

The unbinding transition of mixed fluid membranes

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Abstract. A phenomenological model for the unbinding transition of multicomponent fluid membranes is proposed, where the unbinding transition is described using a theory analogous to Flory-Huggins theory for polymers. The coupling between the lateral phase separation of inclusion molecules and the membrane-substrate potential provides a rich phase behavior. Our model describes the first-order nature of the unbinding transition in multicomponent membranes as was observed in a recent experiment. In particular, we predict different scenarios of phase coexistence between bound and unbound membrane states.

INTRODUCTION

Adhesion of membranes and vesicles is responsible for cell-cell adhesion which plays an important role in all multicellular organisms. In general, bio-adhesion is governed by the interplay of various generic and specific interactions. Specific interactions act between complementary pairs of proteins such as ligand and receptor, or antibody and antigen. The problem of adhesion of multicomponent membranes is intimately related to that of domain formation. In a recent experiment by Marx *et al.* [1], the role of long-range repulsions due to thermal fluctuations (Helfrich repulsion) of adhering membranes has been addressed. Analyzing the probability distribution of the membrane-substrate spacing for various multicomponent membranes, a phase separation between two distinct lipopolymer-poor and lipopolymer-rich states having two different spacings from the substrate was suggested.

In this paper, we propose a simple phenomenological model for a multicomponent (mixed) fluid membrane which can undergo simultaneously a lateral phase separation and an unbinding transition. The model is motivated by the experiment [1] and relies on the coupling between the inclusion concentration and the membrane-substrate spacing. The lateral phase separation of the inclusion affects the second virial coefficient of the unbinding transition which is taken into account in analogy to the Flory-Huggins theory for polymers. Our model exhibits various types of phase coexistence, including a phase separation between bound and unbound states as well as between two unbound ones. The former phase coexistence indicates the first-order nature of the unbinding transition, as was anticipated in the experiment.

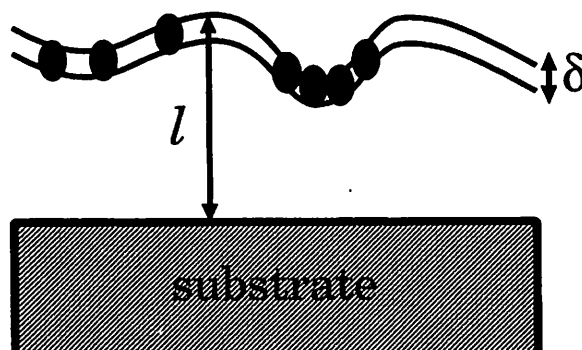


FIGURE 1. A mixed fluid membrane adhering to a substrate. Black filled ovals indicate inclusions such as proteins or lipopolymers. The height of the upper membrane leaflet from the substrate is ℓ , whereas the thickness of the bilayer membrane is δ .

MODEL

Fluid membranes in a lamellar stack or close to a substrate experience steric repulsion arising from their reduced undulation entropy due to the confinement effect. The corresponding interaction energy per unit area has been given by Helfrich as

$$v_s(\ell) = \frac{b(k_B T)^2}{\kappa(\ell - \delta)^2}, \quad (1)$$

where k_B is the Boltzmann constant, T the temperature, κ the bending rigidity of the membrane having thickness δ , and ℓ the average height of the upper membrane lipid leaflet from the substrate (see Fig. 1). While a simple superposition of the Helfrich repulsion, Eq. (1), and other direct interactions gives an incorrect (first-order) description of the unbinding transition, a simple theory

