

## Crumpling and Buckling Transitions in Polymerized Membranes

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We consider polymerized non-self-avoiding membranes fluctuating under constrained boundary conditions. Such constraints prevent the membranes from being crumpled at high temperature and introduce a tension. Upon lowering the temperature this tension reduces, and vanishes at a transition temperature below which the membrane assumes a buckled state. Buckling transition points, depending on boundary conditions, arrange on a line which terminates at the crumpling transition. We study the critical properties of these transitions by various field-theoretical techniques.

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Polymerized membranes, in contrast to linear chains, may exist in an orientationally ordered flat phase even at nonzero temperature.<sup>1,2</sup> The effective phonon-mediated long-range interactions existing in these two-dimensional objects can indeed prevent thermal fluctuations from destroying the orientational order.<sup>3</sup> Recently developed theoretical models<sup>4,5</sup> of polymerized membranes exhibit, at a finite temperature  $T=T_c$ , a crumpling transition<sup>6</sup> between such an ordered flat phase and a disordered crumpled phase. While below  $T_c$  the radius of gyration  $R_g$  of a fluctuating membrane is proportional to its linear size  $L$ , above the transition a non-self-avoiding free membrane is completely crumpled with  $R_g^2 \propto \ln L$ .<sup>1</sup> In the crumpled phase the correlation length of the membrane normals  $\xi_p(T)$  is finite. This persistence length diverges as one approaches the transition point. Thermal fluctuations which drive the crumpling transition also modify the flat ordered phase; for instance, it has been recently recognized by Aronovitz and Lubensky<sup>7</sup> that the fluctuations lead to a nontrivial renormalization of the elastic constants. In this Letter we extend their work to include the case of more general boundary conditions applied to the membrane.

Real polymerized membranes can be submitted to a large variety of boundary conditions among which the free case, considered until now, is only a particular example. Another interesting example which should be experimentally realizable<sup>8</sup> is the case of closed membranes, i.e., vesicles, for which one can vary the internal pressure. In this Letter we consider a somehow idealized situation of a polymerized membrane which spans a rigid frame. This is a simple generalization of the free boundary case: A variation in the size of the frame can induce a homogeneous tension or tangential pressure acting on the membrane. In particular, we shall see how the constraints introduced by the boundary conditions affect the crumpling transition and how they modify the low-temperature flat phase.

Figure 1 summarizes the main results of the paper.<sup>9</sup> The variable  $\zeta$  measures the ratio of the linear size of the frame to the linear internal size  $L$  of the membrane (at

$T=0$  without tension). The case of *free boundary conditions* is recovered by our allowing the size of the frame to vary freely.  $\zeta$  then takes its spontaneous value  $\zeta_{sp}(T)$ , which is positive for  $T < T_c$ . We can therefore consider  $\zeta_{sp}$  as the order parameter for the crumpling transition. If the transition is continuous,<sup>10</sup> one can introduce the usual critical exponents  $\beta$  and  $\nu$ :  $\zeta_{sp} \sim (T_c - T)^\beta$ ,  $T < T_c$  and  $\xi_p(T) \sim (T - T_c)^{-\nu}$ ,  $T > T_c$ . At the critical point the membrane is a self-similar object with Hausdorff dimension  $D_F$  ( $R_g \sim L^{2/D_F}$ ). With *constrained boundary conditions*, the sides of the membrane are fixed to a square frame of side  $\zeta L$ . Therefore, the region  $|\zeta| > \zeta_{sp}(T)$  corresponds to stretched membranes.

The conjugate field is the tension  $f$ , defined as  $f = 1/L^2 (\partial \Gamma / \partial \zeta)$ , where  $\Gamma$  is the effective potential of the membrane. Therefore,  $f$  and  $T - T_c$  appear as the *two* relevant fields for the crumpling transition. If for a fixed temperature  $T \leq T_c$  the constraint is released ( $f \rightarrow 0$ ), the variable  $\zeta$  approaches  $\zeta_{sp}(T)$  (see Fig. 1) as

$$\zeta - \zeta_{sp} \sim f^{1/\delta}, \quad T < T_c, \quad \zeta \sim f^{1/\delta}, \quad T = T_c. \quad (1)$$

