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Complete unbinding of fluid membranes in the presence of short-ranged forces

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Lipid or surfactant bilayers which are bound by an external pressure and interact via an additional short-ranged potential are studied theoretically. If the latter potential is not strong enough to bind the lamellae by itself, it has asymptotically no effect on the (complete) unbinding transition, which occurs in the limit of vanishing pressure; the separation and correlation lengths diverge as power laws as a function of the pressure, with the amplitudes being determined by characteristic amplitude ratios. If the potential strength exceeds the critical value, the bilayers are bound even for zero external pressure (incomplete unbinding). Exactly at the critical potential strength, all length scales again diverge as a function of the pressure. The critical exponents are found to be identical to those for a less attractive potential, but the asymptotic amplitude ratios have different values; also, the fluctuation amplitude, which measures the strength of the fluctuation-induced repulsion between the bilayers, is reduced by a factor of 12 as compared to the subcritical case. These results are obtained directly by Monte Carlo simulations of two fluid membranes and agree with exact calculations for the analogous system of two strings in 1+1 dimensions. Experimentally, the effects of short-ranged van der Waals attraction on the fluctuation amplitude c_{fl} should be observable for suitable systems by small-angle x-ray scattering on lamellar phases.

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I. INTRODUCTION

In many experimental situations, lipid or surfactant systems form ordered stacks of bilayers, in which the lamellae are on average parallel to each other and separated by layers of solvent. The layer spacing in this lamellar phase can be determined either by the total volume fraction of solvent, in which case separations of up to 1 μm are obtainable [1,2], or by a balance of an externally applied pressure and a variety of other direct forces which act between the layers. The limit of zero pressure corresponds to the case where the lamellae are swollen in excess water, i.e., the unconstrained case. Experimentally, the pressure can be varied over many orders of magnitude using mechanical, osmotic, and vapor-pressure techniques [3]. The direct forces comprise the omnipresent van der Waals attraction, in the case of charged membrane electrostatic interactions, and the so-called hydration forces, which are strongly repulsive and act at very short distances [4]. In addition, membranes cannot cross, which is accounted for by a hard-wall interaction at zero separation. Thermally excited shape fluctuations turn out to be of crucial importance in these systems and act to renormalize the direct interactions [5], thus influencing the mean separation between the membranes. Quite generally, shape fluctuations are severely restricted by the presence of neighboring membranes, which leads to an effective repulsion between the membranes, the so-called Helfrich interaction [6].

In a heuristic way, the effect of thermal fluctuations can be incorporated by adding the Helfrich term to the direct interactions and then proceeding in a mean-field manner by simply minimizing the obtained expression. Although this procedure fails in the case of *critical* unbinding [7], it turns out to be accurate for *complete* unbinding driven by external pressure [8]. Using simple scaling ideas, the Helfrich interaction can be shown to be of the form $V_{\mathrm{fl}} = c_{\mathrm{fl}}T^2/Kl^2$, where l is the separation between the membranes and K is the bending rigidity [6]. The universal amplitude c_{fl} measures the strength of the fluctuations and plays a crucial role in the physics of membranes and membrane stacks. This amplitude not only determines the mean membrane separation in the case where the spacing is allowed to equilibrate under the action of an external pressure, but it can also be measured directly using x-ray scattering techniques [9,10]. The estimate due to Helfrich obtained for the pure hard-wall case using a harmonic approximation, $c_{\mathrm{fl}} = 3\pi^2/128 \simeq 0.2313$, agrees surprisingly well with the above mentioned scattering experiments. Former Monte Carlo estimates of this quantity yielded values smaller by a factor of roughly 2 [11,12]. This discrepancy, which is further substantiated by Monte Carlo results obtained in this work, still has to be resolved.

In this article, the effect of an additional short-ranged potential on the (complete) unbinding transition and specifically on the amplitude c_{fl} is considered. The interplay of the short-ranged potential and the external pressure (the latter can be thought of as a linear and thus rather long-ranged potential acting between the membranes) has a number of interesting consequences. As a main result, the amplitude c_{fl} and the amplitudes of all other length scales are identical to the pure hard-wall case in the asymptotic limit of vanishing pressure, as long as the potential is not attractive enough to bind the membranes by itself; this defines the *subcritical* case. The

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amplitudes of the various diverging length scales are connected by characteristic amplitude ratios. If the potential strength exceeds the critical value, constituting the *supercritical* case, the membranes are bound even in the absence of an external pressure and no unbinding transition takes place (incomplete unbinding). In between, exactly at the critical potential strength, referred to as the *critical* case, the fluctuation amplitude c_{fl} is reduced by a factor of 12 and the characteristic amplitude ratios have distinct values. Also, the lateral height-height correlation function is found to be qualitatively different for these cases; the large range of distances over which a membrane is anticorrelated as it is found for the exactly solvable harmonic case is reduced for the supercritical case and actually disappears completely for the subcritical case. These results also apply to the case where the layer spacing is determined by the solvent volume fraction, for which the pressure variable used in the present calculations functions as a Lagrange multiplier fixing the conjugated separation variable [8].

The calculations are always restricted to one separation coordinate, i.e., to the case of just two membranes. In fact, the results should also be valid for stacks of many membranes, as suggested by recent work on the critical unbinding of membrane stacks [13] and by explicit Monte Carlo simulations of stacks of three and four membranes bound by external pressure [14]. In the latter work it was shown that a stack of membranes which are bound by pressure only can asymptotically be described as an assembly of decoupled membrane pairs. If the stack is bound by short-ranged forces acting between the membranes, the situation turns out to be more subtle; however, if the membranes are identical they still unbind simultaneously at a temperature that does not depend on the number of membranes involved.

First, exact calculations for the case of strings in 1+1 dimensions are presented, corresponding to wetting in two dimensions. Following a renormalization-group argument, strings in 1+1 dimensions, governed by line tension, and membranes in 2+1 dimensions, governed by bending rigidity, should exhibit identical critical behaviors [15,16]. Indeed, the extensive Monte Carlo simulations of two fluid membranes presented in Sec. III confirm this expectation; even the amplitude ratios of different length scales turn out to be identical. Section IV summarizes the results and contains possible experimental applications and implications.

