropriate equation describing the relaxation of the order parameter in this case is

$$\partial(\nabla Z)/\partial t = -M[\delta H/\delta(\nabla Z)] + \xi(x, t) \quad (8)$$

where M is a phenomenological kinetic coefficient and ξ denotes the thermal noise. For simplicity, we assume only Gaussian white noise, i.e.

$$\langle \xi(x, t) \xi(x', t') \rangle = 2k_B T \nabla^2 \delta(x - x') \delta(t - t'). \quad (9)$$

So far as the coefficient M is concerned, we assume $M \propto -\nabla^2$ so that the order parameter is conserved.

There is a close relation between the phenomena of roughening and crumpling. In fact, the effective Hamiltonian for both interfaces and membranes is given by $\int d^2 r |\nabla^p Z|^2$; $p=1$ describes interfaces whereas $p=2$ corresponds with amphiphilic membranes. It is quite straightforward to check that all crumpled membranes are rough with a roughening exponent larger than unity (Lipowsky 1990). The phenomenon of roughening has been investigated in great detail over the last 15 years (see Van Beijren and Nolden 1987 for a review). In recent years the phenomenon of kinetic roughening of growing surfaces has become a very active field of investigation.

Our equation for the temporal evolution is similar to some of the recently proposed equations governing surface growth (Sun *et al* 1989, Wolf and Villain 1990, Golubovic and Bruinsma 1991, Das Sarma and Tamborenea 1991); all the latter models are extensions of what is now known as the KPZ model (Kardar *et al* 1986). However, there are also some crucial differences between our equation of motion for the order parameter and the KPZ-type equations mentioned above; the nonlinearities in our equation follows from the potential itself unlike the sources of nonlinearities in the KPZ-type equations.

Note that because of the special form (7) of the effective Hamiltonian the equation (8) can be written as

$$\partial Z(x, t)/\partial t = (M/k^2)[-(\delta H/\delta Z)] + \xi(x, t). \quad (10)$$

To begin with, let us consider the conserved order parameter because in that case both the analytical calculations as well as numerical computations become much simpler. We can, in that case, use the dynamic renormalization group proposed by Bray (1989, 1990) for the dynamics of phase separation. Besides, if the order parameter is conserved, equation (10) further reduces to the simple form

$$\{\partial Z(x, t)/\partial t\} = -[\delta H/\delta Z] + \xi(x, t). \quad (11)$$

Next let us sketch the main steps in our RG analysis. We make a scale change $k \rightarrow k'$ and time rescaling $t \rightarrow t'$ where

$$k = k'/b \quad t = b^z t'. \quad (12)$$

This rescaling leads to a change of the order parameter $\psi_k(t) \rightarrow \psi'_{k'}(t')$. In the case of phase separation

$$S(k, t) \sim k^{-(2-\eta)} g(k^2 t)$$

where g is the universal scaling function. But, in the case of membranes

$$S(k, t) \sim k^{-(4-\eta)} g(k^2 t). \quad (13)$$

Moreover, following Bray (1989, 1990), we impose the constraint that the structure factor is invariant under the scale transformation (12), i.e.

$$S'(k', t') = S(k, t). \quad (14)$$

This requirement fixes the exponent ζ in the equation

$$\psi'_{k'}(t') = b^{-\zeta} \psi_{k/b}(b^z t')$$

namely

$$\zeta = (4 - \eta)/2. \quad (15)$$

Moreover, the free energy functional scales as

$$H(\{b^\zeta \psi'_{k'}\}) = b^y H(\{\psi'_{k'}\}).$$

Since the order parameter under consideration is a vector

$$y = d - 2. \quad (17)$$

Following Bray's method in the case of a zero-temperature fixed point, we obtain

$$2 - z + 2\zeta - y = 0. \quad (18)$$

Substituting (15) into (18) we obtain

$$(2 - z) + (4 - \eta) - y = 0. \quad (19)$$

But, in the case of membranes

$$(4 - \eta) = d \quad (20)$$

and, hence

$$z = d + 2 - y. \quad (21)$$

Substituting (17) into (21) we obtain

$$z = 4. \quad (22)$$

Now we interpret the main result (22). Following a reverse quench of a fluid-like membrane from the zero temperature to a non-vanishing low temperature the linear size of the region over which equilibration takes place in a time t grows as $t^{1/4}$. In other words, the growth law (22) is identical to that in the case of phase separation in systems with vector order parameter, in spite of the special form (7) of the coarse-grained free energy functional F .

The dynamical process of 'healing' of rough crystal surfaces cooled rapidly below the roughening temperature has been investigated both analytically (Villain 1986) as well as through laboratory experiments (Zuo and Wendelken 1993) and computer simulation (Selke 1987). In the analytical work Villain (1986) used the terrace-ledge-kink model of crystal surfaces and formulated the problem of healing in such a way that, at least at the formal level, it became analogous to the problem of phase separation in binary alloys. Therefore, Villain used the classic Lifshitz and Slyozov approach (Lifshitz and Slyozov 1961) for the latter problem. A step on the crystal surface can emit or absorb particles and holes; smaller terraces decay while larger ones grow. Villain (1986) predicted that the surface is flat on length scales smaller than $R(t) \propto t^{1/3}$. However, it is worth emphasizing that in spite of the superficial similarity between the dynamics of roughening (or the reverse process of healing) of crystal surfaces and crumpling of amphiphilic membranes the physical mechanisms are quite different; there are neither terraces nor steps and kinks on membranes. In this paper we have also exploited some formal similarities between the dynamics of crumpling and phase separation in binary alloys; both these processes are described by similar Langevin equations.

As pointed out earlier, equation (11) is equivalent to the original equation (8) for the dynamics of the order parameter provided the latter is conserved. Therefore, for a numerical test of our prediction it may be easier to solve the partial differential equation (11) rather than the original equation (8). Moreover, our prediction may be tested directly through Monte Carlo simulation.

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