For  $T < T_c$ , inside the coexistence region  $|\zeta| < \zeta_{sp}(T)$ , the polymerized membrane *buckles*. We expect it then to be in a nonhomogeneous mixed thermodynamic state, which is a mixture of pure (flat) phases

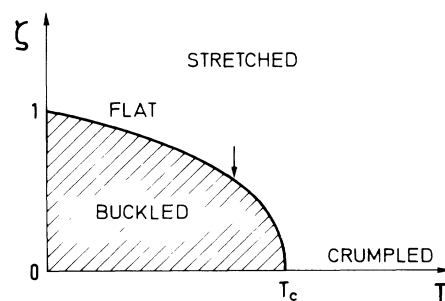


FIG. 1. The  $(\zeta, T)$  plane. The hatched region corresponds to the buckled state, bounded by the line  $\zeta = \zeta_{sp}(T)$ . The buckling transition is obtained by approaching this line for  $T < T_c$ , e.g., along the arrow.

with different orientations. For instance, the buckled membrane could be made out of regions where it is almost flat and unstrained, separated by defects or "domain walls" with high stress. The crossing of the line  $\zeta = \zeta_{sp}(T)$  can be considered as a new transition, which we call *the buckling transition*, with a *single* relevant field. It is characterized by the exponent  $\delta'$  of Eq. (1) and new exponents  $\nu'$ ,  $\eta'$ , and  $\eta_{u'}$  defined by

$$\Gamma_{uu}(q) \sim q^{2+\eta_{u'}}, \quad \Gamma_{hh}(q) \sim q^{4-\eta'}, \quad (f=0, \zeta = \zeta_{sp}), \quad (2a)$$

$$\xi_u \sim \xi_h \sim (\zeta - \zeta_{sp})^{-\nu'}. \quad (2b)$$

The functions  $\Gamma_{uu}^{-1}$  and  $\Gamma_{hh}^{-1}$  are the propagators for the in-plane ( $u$ ) and out-of-plane ( $h$ ) modes<sup>11</sup> which behave as power laws of the distance at  $\zeta = \zeta_{sp}$ , and which decay exponentially with the correlation lengths  $\xi_u$  and  $\xi_h$  for  $\zeta \gtrsim \zeta_{sp}$ . In contrast with Heisenberg spin systems, the "primed" exponents in Eqs. (1) and (2) are governed by a nontrivial fixed point.<sup>12</sup> The fact that  $\eta_{u'}$  is nonzero corresponds to the breakdown of classical elasticity theory<sup>7</sup> for fluctuating polymerized membranes.

We have computed the critical exponents of the crumpling and buckling transitions for a generalized model of  $D$ -dimensional elastic manifolds embedded in  $d$ -dimensional space. The model can be solved for arbitrary  $D$  in the limit  $d = \infty$ , and can be studied for arbitrary  $d$  in

TABLE I. Critical exponents of the buckling transition. The  $\epsilon$ -expansion results hold to first order in  $\epsilon$ .

	$(\delta')^{-1}$	$2 - (\nu')^{-1}$	$\eta'$	$\eta_{u'}$
$d = \infty$				
$2 \leq D \leq 4$	$\frac{D-2}{2}$	$4-D$	0	$4-D$
$D=4-\epsilon$	$1 - \frac{d_c \epsilon}{2d_c + 48}$	$\frac{d_c + 12}{d_c + 24} \epsilon$	$\frac{\epsilon}{2 + d_c/12}$	$\frac{\epsilon}{1 + 24/d_c}$
$d_c = d - D$				

an  $\epsilon$  expansion, with  $\epsilon = 4 - D$ . The upper critical dimension  $D_u$  is equal to 4 for both transitions. The results for the crumpling transition exponents at  $d = \infty$  are given by

$$\beta = \frac{1}{2}, \quad \nu^{-1} = D - 2, \quad D_F = \frac{2D}{4-D}, \quad \delta = \frac{D+2}{D-2}. \quad (3)$$

The results for the buckling transition exponents are summarized in Table I. They satisfy the following scaling laws:  $\eta_{u'} = 4 - D - 2\eta'$ ,  $\delta' = (2 - \eta')\nu'$ .

