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Short Communication

Stretching and buckling of polymerized membranes: a Monte Carlo study

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Abstract. — We study polymerized (elastic) membranes fluctuating under constrained boundary conditions. We show that the low-temperature, “flat” phase is not described by the classical theory of elasticity. Thermal fluctuations induce important modifications to mechanical laws such as Hooke’s law. We study the approach to the buckled state of membranes, verify the finite-size scaling relations and measure related critical exponents. In the presence of thermal fluctuations the buckled state is qualitatively different from its classical counterpart.

Although the existence of a *finite-temperature* crumpling transition [1] for self-avoiding elastic (i.e. polymerized or solid-like) membranes [2, 3] is still an open theoretical [4, 5] and experimental [6] question, the presence at low enough temperatures of an orientationally ordered phase is not put in doubt. In this “flat” phase *the classical theory of elasticity is believed to break down* due to thermal out-of-plane fluctuations [7, 8]. In particular, if one considers an elastic membrane in the “flat” phase whose excitations are decomposed into transverse undulations, h , and internal “phonon-like” modes, u_1 and u_2 :

$$\mathbf{r}(\boldsymbol{\sigma}) = (\sigma_1 + u_1(\boldsymbol{\sigma}), \sigma_2 + u_2(\boldsymbol{\sigma}), h(\boldsymbol{\sigma})) \quad (1)$$

(where $\boldsymbol{\sigma} = (\sigma_1, \sigma_2)$ are the cartesian coordinates in the reference plane of the membrane, and $\mathbf{r}(\boldsymbol{\sigma})$ is the corresponding position in space), then one can show that thermal fluctuations modify the spectrum of these excitations:

$$\begin{aligned} \langle (h(\boldsymbol{\sigma}) - h(0))^2 \rangle >_0 &\sim \|\boldsymbol{\sigma}\|^{2-\eta_h} \\ \langle (u_a(\boldsymbol{\sigma}) - u_a(0))^2 \rangle >_0 &\sim \|\boldsymbol{\sigma}\|^{\eta_u} \end{aligned} \quad (2)$$

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The critical exponents $\eta_h \equiv \eta$ and η_u , describing the nonclassical behavior of two-point correlation functions (Eq. (2)), can be evaluated in different approximation schemes such as the ε - or $1/d$ -expansions [7, 8]. The subscripts appearing in equation (2) mean that the membrane fluctuates under no constraints. However, one can apply in general some non-zero lateral tension f to the membrane whose behavior will be then modified [8]. Under the action of a positive f (i.e. a stretching) the mean values of the excitations h and u_a and their correlation functions take the form:

$$\begin{aligned} \langle h(\sigma) \rangle_f &= 0; \quad \langle u_a(\sigma) \rangle_f = \delta\zeta\sigma_a, \quad \text{with } \delta\zeta \sim f^{1/\delta'} \\ \langle (h(\sigma) - h(0))^2 \rangle_f &\sim \|\sigma\|^{2-\eta_h} \mathcal{F}_h(\|\sigma\|/\xi_h) \\ \langle (u_a(\sigma) - u_a(0))^2 \rangle_f &\sim \|\sigma\|^{\eta_u} \mathcal{F}_u(\|\sigma\|/\xi_u) + (\delta\zeta)^2 \|\sigma\|^2 \end{aligned} \quad (3)$$

where $\xi_h \sim \xi_u \sim (\delta\zeta)^{-\nu'} \sim f^{\nu'/\delta'}$ and \mathcal{F}_h and \mathcal{F}_u are scaling functions which tend to constant values for $\|\sigma\|$ much larger than the correlation lengths ξ_h and ξ_u . These equations define the new exponents δ' and ν' . The perturbative calculations predict the value of the δ' exponent to be bigger than one: this implies that *the classical Hooke law is also invalid* in the “flat” phase [8]. This is a quite surprising prediction of the theory of elastic membranes. It can in principle be verified experimentally by measuring the spectrum of fluctuations of polymerized vesicles or the deformation of the charged polymerized vesicles in an electric field [9].

One of the main goals of this letter is to check this theoretical prediction in a Monte-Carlo study of a tethered membrane [10, 11]. In such a model membrane the centers of N impenetrable circular beads of diameter a are linked by tethers of length $\ell_0 > a$ to form a triangulated net (of a global hexagonal shape of size L). The configurations of the membrane are altered according to the usual local Monte-Carlo dynamics [11], with energy given by the *bending elasticity* term $E_{\text{bend}} = (\kappa/2) \sum_{i=1}^N (1 - \cos \theta_i)$, where θ_i is the angle between two neighbouring triangles and κ is the bending rigidity. We have studied the response of a membrane to a stretching field $f > 0$ and measured the value of the η exponent: $\eta = 0.75 \pm 0.05$. Other critical exponents characterizing the “flat” phase can be obtained from η through the following scaling laws

$$\begin{aligned} \eta_u &= 2(1 - \eta) \\ \delta' &= (2 - \eta)/\eta; \quad \nu' = 1/\eta \end{aligned} \quad (4)$$

which were derived in the renormalization group approach [7, 8]. In particular, the exponent δ' , which can be viewed as describing the so-called *buckling transition* [8], i.e. the approach to the unconstrained state (as $f \rightarrow 0$) below which the membrane buckles, is found to be $\delta' = 1.7 \pm 0.2$. This means that the classical Hooke’s law is indeed invalid.

