

# Parallel structure factors of lateral and transverse phase separations in bilayer biomembranes: Effects of the presence of an interacting wall

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The adsorption of biomolecules such as lipids or proteins on a solid substrate has numerous significant applications in different fields such as drug delivery, pharmaceuticals, and tissue engineering. In such situations, the phase transitions in the lipid membrane are influenced by the presence of an interacting wall. We consider, in this context, a bilayer biomembrane made up of two amphiphiles A and B. Under a variation of a suitable parameter, these systems undergo a phase separation from a homogeneous liquid phase to two distinct liquid phases. We investigate, in this work, the influence of this adsorbing surface on the parallel structure factors  $S(z, z')$  of the lipid mixture. These structure factors are exactly calculated in all cases: below  $T_c$ , above  $T_c$ , and at  $T = T_c$ , as well as their associated local compressibilities  $S(z)$ .

## The model

- We consider a binary fluid membrane formed by a mixture of two lipid molecule species, which compose the two leaflets of the biomembrane. The mixture is assumed to be in a semi-infinite *three-dimensional* space limited by an interacting *2D* -wall.
- The purpose is to explore the influence of this interacting wall on the structure factor of the lipid-mixture. the presence of the adsorbing surface gives rise to drastic changes in the effective interaction governing the transverse and the lateral separations
- The mixing free energy allowing investigation the phase separation is given by:

$$F = F_0 + F_c$$

here,  $F_0$  is the free energy of two plan leaflets. Whereas,  $F_c$  accounts for the part of energy due to the curvature.

- Two fields  $\varphi$  and  $\psi$  (*order parameters*), associated with the two monolayers, are introduced in order to describe the physics. They write as:

$$\begin{aligned} \varphi &= \phi_A^{(1)} - \phi_B^{(1)}, & \text{for leaflet 1} \\ \psi &= \phi_A^{(2)} - \phi_B^{(2)}, & \text{for leaflet 2} \end{aligned}$$

hereabove,  $\phi_\alpha^{(i)}$  stands for the composition of phospholipids  $\alpha = A, B$  on sheet  $i = 1, 2$ .

- The free energy governing physics is given by the following functional of the order Parameters  $\varphi$  and  $\psi$ :

$$\frac{F[\varphi, \psi]}{k_B T} = \frac{1}{a_0^2} \int d^2 \mathbf{r} \left\{ \frac{a_0^2}{2} (\nabla \varphi)^2 + \frac{a_0^2}{2} (\nabla \psi)^2 + \frac{1}{2} \alpha_1 \varphi^2 + \frac{1}{2} \alpha_2 \psi^2 - \Lambda \varphi \psi + \frac{1}{4} u \varphi^4 + \frac{1}{4} v \psi^4 \right\}$$

with the notations

$$\alpha_1 = 1 - \frac{1}{2} \chi_{11} + \frac{1}{16} \bar{\kappa} (a_0 \Delta c_0), \quad \alpha_2 = 1 - \frac{1}{2} \chi_{22} + \frac{1}{16} \bar{\kappa} (a_0 \Delta c_0),$$

$$\Lambda = \chi_{12} - \frac{1}{16} \bar{\kappa} (a_0 \Delta c_0), \quad \chi_{12} = \frac{1}{4} (\chi_{AB} - \frac{\chi_{AA} + \chi_{BB}}{2}).$$

- There  $\mathbf{r}$  is the transverse vector,  $a_0$  the size of polar head. Here,  $\chi_{11}$  and  $\chi_{22}$  stand for the respective segregation parameters describing the effective chemical interactions on leaflets 1 and 2.  $\chi_{12}$  measures the interaction strength between amphiphiles belonging to different sheets.  $\Delta c_0 = c_0^A - c_0^B$  is the asymmetry gap, and the parameter  $\bar{\kappa} = (\kappa/k_B T)$  stands for the reduced bending rigidity constant.

## Structure factor near surface

- The free energy of the system writes:  $F = F_b + F_s$ , where  $F_b$  constitutes the contribution associated with the bulk, and  $F_s$  that relative to the surface.
- We note, here, that the surface energy contributes to the critical properties only through a boundary condition. Therefore, only the bulk contributes to the free energy, and one has  $F \cong F_b$ . Hence, the free energy  $F$  writes :

$$\frac{F}{k_B T} = c \int dz \left\{ \frac{1}{2} \left( \frac{\partial \Psi}{\partial z} \right)^2 + \frac{\tilde{\alpha}}{2} \Psi^2(z) + \frac{\tilde{w}}{4} \Psi^4(z) - (\Delta \mu) \Psi \right\}$$

with :

$$\begin{aligned} \tilde{\alpha} &= (\alpha_1 \eta^2 + \alpha_2 (1 - \eta)^2 - 2\Lambda \eta (1 - \eta)) / c \\ \tilde{w} &= (u \eta^4 + v (1 - \eta)^4 + 2\Omega \eta^2 (1 - \eta)^2) / c \\ c &= \eta^2 + (1 - \eta)^2 \end{aligned}$$