We now outline the derivation of these results.<sup>13</sup> A Euclidean set of coordinates  $\sigma$  is chosen so that the rest membrane configuration  $\mathbf{X}_0(\sigma)$  at  $T=0$  is  $X_0^i(\sigma) = \delta^i \times \sigma^i$  ( $\alpha = 1, \dots, d; i = 1, \dots, D$ ). The energy of the configuration  $\mathbf{X} = \mathbf{X}(\sigma)$  is given by<sup>4,5</sup>

$$\mathcal{H}(\mathbf{X}) = \int d^D \sigma \left[ \frac{1}{2} \kappa_0 (\Delta \mathbf{X})^2 + \frac{1}{4} \mu_0 (\partial_i \mathbf{X} \cdot \partial_j \mathbf{X} - \delta_{ij})^2 + \frac{1}{8} \lambda_0 (\partial_i \mathbf{X} \cdot \partial_i \mathbf{X} - \delta_{ii})^2 \right], \quad (4)$$

where  $\kappa_0$  is the rigidity constant and  $\mu_0, \lambda_0$  are the elastic Lamé coefficients.

We first study this model in the  $d = \infty$  limit for arbitrary  $D$ . It is convenient to transform  $\mathcal{H}$  through a dummy (Hubbard-Stratonovitch) integration over an auxiliary field  $\lambda^{ij}$  to obtain a Hamiltonian  $\mathcal{H}(\mathbf{X}, \lambda^{ij})$  quadratic in  $\mathbf{X}$ . If we then split  $\mathbf{X}$  into its average  $\mathbf{X}_{av}$  and its fluctuations, we can perform explicitly the Gaussian integration over the fluctuations. The integration  $\lambda_{ij}$  may be done by the saddle-point method. As a result, the following expression is obtained for the effective potential  $\Gamma(\mathbf{X}_{av})$ :

$$\Gamma(\mathbf{X}_{av}) = \left[ \tilde{\mathcal{H}}(\mathbf{X}_{av}, \lambda^{ij}) + \frac{1}{2} k_B T d \text{Tr}_\Lambda \ln(\kappa_0 \Delta^2 - 2\partial_i \lambda^{ij} \partial_j) \right]_{\text{SP}}. \quad (5)$$

The subscript SP refers to the saddle point with respect to  $\lambda^{ij}$  at which the expression is evaluated. An uv-cutoff  $\Lambda$  has been introduced to regularize the trace. The equilibrium configuration  $\mathbf{X}_{eq}$  with *free* boundary conditions is obtained as the extremum of  $\Gamma(\mathbf{X}_{av})$ . We obtain  $\mathbf{X}_{eq} = \zeta_{eq} \mathbf{X}_0(\sigma)$ , where  $\zeta_{eq}$  vanishes above  $T_c$  and is equal to  $(T - T_c)^\beta$  with  $\beta = \frac{1}{2}$  below  $T_c$ . The value of the crumpling transition temperature is given by

$$\frac{1}{k_B T_c} = \frac{d}{D \kappa_0} \int^\Lambda \frac{d^D q}{(2\pi)^D} \frac{1}{q^2}. \quad (6)$$

The lower critical dimension  $D_l$  is identified as the one for which  $T_c$  vanishes. The result  $D_l = 2$  holds for  $d = \infty$ ; however, by setting up a renormalization procedure to first order in  $1/d$ ,<sup>4</sup> one can show that  $D_l = 2 - 2/d + O(1/d^2)$ . The inverse effective propagator is obtained by expansion of  $\Gamma(\mathbf{X}_{av})$  around the equilibrium

configuration  $\mathbf{X}_{eq}$ . From its behavior for  $T \geq T_c$ , one extracts in the usual way the crumpling exponent  $\nu$  and the Hausdorff dimension  $D_F$ . For  $T < T_c$ , the membrane is at its buckling transition point; we therefore extract from the inverse propagator behavior the values of  $\eta'$  and  $\eta_{u'}$  quoted in Table I.