Recent independent Monte-Carlo simulations of free tethered membranes [11] have measured the entropic repulsion between the membrane and a impenetrable wall [12]. This repulsive potential varies with the mean distance from the wall, ℓ , as $1/\ell^\tau$. The measured value of the exponent τ was $\tau \approx 3.1 \pm 0.2$. Our result for η provides therefore a good check of the scaling relation [13, 11]: $\tau = 4/(2 - \eta)$. (Indeed, this phenomenological relation leads to $\tau = 3.2 \pm 0.2$.)

When the homogeneous field, f , decreases below zero the membrane will enter a buckled phase, with a non-planar average configuration. The detailed nature of the buckled state is unknown. The buckling phenomenon exists for classical thin shells [14] and one could imagine a buckled state as a membrane fluctuating around its classical (buckled) configuration (which depends on details of the boundary conditions). Thus the simplest question one can ask how different are classical and thermally-excited buckling phenomena? To answer this question we have

extended the range of our simulations to negative values of f and have studied the average configurations in the buckled state of a hexagonally-shaped tethered membrane. Although more quantitative studies are still needed, based on the results of our simulations, we can already say that the average configuration of the buckled state is very different at non-zero temperatures from its classical equivalent, and it does *not* present any “domain-wall” pattern which one could naively expect from the analogy with the ordered phases of magnetic systems [8]. An average buckled configuration has its own pattern with characteristic length which seems to be a function *both* of the size of the membrane (as it is the case for classical buckling) *and* the correlation lengths ξ_h and ξ_u .

We now describe in more detail the results of our numerical simulations and the method we have used. The model considered here was introduced first in reference [10] to verify the existence of the crumpling transition. In fact, in the absence of self-avoiding this model (with $a = 1, \ell_0 = 1.6$) exhibits a crumpling transition at $\kappa = \kappa_{cr} = 0.46 \pm 0.03$ (in units of $k_B T$). In our study of the “flat” phase we have thus considered the values $\kappa > \kappa_{cr}$. We impose in the following two types of boundary conditions:

- (i) the boundary of the net is constrained to vary in the plane $r_3 = 0$. The projected area of the membrane onto this plane, A_3 , is a fluctuating variable;
- (ii) the boundary of the net is fixed on a hexagon of a linear size ζL ($1 < \zeta < 1.6$) in the plane $r_3 = 0$. The projected area has its value fixed at $A_3 = N_\Delta \zeta^2 \sqrt{3}/4$, where $N_\Delta(L)$ is the number of triangles in the net.

In order to study the approach to the buckling transition we have chosen the type-(i) boundary conditions and put $\kappa = 1$ [15]. The linear size of the network varies from $L = 2$ ($N_\Delta = 24$) to $L = 9$ ($N_\Delta = 486$) and we use the finite size scaling relations (valid for L large enough):

$$\begin{aligned} \langle A_3 \rangle_f - \langle A_3 \rangle_0 &= L^{2-1/\nu'} \mathcal{F}(fL^{\delta'/\nu'}) \\ &= L^{2-\eta} \mathcal{F}(fL^{2-\eta}) \end{aligned} \tag{5}$$

Where $\mathcal{F}(x) \sim x^{1/\delta'}$ as $x \rightarrow \infty$, and $\mathcal{F}(x) \sim x$ as $x \rightarrow 0$. We have used here the scaling relations (4), proven in the renormalization group framework. It is convenient to compute $\langle A_3 \rangle_f$ through

$$\langle A_3 \rangle_f = \frac{\langle A_3 e^{fA_3} \rangle_0}{\langle e^{fA_3} \rangle_0} \tag{6}$$

which can be viewed as a generalization of the fluctuation-dissipation theorem. Linearizing these last two equations for small values of f ($f > 0$) leads to the following simple formula

$$\langle A_3^2 \rangle_0 - \langle A_3 \rangle_0^2 \sim L^{4-2\eta} \sim N_\Delta^{2-\eta}, \tag{7}$$

which allows us to measure the critical exponent η .