- The model is written as an effective theory in terms of the overall composition fluctuation  $\Psi = \varphi + \psi$ , and the rate of the composition fluctuation on the leaflet, , we have noted  $\eta = \varphi / \Psi$ . The chemical potential difference is denoted  $\Delta \mu$ :  $\Delta \mu = (\mu_A^i - \mu_B^i) / k_B T$ ,  $i = 1, 2$ . Explicitly  $\Psi$  and  $\eta$  write:

$$\Psi = (\phi_A^{(1)} - \phi_B^{(1)}) + (\phi_A^{(2)} - \phi_B^{(2)})$$

$$\eta = \frac{(\phi_A^{(1)} - \phi_B^{(1)})}{\Psi}$$

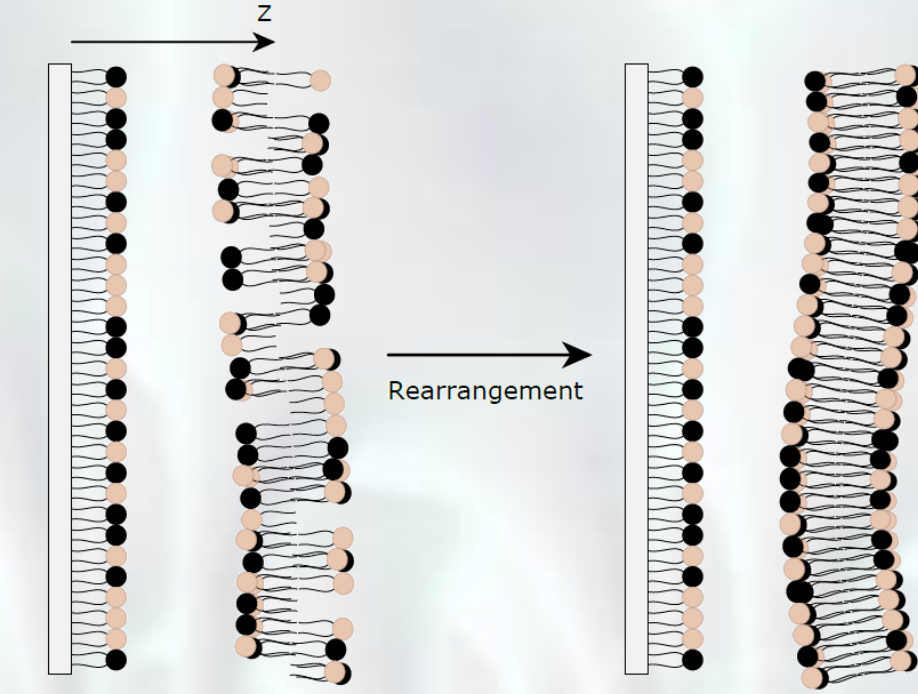


Fig.1- The fluid membrane in the presence of an *2D* -adsorbing wall.

## Parallel structure factor in the high phase temperature

- To determine the parallel structure factor in the disordered phase ( $T > T_c$ ), we determined in advance the parallel structure factor  $S(\mathbf{q}; z, z')$  which is solution of the following differential equation:

$$[\mathbf{q}^2 + \tilde{\alpha} + 3\tilde{w}\Psi^2(z) - \partial_z^2] S(\mathbf{q}; z, z') = \delta(z - z')$$

with the boundary conditions

$$S(\mathbf{q}; z, z') \Big|_{z \text{ or } z' = 0} = 0$$

$$S(\mathbf{q}; z, z') \Big|_{z \text{ or } z' = \infty} = 0$$

hereabove,  $\mathbf{q}$  is the *two* -dimensional parallel wave vector,  $z$  and  $z'$  denote the distances of both points from the surface.

- Using the standard image method, we find the exact expression of the structure factor at *zero* -scattering angle. It is given by:

$$\begin{aligned} S(z, z') &= A(z, z') \left\{ \frac{3}{4\xi_b^+} [|z - z'| - (z + z')] + \frac{1}{4} \sinh \left( \frac{[(z + z') - |z - z'| + 2l_0]}{\xi_b^+} \right) \right. \\ &\quad \left. + \tanh \left( \frac{[(z + z') - |z - z'| + 2l_0]}{2\xi_b^+} \right) - \frac{1}{4} \sinh \left( \frac{2l_0}{\xi_b^+} \right) - \tanh \left( \frac{l_0}{\xi_b^+} \right) \right\} \end{aligned}$$

where:

$$A(z, z') = \xi_b^+ \frac{\cosh \left( \frac{z + l_0}{\xi_b^+} \right) \cosh \left( \frac{z' + l_0}{\xi_b^+} \right)}{\sinh^2 \left( \frac{z + l_0}{\xi_b^+} \right) \sinh^2 \left( \frac{z' + l_0}{\xi_b^+} \right)}$$

with the following simplified notations:

$$\begin{aligned} S(z, z') &= S(\mathbf{q} = 0; z, z') \\ S(z) &= S(z, z' = z) \end{aligned}$$