II. EXACT RESULTS FOR STRINGS

Here the unbinding of strings in 1+1 dimensions is considered under the action of both external pressure and a short-ranged potential. The calculation can be done exactly using the Schrödinger-equation formalism. It turns out that the results can be carried over directly to the case of fluid membranes, as will be expounded in Sec. III.

To proceed, consider the effective Hamiltonian appropriate for the unbinding of strings

$$\mathcal{H} = \int dx \left\{ \frac{K}{4} (\nabla l)^2 + V(l) + Pl \right\}, \tag{1}$$

where the coordinate l corresponds to the separation of one string with line tension $K/2$ from a wall or to the separation of two strings with line tension K each after extraction of the center-of-mass coordinate. For simplicity, the potential $V(l)$ is taken to be of a square well form

$$V(l) = \begin{cases} \infty & \text{for } \quad l \le 0 \\ V^0 & \text{for } 0 < \; l < l^0 \\ 0 & \text{for } l^0 \le \; l. \end{cases} \tag{2}$$

Attraction corresponds to $V^0 < 0$, repulsion to $V^0 > 0$. The thermodynamic properties of this problem can be obtained using transfer-matrix methods. For a finite short-distance cutoff, one has to determine the eigenvalues and eigenfunctions of the transfer matrix from an integral equation [16]. In the limit of zero cutoff, this integral equation reduces to the Schrödinger-type equation [17] as given by

$$\left[-\frac{T^2}{K} \frac{\partial^2}{\partial l^2} + Pl - E_n + V^0 \right] \Phi_n(l) = 0 \quad \text{for } l < l^0,$$

$$\left[-\frac{T^2}{K} \frac{\partial^2}{\partial l^2} + Pl - E_n \right] \Phi_n(l) = 0 \quad \text{for } l > l^0. \tag{3}$$

In order to simplify the notation, one introduces the rescaled variables

$$\begin{aligned} y &\equiv (PK)^{1/3} T^{-2/3} l, \\ y_0 &\equiv (PK)^{1/3} T^{-2/3} l^0, \\ \varepsilon_n &\equiv (PT)^{-2/3} K^{1/3} E_n, \\ v &\equiv -(PT)^{-2/3} K^{1/3} V^0. \end{aligned} \tag{4}$$

Then the Schrödinger equations (3) can be written as

$$\left[-\frac{\partial^2}{\partial y^2} + y - \varepsilon_n - v \right] \Psi_n(y) = 0 \quad \text{for } y < y_0,$$

$$\left[-\frac{\partial^2}{\partial y^2} + y - \varepsilon_n \right] \Psi_n(y) = 0 \quad \text{for } y > y_0. \tag{5}$$

Within the transfer-matrix formalism, the unbinding of the string is given by the unbinding of the ground-state Ψ_0, which undergoes a transition from a bound to a scattering state. The general solution of (5), which has to be normalizable, is given by

$$\Psi_n(y) = A\,\mathrm{Ai}(y - \varepsilon_n - v) + B\,\mathrm{Bi}(y - \varepsilon_n - v) \tag{6}$$

for $y < y_0$ and

$$\Psi_n(y) = C\,\mathrm{Ai}(y - \varepsilon_n) \tag{7}$$

for $y > y_0$. The latter form ensures that $\Psi_n(y)$ vanishes as $y \to \infty$. The hard wall at $y = 0$ forces $\Psi_n(0) = 0$, which together with (6) leads to

$$B = -A \frac{\mathrm{Ai}(-\varepsilon_n - v)}{\mathrm{Bi}(-\varepsilon_n - v)}. \tag{8}$$

The usual matching condition requires that $\Psi_n(y)$ and $\partial \Psi_n(y)/\partial y$ are continuous at $y = y_0$ and takes the final form

$$\mathcal{G} \equiv \frac{\mathrm{Ai}'(y_0-\varepsilon_n-v)-\mathrm{Bi}'(y_0-\varepsilon_n-v)\frac{\mathrm{Ai}(-\varepsilon_n-v)}{\mathrm{Bi}(-\varepsilon_n-v)}}{\mathrm{Ai}(y_0-\varepsilon_n-v)-\mathrm{Bi}(y_0-\varepsilon_n-v)\frac{\mathrm{Ai}(-\varepsilon_n-v)}{\mathrm{Bi}(-\varepsilon_n-v)}}$$

$$= \frac{\mathrm{Ai}'(y_0-\varepsilon_n)}{\mathrm{Ai}(y_0-\varepsilon_n)}, \tag{9}$$

from which the energies ε_n can be determined. The prime denotes the derivative of the Airy function.

A. Zero potential

In the limit of vanishing potential, i.e., $v = 0$, Eq. (9) reduces to $\mathrm{Ai}(-\varepsilon_n) = 0$ [17–21]. Consequently, the energy spectrum of the Schrödinger equation is determined by the zeros of the Airy function $\varepsilon_0 = 2.3381$, $\varepsilon_1 = 4.0879$, and so on. Within the transfer-matrix formalism, the free energy of the problem defined by (1) and (2) is given by

$$f \equiv E_0 = \varepsilon_0 (PT)^{2/3} K^{-1/3}. \tag{10}$$

Using standard thermodynamic relations, the internal energy U and the mean separation $\langle l \rangle$ can be calculated from the free energy f,

$$U \equiv \langle \mathcal{H} \rangle = -T^2 \frac{\partial f/T}{\partial T} = \frac{1}{3} f, \tag{11}$$

$$\langle l \rangle = T \frac{\partial f/T}{\partial P} = \frac{2}{3} f/P = \varepsilon_0 \frac{2}{3} \left(\frac{T^2}{KP} \right)^{1/3}. \tag{12}$$

With the definition of the free energy $f = U - TS$, the entropic contribution to the free energy turns out to be

$$TS = -T \frac{\partial f}{\partial T} = -\frac{2}{3} f. \tag{13}$$

For the elastic contribution one obtains

$$\langle \mathcal{H}_{\mathrm{el}} \rangle \equiv U - P \langle l \rangle = -f/3, \tag{14}$$

which means that the elastic free energy is maximized in the unbound state. The total fluctuation-induced part of the free energy, which is defined as the sum of the elastic and entropic contributions, is given by

$$V_{\mathrm{fl}} \equiv \langle \mathcal{H}_{\mathrm{el}} \rangle - TS = f/3. \tag{15}$$