The equilibrium configuration with *constrained* boundary conditions is given by  $\mathbf{X}_{eq} = \zeta \mathbf{X}_0(\sigma)$  provided  $\zeta > \zeta_{sp}$ . Since  $\Gamma(\mathbf{X}_{av})$  is not stationary at  $\mathbf{X}_{eq}$ , a linear term will appear in its expansion, yielding the nonzero tension  $f$ . The equation of state relating  $\zeta$  to  $f$  for small  $f$  (if we omit regular terms in  $f$ ) has the simple form,

$$\zeta^2 = (T_c - T)/T_c + CT(f/\zeta)^{(D-2)/2}, \quad (7)$$

where  $C$  is a constant. For  $T$  approaching  $T_c$  from above, we obtain thus  $\zeta = \chi f$ , with  $\chi \sim |T - T_c|^{-\gamma}$

where  $\gamma = 2/(D-2)$ . At the transition  $\zeta \sim f^{1/\delta}$  with  $\delta$  given by Eq. (3). Finally, for  $T < T_c$  and  $f \rightarrow 0$ , Eq. (7) yields the exponent  $\delta'$  quoted above. From Eq. (7) we also see that four is the upper critical dimension for both the crumpling and buckling transitions: above  $D_u = 4$ , the term on the right-hand side vanishes more rapidly than  $f$ , and thus the regular terms omitted in (7) dominate.

In order to study the properties of the flat phase and the buckling transition for general  $d$ , we can therefore exploit the  $\epsilon$  expansion, with  $\epsilon = 4 - D$ .<sup>7</sup> We consider the statistics of fluctuations around a stretched configuration  $\mathbf{X}_s = \zeta \mathbf{X}_0$ , and redefine the coordinates  $\sigma$  so that  $X_s^a = \delta_i^a \sigma^i$ . We split the small fluctuations  $\delta \mathbf{X} = \mathbf{X} - \mathbf{X}_s$  into  $u$  and  $h$  modes. Near  $D_u = 4$  we can drop irrelevant terms by power counting and obtain the following expression for the effective Hamiltonian<sup>3,7</sup>

$$\mathcal{H}_s = \int d^D \sigma [\tau_0 u_{ii} + \frac{1}{2} \kappa_0 (\Delta h)^2 + \mu_0 u_{ij}^2 + \frac{1}{2} \lambda_0 u_{ii}^2], \quad (8)$$

where  $u_{ij} = \frac{1}{2} (\partial_i u_j + \partial_j u_i + \partial_i \mathbf{h} \cdot \partial_j \mathbf{h})$  and the coefficients  $\kappa_0, \mu_0, \lambda_0$  are related to those of Eq. (4) by simple  $\zeta$ -dependent rescalings. Since  $\mathbf{X}_s$  is not necessarily an extremum of  $\mathcal{H}$ , a linear term  $\tau_0 u_{ii}$  appears in Eq. (8). By our neglecting the irrelevant terms, the Euclidean symmetry of the original Hamiltonian<sup>5</sup> has been explicitly broken. Nevertheless,  $\mathcal{H}_s$  is invariant with respect to the residual symmetry transformations given (for any set of  $D$  vectors  $\mathbf{A}_i$  with  $d_c = d - D$  components) by the following expressions:

$$\begin{aligned} \mathbf{h}(\sigma) &\rightarrow \mathbf{h}(\sigma) + \mathbf{A}_i \sigma^i, \\ u_i(\sigma) &\rightarrow u_i(\sigma) - \mathbf{A}_i \cdot \mathbf{h}(\sigma) - \frac{1}{2} \mathbf{A}_i \mathbf{A}_j \sigma^j. \end{aligned} \quad (9)$$