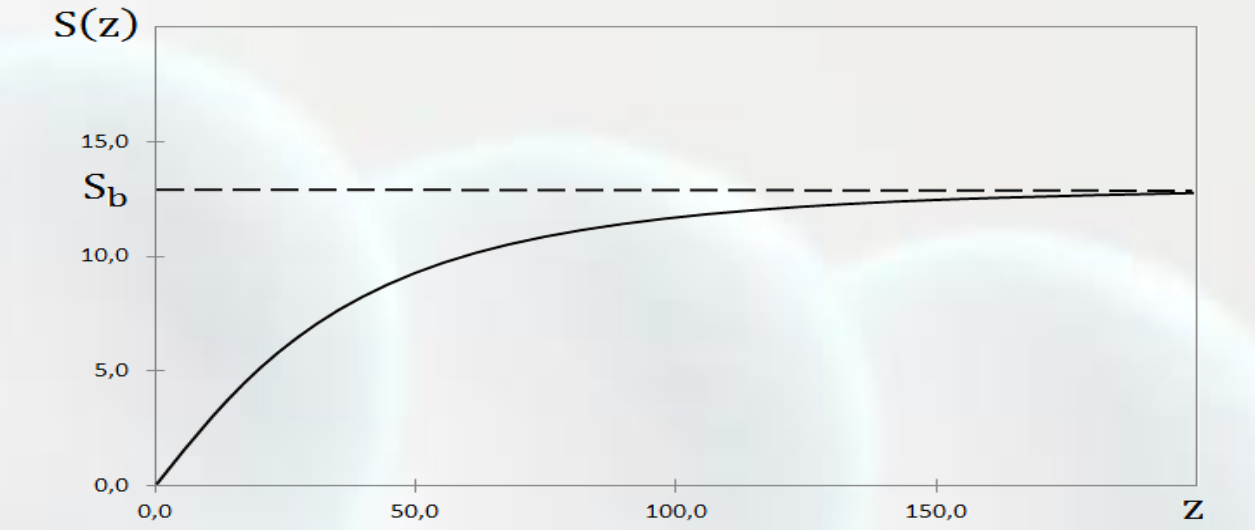


Fig.2- Plot of the local compressibility, as a function of the distance  $z$  to the surface, in the one-phase region ( $T > T_c$ ).

## Parallel structure factor in the two-phase region

- Regime ( $T < T_{comp}$ ): the parallel structure factor is given by

$$\begin{aligned} S(z, z') &= C(z, z') \left\{ 3 \left[ \frac{[(z + z') - |z - z'|]}{16\xi_b^-} \right] + \frac{1}{4} \sinh \left( \frac{[(z + z') - |z - z'| + 2l_2]}{\xi_b^-} \right) \right. \\ &\quad \left. + \frac{1}{4} \sinh \left( \frac{[(z + z') - |z - z'| + 2l_2]}{\xi_b^-} \right) + \frac{1}{4} \sinh 2 \left( \frac{[(z + z') - |z - z'| + 2l_2]}{\xi_b^-} \right) \right. \\ &\quad \left. - \frac{1}{4} \sinh \left( \frac{2l_2}{\xi_b^-} \right) - \frac{1}{32} \sinh \left( \frac{4l_2}{\xi_b^-} \right) \right\} \end{aligned}$$

where

$$C(z, z') = \xi_b^- / \left( \cosh^2 \left( \frac{z + l_2}{\xi_b^-} \right) \cosh^2 \left( \frac{z' + l_2}{\xi_b^-} \right) \right)$$

- Regime ( $T = T_{comp}$ ): in this situation, the corresponding parallel structure factor is of Dirichlet type

$$S(z, z') = \frac{1}{2\kappa} \left\{ e^{-\kappa|z - z'|} - e^{-\kappa(z + z')} \right\}, \quad \kappa = \sqrt{2\tilde{w}\Psi_0^2}$$

## Conclusions

- We have evaluated the effect of the presence of an adsorbing surface on the parallel structure factor  $S(z, z')$ , and the local compressibilities in high-phase temperature, at  $T = T_c$ , and in the two-phase region.
- In the two-phase region, the presence of a compensation temperature  $T_{comp}$ , for which one has equality between the surface and the bulk composition fluctuations  $\Psi_b = \Psi_0$ . led us to distinguish three regimes: above  $T_{comp}$ , below  $T_{comp}$ , and at  $T = T_{comp}$ .

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