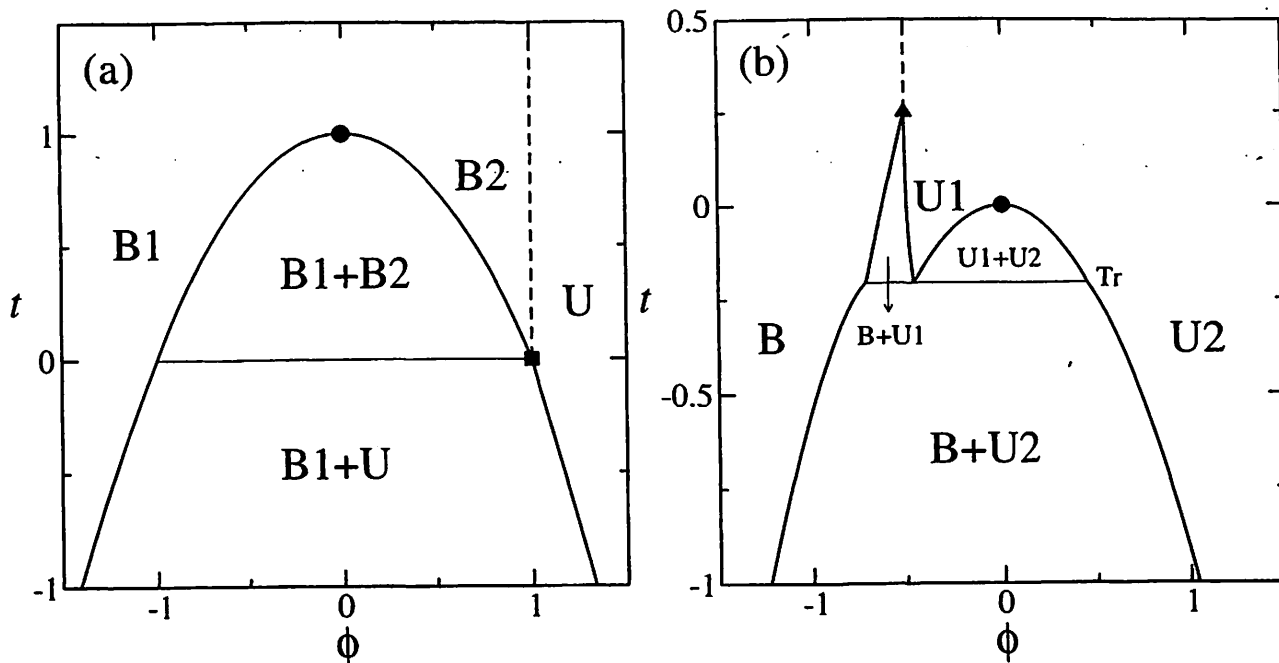


FIGURE 2. The phase diagrams for (a) $\chi = 1$ and (b) $\chi = -0.5$ as a function of ϕ and t when $\gamma = 1$. The continuous line is a first-order line, whereas the dashed line is a second-order one. The critical point, the critical end-point, and the tricritical point are indicated by a circle, square, and triangle, respectively. The bound and the unbound phases are denoted by B and U, respectively.

for the unbinding transition in a bulk of lamellar phase was considered by Milner and Roux [2]. Following the spirit of Flory-Huggins theory for polymers, the Helfrich estimate of the entropy is taken into account accurately, whereas the other interactions are approximately incorporated via a second virial term. Then the free energy *per unit area* of a single membrane can be expressed as

$$f(w) = -k_B T \delta \bar{\chi} w + \frac{b(k_B T)^2}{\kappa \delta^2} w^2, \quad (2)$$

where $w = \delta/\ell \geq 0$ cannot be negative, $\bar{\chi}$ is the second virial coefficient. Minimization of $f(w)$ with respect to w shows that this free energy has a continuous second-order transition at $\bar{\chi} = 0$ between a bound state ($w > 0$) for $\bar{\chi} > 0$ and an unbound state ($w = 0$) for $\bar{\chi} < 0$.

We now consider a two-component membrane adhering to a substrate as in Fig. 1. The overall membrane state is characterized by its average distance ℓ from the substrate. The internal degree of freedom, on the other hand, corresponds to the membrane composition. The interaction between two inclusions is attractive leading to a condensation transition, and the concentration with respect to the critical concentration is defined as ϕ . The proposed free energy *per unit area* of a single mixed membrane undergoing the unbinding transition is

$$f(\phi, w) = -\mu\phi + \frac{1}{2}t\phi^2 + \frac{1}{4}\phi^4 - \chi w + \frac{1}{2}w^2 + \gamma\phi w, \quad (3)$$

with the constraint $w \geq 0$. Here all energy terms have been scaled by $2b(k_B T)^2/\kappa\delta^2$, and are now dimension-

less. The first three terms in Eq. (3) depend only on ϕ , and correspond to the Landau free energy of a two-component membrane undergoing a lateral inclusion-lipid phase separation, μ is the chemical potential conjugate to ϕ , and $t \sim (T - T_c)$ the reduced temperature. The next two terms depend only on w , and represent the unbinding transition of a single membrane as have been described by Eq. (2). In the above, χ is the scaled virial coefficient. The last term is the lowest order coupling term between ϕ and w with a dimensionless coupling coefficient $\gamma > 0$. The physical meaning of this bilinear term is as follows. When the mixed membrane is quenched into the two-phase region, an inclusion-poor phase ($\phi < 0$) coexists with an inclusion-rich phase ($\phi > 0$). This can lead to different direct interactions and hence different second virial coefficients χ for each of the domains. We model this situation by considering an effective virial term in Eq. (3) as $-\chi_{\text{eff}} w = -(\chi - \gamma\phi)w$, which leads to the coupling term ϕw . A typical example of the obtained phase diagrams for $\chi = 1$ and -0.5 are given in Fig. 2 when $\gamma = 1$. Our theory describes the coexistence between the two unbound states.

REFERENCES

1. Marx, S., Schilling, J., Sackmann, E., and Bruinsma, R., *Phys. Rev. Lett.* **88**, 1381021-1381024 (2002).
2. Milner, S.T., and Roux, D., *J. Phys. I France* **2**, 1741-1754 (1992).