

## Lateral polarization of lipid head dipoles by surface charges

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The lipid bilayers, constituting the frame of native biological membranes, are mix of neutral and charged lipids. The fundamental question is: do the surface charges induce bilayer stretching or do they induce bilayer compression? Despite to the long history of the investigation on this topic, the reply to the question is not totally clear at present.

We consider here the phenomenon which may exist in multi-component lipid bilayers consisted on charged and neutral lipids. This is the lateral polarization of the neutral lipid molecule heads by nearby surface charges. Our idea is the following. Any molecule in lipid bilayer is mobile enough to oscillate (and/or rotate) around its longitudinal axis. The neutral lipid heads possess a large electric dipole moment of about  $20 D$ . There is no preferred lateral orientation of lipid heads in pure neutral bilayer. Their orientation is arbitrary at any given moment. The time-averaged projection of head dipole on any lateral direction is equal to 0. But in charged membranes the electrostatic interaction of surface charges with the dipoles of neighboring neutral lipid molecules tends to hold the heads of the latter in such a manner that the lateral component of the head dipole is directed parallel to the charge-dipole line. Hence, the presence of the charge can induce a local lateral ordering of the neutral lipids. We call this phenomenon lateral polarization. Such an orientation results in charge-dipole attraction, and hence this phenomenon can generate local bilayer compression. The questions arise: Is the electrostatic interaction strong enough to hold the neutral lipid molecules in a given orientation for a long time? If so, when do surface charges induce membrane stretching and when do they induce membrane compression?

We have analyzed theoretically this hypothetical phenomenon and obtained that the energy of this interaction can be enough strong to hold these molecules in a certain thermally averaged orientation. The polarization results in local bilayer compression. Each surface charge induced the formation of microclusters with mechanical characteristics different from those of other parts of the bilayer. We demonstrated that the depth of charge immersion inside the bilayer interface is key parameters determining whether polarization effects dominated over long-range electrostatic repulsion. The

polarization effects are stronger if the charge is located deeper in the bilayer. Probably, this explains the difference between positive and negative surface charges effects. The negative charges correspond to shallow surface charges. They do not induce the polarization. On the contrary, the positive charges correspond to deeply immersed charges. They induce strong polarization.

As an application of our theory we compared our results with published data for DMTAP/DMPC mixed bilayers, which can not be explained in framework of the classical membrane electrostatics. The dependence of averaged area per molecule and chain melting temperature (Fig.1) on mole fraction of DMTAP were considered, and the theoretical predictions obtained in the present study were consistent with these data.

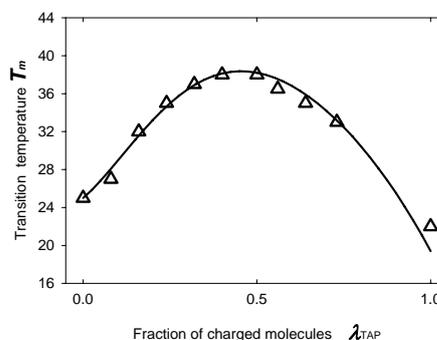


Fig.1 Chain-melting transition temperature of a DMTAP/DMPC mixed bilayer as a function of the DMTAP mole fraction. Triangles correspond to experimental data from Zantl, R., Baicu, L., Artzner, F., Sprenger, I., Rapp, G., J.O. Raldler, J.O. J. Phys. Chem. B. 1999, 103, 10300. The solid curve shows theoretical calculations.