Using (12) this fluctuation potential can be written as a function of the mean separation $\langle l \rangle$,

$$V_{\mathrm{fl}}(\langle l \rangle) = \frac{c_{\mathrm{fl}} T^2}{K \langle l \rangle^2}, \tag{16}$$

with the fluctuation amplitude c_{fl} having the value

$$c_{\mathrm{fl}} = (2\varepsilon_0/3)^3/2 = 1.8936. \tag{17}$$

The total free energy f can now be written as a function of the mean separation $\langle l \rangle$ and separates into the fluctuation contribution $V_{\mathrm{fl}}(\langle l \rangle)$ and the potential energy $P\langle l \rangle$,

$$f(\langle l \rangle) = P \langle l \rangle + \frac{c_{\mathrm{fl}} T^2}{K \langle l \rangle^2}. \tag{18}$$

Minimization of this free-energy expression with respect to the mean separation $\partial f(\langle l \rangle)/\partial \langle l \rangle$ leads back to (12); the separation into the direct potential and the fluctuation potential is therefore exact. This is a direct consequence of the long-ranged character of the pressure interaction. Indeed, such a superposition is expected to be accurate for all potentials that decay for large separations slower than the fluctuation potential itself [19]. It is to be noted that the fluctuation potential as given by (16) depends on the mean separation and not on the separation itself.

The parallel correlation length $\xi_{\parallel}$, which measures the decay of correlations parallel to the string, is given by

$$\xi_{\parallel} = \frac{T}{E_1 - E_0} = \left(\frac{TK}{P^2} \right)^{1/3} \frac{1}{\varepsilon_1 - \varepsilon_0}, \tag{19}$$

with $\varepsilon_1 - \varepsilon_0 = 1.7498$. For the ratio of separation and parallel correlation length one obtains the characteristic value

$$\frac{\langle l \rangle}{\xi_{\parallel}^{1/2}} = \frac{2\varepsilon_0 \sqrt{\varepsilon_1 - \varepsilon_0}}{3} \left(\frac{T}{K} \right)^{1/2} = 2.0619 \left(\frac{T}{K} \right)^{1/2}. \tag{20}$$

In order to determine the roughness or perpendicular correlation length $\xi_{\perp}$ one first calculates

$$\langle y^2 \rangle = \frac{\int_0^\infty y^2 \mathrm{Ai}^2(y - \varepsilon_0) dy}{\int_0^\infty \mathrm{Ai}^2(y - \varepsilon_0) dy} = \varepsilon_0^2 \frac{8}{15}, \tag{21}$$

from which the roughness follows to be

$$\xi_{\perp} \equiv \sqrt{\langle l^2 \rangle - \langle l \rangle^2} = \varepsilon_0 \frac{2}{3\sqrt{5}} \left(\frac{T^2}{KP} \right)^{1/3}. \tag{22}$$

The ratio of separation and roughness is thus given by

$$\frac{\langle l \rangle}{\xi_{\perp}} = \sqrt{5}. \tag{23}$$

B. Repulsive potential

One is interested in the asymptotic regime where the pressure P goes to zero while the potential strength V^0 is a positive constant; in this limit, the rescaled potential strength v is negative and diverges, the rescaled potential range y_0 goes to zero, and the rescaled energies ε_n approach constant values. Thus the arguments of the Airy functions on the left-hand side of (9) are positive divergent. For $x \to \infty$, the Airy functions $\mathrm{Ai}(x)$ and $\mathrm{Bi}(x)$ are given by [20]

$$\mathrm{Ai}(x) \sim \frac{1}{2} \pi^{-1/2} x^{-1/4} e^{-2x^{3/2}/3},$$

$$\mathrm{Bi}(x) \sim \pi^{-1/2} x^{-1/4} e^{2x^{3/2}/3}. \tag{24}$$

Expanding the separate terms of $\mathcal{G}$ around $(-v - \varepsilon_n)$ up to second order in y_0 one obtains

$$\mathcal{G} \simeq \frac{1}{y_0} - \frac{y_0}{2}(v + \varepsilon_n) \simeq \frac{1}{y_0}\left(1 - \frac{u}{2}\right), \tag{25}$$

where the pressure-independent rescaled potential strength

$$u \equiv v y_0^2 = -K V^0 (l^0/T)^2 \tag{26}$$

has been used. For vanishing pressure, i.e., for $y_0 \to 0$, the expression (25) diverges. From (9) it follows that $\mathrm{Ai}(y_0 - \varepsilon_n) = 0$ holds in this limit, so one is led back to the results for zero potential; in fact, a repulsive potential is asymptotically irrelevant and the ground-state energy is given by $\varepsilon_0 = 2.3381$ for vanishing pressure. In order to extract the leading correction to the asymptotic behavior, one expands the inverse of the right-hand side of (9) around $y_0 - \varepsilon_0 = 2.3381$,

$$\frac{\mathrm{Ai}(y_0 - \varepsilon_0)}{\mathrm{Ai}'(y_0 - \varepsilon_0)} \simeq 2.3381 + y_0 - \varepsilon_0, \tag{27}$$

and equates it with the inverse of (25), obtaining for the ground-state energy to lowest order in y_0 and u

$$\varepsilon_0 \simeq 2.3381 - y_0 u/2. \tag{28}$$

For nonzero pressure, the ground-state energy increases, as expected for a repulsive potential. For infinitely repulsive potential $V^0 = \infty$ (this amounts to a shift of the hard wall), the ground-state energy is given by

$$\varepsilon_0 = 2.3381 + y_0. \tag{29}$$

C. Attractive potential

In this case, v is a positive number, which diverges for vanishing pressure. For $x \to -\infty$, the Airy functions $\mathrm{Ai}(x)$ and $\mathrm{Bi}(x)$ are given by [20]

$$\mathrm{Ai}(x) \sim \pi^{-1/2}(-x)^{-1/4} \sin\left(\frac{2}{3}(-x)^{3/2} + \frac{\pi}{4}\right)$$
$$\mathrm{Bi}(x) \sim \pi^{-1/2}(-x)^{-1/4} \cos\left(\frac{2}{3}(-x)^{3/2} + \frac{\pi}{4}\right). \tag{30}$$