The associated Ward identities for the effective potential are sufficient to prove the renormalizability of  $\mathcal{H}_s$ . We stress that symmetry does *not* rule out the  $\tau_0 u_{ii}$  term in Eq. (8): even if it is not present at the beginning it would be generated by the renormalization.<sup>14</sup> For constrained boundary conditions thermal fluctuations will, in general, introduce tension in the membrane. The theory of Ref. 7 corresponds therefore to vanishing *renormalized*  $\hat{\tau}$ , and hence to the critical theory at the buckling transition.<sup>11</sup> The renormalization scheme, analogous to that in Ref. 7, introduces in addition the renormalization of  $\tau_0$ :  $\hat{\tau} = Z Z_\tau^{-1} \tau_0 / \kappa_0$ . We obtain the corresponding new exponent function  $\gamma_\tau$ , given to lowest order by

$$\gamma_\tau = (16\pi^2)^{-1} [d_c (\hat{\lambda} + \hat{\mu}/2) + 5A\hat{\mu}], \quad (10)$$

where  $A = (\hat{\lambda} + \hat{\mu})/(\hat{\lambda} + 2\hat{\mu})$ , and  $\hat{\lambda}, \hat{\mu}$  are the renormalized dimensionless Lamé coefficients as in Ref 7. The free membrane and the buckling transition are described by the unstable  $\hat{\tau} = 0$  plane. If we analyze in a standard fashion the corresponding fixed point, the critical exponents  $\eta'$  and  $\eta_u'$  of Table I can be deduced.<sup>11</sup> For a stretched membrane the effective tension  $f$  can be ob-

tained from the inverse propagation  $\Gamma_{hh}$ , as a consequence of the Ward identities:

$$f \sim (\partial \Gamma_{hh} / \partial q^2) |_{q^2=0} \sim \hat{\tau}^{(2-\eta')/(2-\gamma_\tau^*)}, \quad (11)$$

where  $\gamma_\tau^*$  is  $\gamma_\tau$  evaluated at the nontrivial buckling fixed point.<sup>11</sup> The exponent  $\delta'$  can also be obtained from this equation if we assume that  $\zeta - \zeta_{sp}$  is a regular function of  $\hat{\tau}$ . Other buckling exponents can be obtained from scaling relations, whose derivations are not difficult.<sup>13</sup>

Further work to understand the effects of interactions and the nature of the buckled state is clearly needed. The present status of the theory, however, already allows us to expect a rich thermodynamic behavior of polymerized membranes, making experimental investigations of these systems all the more urgent. One possibility is to investigate the polymerized vesicles of Ref. 8, where phenomena which resemble buckling have indeed been observed.<sup>15</sup>

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*Note added.*—In a recent preprint by J. Arnowitz, L. Golubovic, and T. C. Lubensky, the authors derive an extra scaling law:  $\delta'(D-2+\eta') = 2 - \eta'$  which reduces the number of independent exponents. Indeed, this relation can be obtained in the present formalism.<sup>13</sup>

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<sup>1</sup>See Proceedings of the Fifth Jerusalem Winter School, 28 December 1987–6 January 1988, edited by D. R. Nelson, T. Piran, and S. Weinberg (to be published).

<sup>2</sup>Although no systematic experiments have been performed on real polymerized membranes, such a flat phase has indeed been observed in recent numerical simulations. Y. Kantor and D. R. Nelson, Phys. Rev. Lett. **58**, 2774 (1987).

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<sup>9</sup>The phase diagram presented in Fig. 1 is obtained in the thermodynamic limit  $L \rightarrow \infty$ , with  $\zeta$  fixed.

<sup>10</sup>The arguments of Ref. 5 suggest that the crumpling transition could be weakly first order for small embedding dimensions.

<sup>11</sup>The exponents  $\eta'$  and  $\eta_u'$  were already introduced and computed in Ref. 7. However, in this paper the authors did not

consider the possibility of constrained boundary conditions and therefore did not explore the *unstable* direction in the renormalization-group flow, characterized by  $\delta'$  or  $\nu'$ . The apparently stable fixed point (Ref. 4) of this reference corresponds, in fact, to the fixed point describing the buckling transition.

<sup>12</sup>In fact, the possibility of the crumpling transition for two-

dimensional systems is in itself an evidence for such a nontrivial fixed point, since a Gaussian trivial fixed point would be unstable (in  $D=2$ ) according to the Mermin-Wagner theorem.

<sup>13</sup>Details of the calculations will be published elsewhere.

<sup>14</sup>The fact that  $\hat{\tau}$  is proportional to  $\tau_0$  in our calculation is an artifact of the dimensional regularization scheme we adopt.

<sup>15</sup>E. Sackmann, private communication.