In our Monte-Carlo simulation we have used a standard Metropolis algorithm, however, we have divided the network into four independent sublattices and we have moved simultaneously all beads belonging to each of the sublattices (with a step $s < 0.2$ and random directions). The simulations were performed on the Cray XMP. We have first checked that the mean projected area $\langle A_3 \rangle_0$ is proportional to N_Δ (the condition for the “flat” phase). The $L = 9$ network has well been equilibrated after the total of $T = 4 \times 10^7$ steps (i.e. $\sim 10^5$ steps/point). This corresponds roughly to 10 hours CPU.

The inset in figure 1 presents the numerical check of the finite-size scaling relation (5). We have presented here the extension of the membrane $\langle A_3 \rangle_f - \langle A_3 \rangle_0$ as the function of the applied tension f (appropriately rescaled). We can see that the relation (5) hold already for sizes

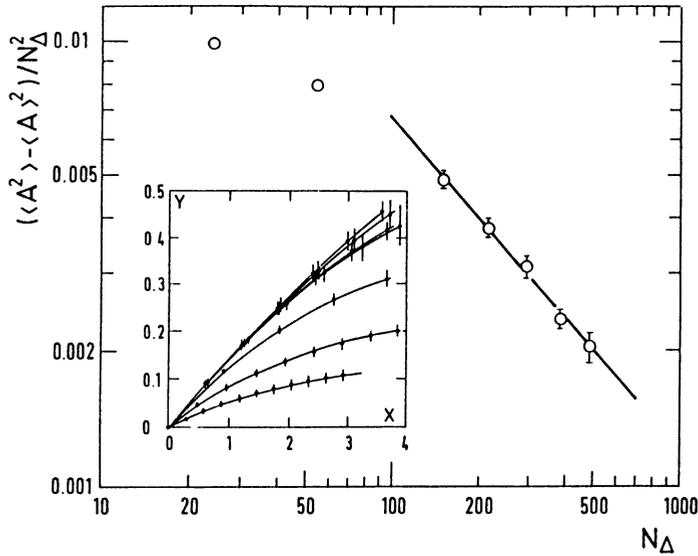


Fig. 1. — Variation of the area fluctuations with the size of the membrane. The inset shows the plot of $Y \equiv (\langle A \rangle_f^{1/2} - \langle A \rangle_0^{1/2}) / N_\Delta^{(1-\eta)/2}$ vs. $X \equiv f N_\Delta^{(2-\eta)/2}$. The curves correspond to linear sizes $L = 3, 4, 6, 7, 8, 9$, and $/10/$ (starting from the lowest one).

$L > 4$. The same value $\eta = 0.75$ used in this scaling plot corresponds also to the one obtained from the curve shown in figure 1, where the fluctuations of the projected area at $f = 0$ are plotted as function of the size of the membrane.

Using scaling relations (4) we obtain from the result $\eta = 0.75 \pm 0.05$ the following critical exponents of the “flat” phase:

$$\begin{aligned} \eta &= 0.75 \pm 0.05 ; \eta_u = 0.5 \pm 0.1 \\ \delta' &= 1.7 \pm 0.2 ; \nu' = 1.3 \pm 0.1 \end{aligned} \quad (8)$$

It is interesting to notice that the values of these exponents characterizing the “flat” phase and the buckling transition are close to the ones obtained through the $1/d$ -expansion [8]. The uncertainties of (8) are estimated by standard statistical methods but should be taken with caution due to the smallness of the simulated systems.

In order to explore the nature of the buckled state we have mainly used the type-(ii) boundary conditions (although we have checked the main conclusions in the fluctuating A_3 case). By varying ζ below the spontaneous ($f = 0$) value ζ_{sp} one modifies the average configurations of the membrane. Figure 2 presents these configurations for a $L = 7$ net for different values of $\kappa/k_B T$ and ζ . In order to represent the three-dimensional configurations of the surface we have chosen to plot the average values of the Gaussian curvature [16]: $K = \sum_\alpha (\pi - \theta_\alpha)$, where the sum is done over all angles α between neighbouring triangles for a given vertex. The Gauss-Bonnet theorem implies that the sum of the values of K over the whole network is zero, we have therefore to concentrate on the *distributions* over the surface of the Gaussian curvature. The averages can be performed over the configurations emerging from different initial conditions after the relaxation time τ , or over various configurations in a single long run ($t \gg \tau$). Typically we have averaged over several thousands of independent configurations.