Using the derivatives of these expressions

$$\mathrm{Ai}^{(i)}(x) = \begin{cases} (-1)^{\frac{i+1}{2}}(-x)^{\frac{i}{2}}\mathrm{Bi}(x) & \text{for} \quad i \quad \text{odd} \\ (-1)^{\frac{i}{2}}(-x)^{\frac{i}{2}}\mathrm{Ai}(x) & \text{for} \quad i \quad \text{even,} \end{cases} \tag{31}$$

$$\mathrm{Bi}^{(i)}(x) = \begin{cases} (-1)^{\frac{i-1}{2}}(-x)^{\frac{i}{2}}\mathrm{Ai}(x) & \text{for} \quad i \quad \text{odd} \\ (-1)^{\frac{i}{2}}(-x)^{\frac{i}{2}}\mathrm{Bi}(x) & \text{for} \quad i \quad \text{even,} \end{cases} \tag{32}$$

the expansion of the left-hand side of (9) around $(-\varepsilon_n - v)$ becomes exactly summable and leads to

$$\mathcal{G} \simeq \frac{\sum_{i=0}^{\infty}\left\{\left[-y_0^2(\varepsilon_n + v)\right]^i/(2i)!\right\}}{y_0 \sum_{i=0}^{\infty}\left\{\left[-y_0^2(\varepsilon_n + v)\right]^i/(2i+1)!\right\}}. \tag{33}$$

With the help of the identities

$$\cos\sqrt{x} = \sum_{i=0}^{\infty} \frac{(-x)^i}{(2i)!}, \tag{34}$$

$$\frac{\sin\sqrt{x}}{\sqrt{x}} = \sum_{i=0}^{\infty} \frac{(-x)^i}{(2i+1)!}, \tag{35}$$

one finally obtains

$$\mathcal{G} \simeq \frac{\sqrt{\varepsilon_n + v}}{\tan(y_0\sqrt{\varepsilon_n + v})} = \frac{\mathrm{Ai}'(y_0 - \varepsilon_n)}{\mathrm{Ai}(y_0 - \varepsilon_n)}, \tag{36}$$

which determines the energy spectrum ε_n. The behavior depends on the potential strength v and can be conveniently classified using the parameter $u \equiv v y_0^2$; see (26). In the limit of vanishing pressure, i.e., for $y_0 \to 0$, the expression $\mathcal{G}$ takes the values

$$\mathcal{G} = \begin{cases} +\infty & \text{for} \ \ u < \pi^2/4 \\ 0 & \text{for} \ \ u = \pi^2/4 \\ -\infty & \text{for} \ \ u > \pi^2/4. \end{cases} \tag{37}$$

As expected, the potential strength $u = \pi^2/4$ presents a border line, since for this value the string undergoes a critical unbinding transition in the absence of external pressure [21,22]. The three cases defined by (37) will now be separately discussed.

For $u < \pi^2/4$, the subcritical case, it is immediately evident from (36) and (37) that $\mathrm{Ai}(y_0 - \varepsilon_n) = 0$ holds. Consequently, the ground-state energy ε_0 for $y_0 \to 0$ is again given by the first zero of the Airy function, i.e., $\varepsilon_0 = 2.3381$, as for zero potential; here it is important to note that $\mathrm{Ai}'(-\varepsilon_0)$ is indeed positive definite. To leading order in y_0 one obtains

$$\varepsilon_0 \simeq 2.3381 - y_0\left(\frac{\tan\sqrt{u}}{\sqrt{u}} - 1\right), \tag{38}$$

where the linear term becomes infinitely large for $u \to \pi^2/4$. This dependence of ε_0 on y_0 is a subdominant correction and the asymptotic thermodynamic behavior does correspond to the case of zero potential.

For $u = \pi^2/4$, the critical case, it follows from (36) and (37) that the energy spectrum is determined by $\mathrm{Ai}'(y_0 - \tilde{\varepsilon}_n) = 0$. The energy levels are given by the zeros of the first derivative of the Airy function, i.e., $\tilde{\varepsilon}_0 = 1.018\,79$ and $\tilde{\varepsilon}_1 = 3.248\,19$. In analogy to the calculation in Sec. II A, the free energy is given by

$$f \equiv E_0 = \tilde{\varepsilon}_0 (PT)^{2/3} K^{-1/3} \tag{39}$$

and the expressions (11)–(16) hold similarly. This means that the simple superposition ansatz (18) still holds in the asymptotic limit of vanishing pressure, which is a rather unexpected result. Specifically, the separation is given by

$$\langle l \rangle = T\frac{\partial f/T}{\partial P} = \frac{2}{3}f/P = \tilde{\varepsilon}_0 \frac{2}{3}\left(\frac{T^2}{KP}\right)^{1/3}. \tag{40}$$

For the ratio of the separations for the subcritical and the critical case one obtains

$$\frac{\langle l(u=0)\rangle}{\langle l(u=\pi^2/4)\rangle} = \frac{\varepsilon_0}{\tilde{\varepsilon}_0} = 2.2950, \tag{41}$$

a value that is also found for membranes using Monte Carlo simulations (see Sec. III). The fluctuation potential is given by the general form (16), but the fluctuation amplitude now has the value

$$c_{\rm fl} = (2\tilde{\varepsilon}_0/3)^3/2 = 0.1567, \tag{42}$$

which is 12 times smaller than that for the subcritical case, Eq. (17). The fluctuations are therefore drastically reduced by the presence of a critical short-ranged potential. For $y_0 \to 0$, the expectation value $\langle y^2\rangle$ can be calculated in analogy to Sec. II A, yielding

$$\langle y^2\rangle = \frac{\int_0^\infty y^2 \mathrm{Ai}^2(y-\tilde{\varepsilon}_0)dy}{\int_0^\infty \mathrm{Ai}^2(y-\tilde{\varepsilon}_0)dy} = 0.749\ 878. \tag{43}$$

For the roughness one obtains

$$\xi_\perp \equiv \sqrt{\langle l^2\rangle - \langle l\rangle^2} = 0.537\ 189 \left(\frac{T^2}{KP}\right)^{1/3}. \tag{44}$$

The ratio of the separation to the roughness is given by

$$\frac{\langle l\rangle}{\xi_\perp} = 1.2643. \tag{45}$$

This ratio is substantially smaller than that for the subcritical case, the string is rougher. For the parallel correlation length $\xi_\parallel$ one obtains

$$\xi_\parallel = \left(\frac{TK}{P^2}\right)^{1/3} \frac{1}{\tilde{\varepsilon}_1 - \tilde{\varepsilon}_0}, \tag{46}$$

with $\tilde{\varepsilon}_1 - \tilde{\varepsilon}_0 = 2.2294$. The ratio of the separation to the parallel correlation length is given by

$$\frac{\langle l\rangle}{\xi_\parallel^{1/2}} = \frac{2\tilde{\varepsilon}_0\sqrt{\tilde{\varepsilon}_1 - \tilde{\varepsilon}_0}}{3}\left(\frac{T}{K}\right)^{1/2} = 1.0141\left(\frac{T}{K}\right)^{1/2}. \tag{47}$$

For $u > \pi^2/4$, the supercritical case, the ground-state energy is given by $\varepsilon_0 = -\infty$ for vanishing pressure, i.e., $y_0 \to 0$, as follows from (36). However, the expression $\varepsilon_0 y_0^2 = Kf(l^0/T)^2$, which is proportional to the free energy, approaches a negative but finite constant as $y_0 \to 0$. This corresponds to an incomplete unbinding transition, for which the string remains bound by the short-ranged potential even for zero pressure. In this limit and as u approaches $u_c = \pi^2/4$, the string unbinds via a critical unbinding transition with the separation asymptotically given by

$$\langle l\rangle = l^0(u-u_c)^{-1}. \tag{48}$$

For $P = 0$ and as $u \to u_c$, the various length scales are connected by the simple expression

$$\xi_\perp = \langle l\rangle = \frac{1}{2}\left(\frac{T}{K}\right)^{1/2} \xi_\parallel^{1/2}. \tag{49}$$

III. MEMBRANES

The appropriate effective Hamiltonian for the unbinding of fluid membranes reads

$$\mathcal{H} = \int d^2\mathbf{x} \left\{\frac{K}{4}\left(\nabla^2 l\right)^2 + V(l) + Pl\right\}, \tag{50}$$

where the coordinate l corresponds again to the separation of one membrane with bending rigidity $K/2$ from a flat substrate or to the separation between two membranes with bending rigidity K each. The potential $V(l)$ is again given by (2).

For the Monte Carlo studies, the $\mathbf{x}$ coordinate is discretized on a square lattice with sites $\{x_{i,j}\}$ and lattice constant Δx; the membrane configuration is then specified by the discrete set $l_{i,j} \equiv l(x_{i,j})$. Using dimensionless variables $z_{i,j} = l_{i,j}\sqrt{K/T}/\Delta x$, $p = P\Delta x^3/\sqrt{KT}$, and the rescaled potential $U(z)$ with depth $U^0 = V^0\Delta x^2/T$ and range $z^0 = l^0\sqrt{K/T}/\Delta x$, the Hamiltonian takes the form

$$\mathcal{H}/T = \sum_{i,j} \frac{1}{4}\left(\nabla_d^2 z_{i,j}\right)^2 + U(z_{i,j}) + pz_{i,j}, \tag{51}$$

where ∇_d^2 denotes the discretized version of the Laplacian.

In the simulations, a square lattice consisting of 3920 discrete sites is partitioned into five sublattices; each sublattice is updated independently using a fully vectorized code employing the standard Metropolis algorithm [23,13]. In most runs, averages are calculated from $\simeq 10^7$ Monte Carlo steps per site, which typically gives a statistical error of less than 1%. The rescaled potential range was always set to $z^0 = 0.0262$. Among the calculated averages are the mean separation $\langle z\rangle$, the roughness or perpendicular correlation length $\xi_\perp \equiv \langle z^2\rangle_c^{1/2} \equiv \sqrt{\langle z^2\rangle - \langle z\rangle^2}$, and the parallel correlation length $\xi_\parallel$, which was estimated using $\xi_\parallel = \Delta x \exp[2\pi\langle(\nabla l)^2\rangle/T]$ [23].

In analogy to the exact calculation for strings, the free energy can be separated into a fluctuation part and a part due to the direct interaction; for zero potential it has the form

$$F(\langle l\rangle)/T = \frac{P\langle l\rangle}{T} + c_{\rm fl}\frac{T}{K\langle l\rangle^2}\ ,$$
$$\hat{F}(\langle z\rangle) \equiv \Delta x^2 F(\langle l\rangle)/T = p(\langle z\rangle) + \frac{c_{\rm fl}}{\langle z\rangle^2}\ . \tag{52}$$

After minimization $\partial F(\langle l\rangle)/\partial\langle l\rangle = 0 = \partial\hat{F}(\langle z\rangle)/\partial\langle z\rangle$, one obtains

$$\langle l\rangle = \left(\frac{2c_{\rm fl}T^2}{KP}\right)^{1/3},\quad \langle z\rangle = \left(\frac{2c_{\rm fl}}{p}\right)^{1/3}. \tag{53}$$