At low temperatures (or large κ) the phenomenon of buckling is purely mechanical (Fig. 2a): when ζ decreases the membrane buckles into a “bell-like” shape with positive K around the

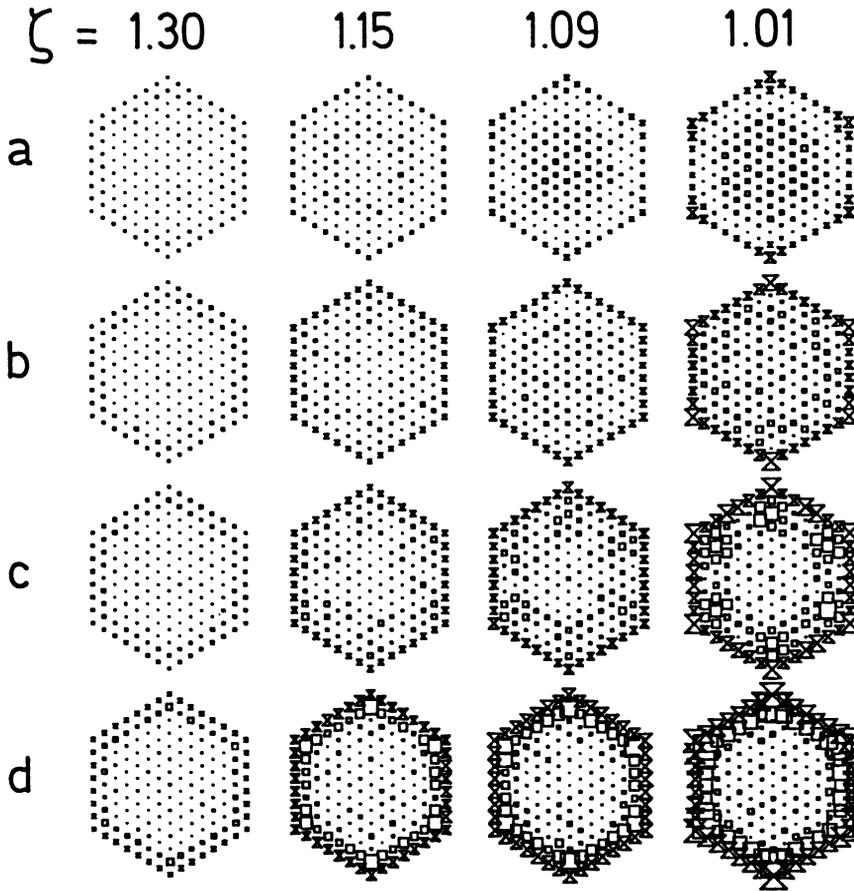


Fig. 2. — Thermally excited buckling of a tethered membrane ($L = 7$): the squares (crosses) correspond to positive (negative) Gaussian curvatures, K , while the size of each symbol is proportional to the magnitude of K . The values of $\kappa/k_B T$ are: (a) 5; (b) 3; (c) 2; (d) 1.

center of the membrane. This shape can easily be calculated in $T \rightarrow 0$ limit by minimizing the curvature energy E_{bend} . If we now increase the temperature (decrease κ) the thermal fluctuations start to modify the buckled state (Fig. 2b). At $\kappa/k_B T \leq 2.7$ we observe a completely new picture: instead of a single region of positive K (in the center) there are six localized regions with $K > 0$ (“spikes”) near the vertices of the hexagon, whereas the center becomes flat (Fig. 2c). Finally, for small κ the membrane is fluctuating wildly, is flat on average and the buckling takes place only on the boundary (Fig. 2d). The characteristic length, Λ , of (between) the curved regions (e.g. the spikes) depends of course on $\kappa/k_B T$, and therefore on the correlation length ξ . Although we could not look at the buckling of membranes bigger than $L = 9$, it seems from the comparison of the buckled states for different sizes (in particular Fig. 2c), that Λ depends also strongly on $L\ell_0$ and is *not* a simple function of κ . It is also remarkable that the *buckling patterns* seem insensitive to the value of ζ for $\zeta < \zeta_{\text{sp}}$; only the amplitude of the curved regions vary with ζ . It would be very interesting to explore more quantitatively the dependence on L and ξ , as well as the role played by the boundary conditions (e.g. the hexagonal shape). For the moment we can only conclude

that there exists a phenomenon of *thermal buckling* qualitatively very different from its mechanical counterpart.

We would like finally to discuss briefly the possible relevance of this work for the experiments in elastic membranes. Good candidates for the study of the stretching and buckling phenomena in thermally excited regime are polymerized vesicles [9] and red blood cells [17]. Although some phenomena resembling the buckling were indeed observed in crosslinked phospholipid bilayers [9], more experimental effort is still needed. In particular, in order to do more quantitative studies on polymerized vesicles one has to find the way to polymerize a whole vesicle (or at least a large part of it). Nature, on the other hand, *did* develop such methods for numerous protein networks [18], for instance for the spectrin networks of red blood cells [17]. It would be thus interesting to continue the quantitative analysis of the flickering behavior of the erythrocytes [19], and concentrate on the elastic properties of the spectrins. If the modifications of the classical-mechanical laws described in this Letter do apply to erythrocyte membranes then one has to reconsider many of the mechanical analyses performed in these systems.

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