The fluctuation amplitude $c_{\rm fl}$ can thus be determined from the mean separation. In Fig. 1 the mean separation $\langle z\rangle$ is plotted as a function of reduced pressure p for four different values of the reduced potential strength U^0. From the top, the values are given by $U^0 = 0$, $U^0 = U_c^0/2$, $U^0 = U_c^0$, and $U^0 = 1.1U_c^0$. The vertical bars inside the data points denote the numerical error. For the critical potential strength the estimate $U_c^0 = 2.78 \pm 0.01$ is used [13]. For the case $U^0 = 1.1U_c^0$ one obtains a bound state even for vanishing pressure, which approaches the separation found for zero pressure $\langle z\rangle = 0.562 \pm 0.005$, denoted by a straight horizontal line, rather rapidly. This scenario corresponds to an incomplete unbinding of the membrane. For the two subcritical potential strengths $U^0 = 0$ and $U^0 = U_c^0/2$, the data converge quickly for small pressure and scale accurately like $\sim p^{-1/3}$, as denoted by the straight line. As for the case of strings, a subcritical potential is asymptotically irrelevant. From the amplitude of the separation and following (53), one obtains the estimate for the fluctuation amplitude $c_{\rm fl} = 0.116 \pm 0.002$ [14], which is very close to $3\pi^2/256$ and thus exactly half the so far accepted estimate [6]. For the critical case, the separation scales with the same exponent, but the amplitude is greatly reduced as compared to the subcritical case. The two parallel straight lines in Fig. 1 are drawn exactly with the amplitude ratio as calculated for strings (41) and match the two sets of data quite accurately. For the critical case, the data seem to deviate from the straight line for very small pressures, which simply reflects the fact that the value used as an estimate for U_c^0, which is determined from an independent Monte Carlo simulation for the critical unbinding of two membranes [13], is actually smaller than the true critical value.

In Fig. 2 the ratio of the separations for zero potential and critical potential $\langle z(U^0 = 0)\rangle / \langle z(U^0 = U_c^0)\rangle$, is plotted. The prediction of the string calculation Eq. (41) is denoted by a broken line. For intermediate pressures, the agreement is good. For smaller pressures there are deviations, again due to the uncertainty in the critical potential depth, and the ratio crosses over to unity, as expected for the subcritical case Eq. (49).

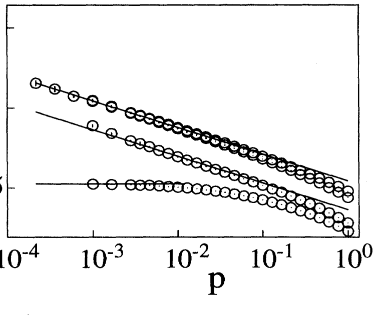

FIG. 1. Monte Carlo results for the separation between two membranes for four different values of the rescaled potential strength, from bottom to top: $U^0 = 1.1U_c^0$, $U^0 = U_c^0$, $U^0 = U_c^0/2$, and $U^0 = 0$, where U_c^0 has been determined independently. Data for the supercritical case $U^0 = 1.1U_c^0$ approach a finite separation of $\langle z\rangle = 0.562 \pm 0.005$ for vanishing pressure p, as denoted by the horizontal straight line. The other data scale asymptotically like $\sim p^{-1/3}$, as indicated by straight lines.

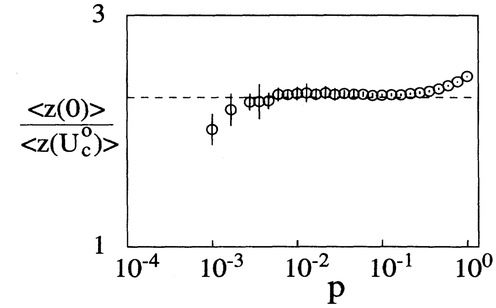

FIG. 2. Ratio of mean separations for zero potential and critical potential strength; the exact result for the corresponding system of strings $\langle z(U^0 = 0)\rangle$ / $\langle z(U^0 = U_c^0)\rangle = 2.295$ is denoted by a broken line. For intermediate values of the pressure p, the Monte Carlo data agree with the string result; for very small values of the pressure, the data progressively deviate from the straight line, indicating that the estimate used for U_c^0 is actually smaller than the true critical potential strength.

Figure 3 shows the ratios of separation and roughness $\langle z\rangle/\langle z^2\rangle_c^{1/2}$ for three different potential strengths $U^0 = 1.1U_c^0$, $U^0 = U_c^0$, and $U^0 = U_c^0/2$ (from the bottom). The data are compared with the results of the string calculations, giving $\langle z\rangle/\langle z^2\rangle_c^{1/2} = 1$, 1.2643, and $\sqrt{5}$, respectively, as given by Eqs. (49), (45), and (23); the agreement is rather good.

At this point it is appropriate to comment on the observed similarity between membranes and strings. In the limit of vanishing rescaling factor, the functional renormalization of the direct interactions leads to identical fixed-point structures for strings and membranes as described by the Hamiltonians (1) and (50) [15]. This is the case even for coupled systems of many strings or membranes [16]. As a consequence, phase diagrams are iden-

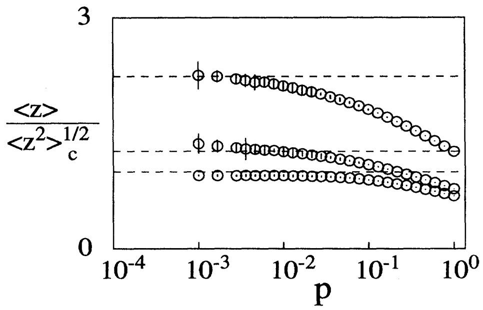

FIG. 3. Ratio of mean separation $\langle z\rangle$ and roughness $\xi_\perp = \langle z^2\rangle_c^{1/2}$ for three different values of the potential strength, from bottom to top: $U^0 = 1.1U_c^0$, $U^0 = U_c^0$, and $U^0 = U_c^0/2$. The results for strings for the three different cases $\langle z\rangle/\xi_\perp = 1$, $\langle z\rangle/\xi_\perp = 1.2643$, and $\langle z\rangle/\xi_\perp = \sqrt{5}$ are denoted by broken lines.

tical and critical exponents are related in a simple way, as indeed observed for the critical unbinding of stacks of membranes and strings [13]. The results presented above show that the same is actually true for amplitude ratios between different length scales; these ratios depend in a very categorical manner on the potential that confines the membrane (which will be discussed below), but do not seem to depend on whether one considers membranes or strings. Based on these notions, one is led to formulate the general relation between the different length scales considered here

$$\mathcal{C}_\perp \xi_\perp = \langle l \rangle = \mathcal{C}_\parallel \left(\frac{T}{K}\right)^{1/2} \left(\frac{\xi_\parallel}{\mathcal{C}_0}\right)^\zeta . \tag{54}$$

The roughness exponent ζ is given by $\zeta = 1/2$ for strings and $\zeta = 1$ for membranes. The values for the constants $\mathcal{C}_\perp$ and $\mathcal{C}_\parallel$ are listed in Table I and do depend on the potential which confines the membrane or string. In contrast, the scale factor $\mathcal{C}_0$ should be independent of the potential and can be determined exactly using a harmonic analysis.

To this end, consider the Hamiltonian (50) for membranes with $P = 0$ and $V(l) = ml^2/2$. The correlation function $C(\mathbf{x}) \equiv \langle [l(\mathbf{x}) - \langle l \rangle][l(\mathbf{0}) - \langle l \rangle] \rangle$ is given by

$$\begin{aligned} C(\mathbf{x}) &= T \int_{-\infty}^{\infty} \frac{d^2\mathbf{q}}{(2\pi)^2} \frac{e^{i\mathbf{q}\cdot\mathbf{x}}}{K\mathbf{q}^4/2 + m} \\ &= \frac{2T}{K} \int_{-\infty}^{\infty} \frac{d^2\mathbf{q}}{(2\pi)^2} \frac{e^{i\mathbf{q}\cdot\mathbf{x}}}{\mathbf{q}^4 + 4\xi_\parallel^{-4}}, \end{aligned} \tag{55}$$

where the correlation length is defined by $\xi_\parallel = (2K/m)^{1/4}$. The integral can be evaluated exactly and leads to

$$C(x) = -\frac{T\xi_\parallel^2}{2\pi K} \mathrm{kei}(\sqrt{2}x/\xi_\parallel). \tag{56}$$

Using the asymptotic expansion for large arguments of the Thomson function $\mathrm{kei}(\sqrt{2}x/\xi_\parallel)$ [24], one obtains the expression valid for large values of x

$$C(x) \simeq \frac{T}{K} \frac{\xi_\parallel^{5/2}}{2\sqrt{\sqrt{2}\pi x}} e^{-x/\xi_\parallel} \sin(x/\xi_\parallel + \pi/8). \tag{57}$$

The exponential factor justifies the definition of the correlation length. Using $\mathrm{kei}(0) = -\pi/4$, one obtains a relation between the roughness and the parallel correlation length

$$C(0) = \xi_\perp^2 = \frac{T}{K} \frac{\xi_\parallel^2}{8}. \tag{58}$$

Similarly for strings, using the Hamiltonian (1) with $P = 0$ and $V(l) = ml^2/2$, one arrives at

$$C(x) = T \int_{-\infty}^{\infty} \frac{dq}{2\pi} \frac{e^{i\mathbf{q}\cdot\mathbf{x}}}{Kq^2/2 + m} = \frac{2T}{K} \int_{-\infty}^{\infty} \frac{dq}{2\pi} \frac{e^{i\mathbf{q}\cdot\mathbf{x}}}{q^2 + \xi_\parallel^{-2}}, \tag{59}$$

with the parallel correlation length defined by $\xi_\parallel = (K/2m)^{1/2}$. The integral leads to

$$C(x) = \frac{T}{K} \xi_\parallel e^{-x/\xi_\parallel}, \tag{60}$$

which gives, for the relation between roughness and parallel correlation length,

$$C(0) = \xi_\perp^2 = \frac{T}{K} \xi_\parallel . \tag{61}$$

Comparing (58) and (61), the scale factor appearing in (54) is determined to be

$$\mathcal{C}_0 = 8. \tag{62}$$

In order to determine the amplitude ratio $\mathcal{C}_\parallel$ for membranes from the Monte Carlo simulation, the full correlation function has also been calculated. In Fig. 4 the normalized correlation function $C(x)/C(0)$ is plotted for (a) $U^0 = 0$ and $p = 0.01$, (b) $U^0 = U_c^0$ and $p = 0.01$, and (c) $U^0 = 1.05U_c^0$ and $p = 0$. The correlation function for $U^0 = 0$ [Fig. 4(a)] resembles the resultant function for the harmonic potential (56) in that it oscillates and shows a wide range of distances for which the membrane is actually anticorrelated. The correlation function for $U^0 = 1.05U_c^0$ [Fig. 4(c)] is rather close to a purely exponential decay [25]. The function in Fig. 4(b) is intermediate between the two other curves. In order to extract the parallel correlation length, the estimate $\xi_\parallel = x/\ln[C(0)/C(x)]$ with $C(x)/C(0)$ chosen close to $e^{-3/2}$ is being used. The data points used for calculating $\xi_\parallel$ are drawn as black circles. This estimate is exact both for purely exponential decay and for a decay governed by (56); it is suggested that it gives reliable values for $\xi_\parallel$

TABLE I. Amplitude ratios of the mean separation $\langle l \rangle$, the roughness $\xi_\perp$, and the parallel correlation length $\xi_\parallel$ for the three cases distinguished by the strength U^0 of the short-ranged potential and for a harmonic potential.

	$\mathcal{C}_\perp = \langle l \rangle/\xi_\perp$	$\mathcal{C}_\parallel = \left(\frac{K}{T}\right)^{1/2} \langle l \rangle/(\xi_\parallel/\mathcal{C}_0)^\zeta$	$\mathcal{C}_\parallel/\mathcal{C}_\perp$	$\mathcal{C}_\parallel$ (MC)
$U^0 < U_c^0$	$\sqrt{5}$	5.8319	2.6081	$\simeq 4.5$
$U^0 = U_c^0$	1.2643	2.8683	2.2691	$\simeq 2.3$
$U^0 > U_c^0$	1	$\sqrt{2}$	$\sqrt{2}$	$\simeq 1.6$
harmonic potential			$\sqrt{8}$	

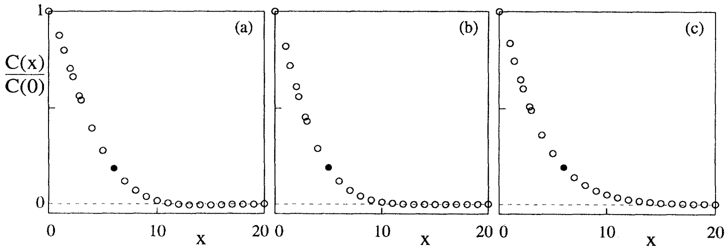

FIG. 4. Normalized correlation function $C(x)/C(0)$ for three different potential strengths (a) $U^0 = 0$, (b) $U^0 = U_c^0$, and $U^0 = 1.05U_c^0$. The distance x is measured in units of the lattice constant Δx. One notices that the region of distances over which the membrane is anticorrelated becomes smaller for the critical potential strength and vanishes altogether for the supercritical case $U^0 = 1.05U_c^0$.

also for the data shown in Fig. 4. The resultant values for $\mathcal{C}_{\|}$ are given in the last column of Table I. The deviations from the exact values for strings are in the limits of uncertainty of the method used for estimating the parallel correlation length. It is therefore plausible that the different values of $\mathcal{C}_{\|}$ found for the different potential scenarios in the string calculation also apply to the case of membranes.

IV. SUMMARY AND DISCUSSION

The complete (i.e., pressure-induced) unbinding of membranes has been investigated, where in addition to the external pressure a short-ranged potential is present. In the calculations, this potential was taken to be of a square-well form, but the results should actually apply to all potentials which contain a hard wall and fall off faster than the fluctuation-induced interaction $V_{\mathrm{fl}} \sim 1/l^2$ [19]. This is the case for the van der Waals attraction as well as for screened electrostatic interactions and the so-called hydration repulsion. The asymptotic behavior, which means the behavior in the limit of vanishing pressure, is divided into three cases: (i) the subcritical case for potentials which are not attractive enough to bind the membranes for zero pressure, including repulsive interactions; (ii) the critical case, where the short-ranged potential has exactly the critical strength; and (iii) the supercritical case, for which the membranes are bound by the potential and where scaling relations hold in the limit where the potential strength approaches the critical value (critical unbinding). For all three scenarios, the relevant length scales are connected by characteristic amplitude ratios, as given by (54) and Table I. The amplitude of the fluctuation-induced Helfrich interaction, which together with the direct interaction determines the unbinding behavior for the complete unbinding cases (i) and (ii), is found to be 12 times smaller for the critical case as compared to the subcritical case.

Experimentally, the fluctuation amplitude c_{fl} has been determined for stacks of surfactant bilayers using small-angle x-ray scattering. In these experiments, the separation between the bilayers is fixed by the amount of solvent added; the pressure P, which appears in the present calculations, connects to the layer spacing via the expression (53). For these systems, the Bragg peaks are replaced by power law singularities $|q_{\|} - q_m|^{\eta_m - 2}$ centered around $q_m = 2\pi m/d$ and characterized by exponents

$$\eta_m = \pi T m^2 [d^4 K \partial^2 f(d)/\partial d^2]^{1/2}/2, \tag{63}$$

where $d = l + \delta$ is the repeat distance of the layers, δ is the layer thickness, and $f(d)$ is the total interaction between the layers (including fluctuation-induced contributions) as a function of the repeat distance d [8]. Using the free-energy expression (52) the exponent η_m is given by

$$\eta_m = \frac{\pi m^2}{2\sqrt{6 c_{\mathrm{fl}}}} \left(1 - \frac{\delta}{d}\right)^2 = \eta^\infty m^2 \left(1 - \frac{\delta}{d}\right)^2 . \tag{64}$$

Taking the Helfrich estimate $c_{\mathrm{fl}} = 3\pi^2/128$ [6], one obtains $\eta^\infty = 4/3$, which is in quantitative agreement with experimental results; for large spacings, only the first Bragg singularity remains [9]. However, the results presented here and in [14] show, however, that c_{fl} is in fact given by $c_{\mathrm{fl}} \simeq 3\pi^2/256$. Without discussing the reason for the apparent agreement between the experimental results and the erroneous value for c_{fl} accepted so far, it is clear that a reduced value of c_{fl} in the case of a critical short-ranged potential would have drastic consequences on the scattering pattern: From the above calculated reduction of c_{fl} by a factor of 12 (and accepting the experimentally measured value $\eta^\infty = 4/3$), one obtains $\eta^\infty = 4\sqrt{12}/3 \simeq 4.6$ for the critical case, from which it follows that there are no Bragg singularities present at all for large distances between the layers.

For which experimental systems could such a critical potential be observed? For surfactant systems, the van der Waals attraction is not strong enough to bind the layers and these systems can be swollen beyond any limits; this is mostly due to the small bending rigidity of the surfactant layers, leading to a strong fluctuation-induced repulsion. Here one is in the subcritical regime; however, for small spacings even for these systems the results presented in this article could be observed experimentally, as will be discussed below. Lamellar systems consisting of lipid bilayers, however, typically show a finite layer spacing even in the absence of external pressure; here fluctuations are weaker due to a much larger bending rigidity. The van der Waals attraction for these systems belongs to the supercritical regime. However, changing the composition of the system, it is fairly easy to approach the critical potential strength. This can be done either by adding a cosurfactant to the bilayers, thereby reducing the bending rigidity [27], or by changing the solvent composition, which influences both the strength of the van der Waals attraction and the bending rigidity of the bilayer [28,29]. In fact, by one of these methods one should actually be able to tune the system to the critical case, for which the reduction of the fluctuation amplitude should be easily measurable.

But even for subcritical potentials, interesting behavior for finite spacings is expected; experimentally, one never reaches the asymptotic limit and corrections to the asymptotic scaling become important. Combining (38), (4), (12), and (17), it follows that $c_{\text{fl}} \sim [1 - c(u) l^0 / \langle l \rangle]^3$ for $\langle l \rangle / l^0 \gg c(u)$, where $c(u)$ diverges as the potential strength approaches the critical value. As follows from (63) by an asymptotic analysis for large repeat distance d, the exponent η_m then attains a maximum value for finite d and reaches the value given by (64) only for larger separations; for a certain range of (subcritical) potential strengths, it follows that actually no Bragg singularities are present for an intermediate range of separations, with the first Bragg singularity reappearing at larger separations [30].

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[30] A behavior similar to the one predicted here has been reported for a surfactant system, where no Bragg singularities were observed for intermediate separations, but actually three peaks were present for very large separations [2]. However, it seems questionable whether van der Waals forces are strong enough in these systems to produce such effects, and no explanation for the reappearance of three Bragg peaks (instead of one) can be offered.