SUPPLEMENTARY INFORMATION

Self-consistent field Modelling of mesomorphic Phase changes of Monoolein and Phospholipids in response to additives.

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1 Isotherms

In the three-component systems used in this paper, that is in bilayers in which additives are included, the amount of additive in the system is a free parameter. This means that one can make so-called absorption isotherms, that is the amount of additive that is absorbed in the membrane per unit area θ^σ as a function of the bulk concentration of this additive. Here $\theta^\sigma = \sum_z \varphi_2(z) - \varphi_2^b$. For good reasons we are typically interested in absorbed amounts which keeps the additive as a minority component in the membrane. That is why we in most cases below the isotherms were discontinued as soon as either the bulk concentration or the concentration of the component in the membrane passed some threshold value.

Absorption isotherms are informative about (hidden) solubility limitations that might pop up in cases where the additive my have some solubility gap with the aqueous solution. In such a case the absorption isotherms will feature some type of Van der Waals loop. Below we avoid these regions and always keep the amount of additive below the bulk binodal value. These constraints do not pose major restrictions, because our primary interest is in predicting (initial) trends in how additives may push a membrane system towards potential phase changes, rather than to pinpoint a threshold composition for such mesomorphic variation or where the membrane falls apart (e.g. something which happens when large amounts of Triton is used). The slope of this absorption isotherm can be related to a membrane partition coefficient[?] , but we will not dwell on this quantity.

The absorption isotherms for the three alcoholic solvents used in this study are given in figure S1.

Intuitively one would expect isotherms to show positive accumulated amounts as a function of the concentration of the additive. However for rather hydrophilic compounds that accumulate weakly in the head group or glycerol-backbone region, may show a strong depletion in the core region, rendering the overall absorption to be negative. Inspection of Fig. S1 this is the case for butanediol and for ethanol for both bilayers. Only for t-butanol modest positive absorbed amounts are predicted. For the MO system, we could not exceed the limit of $\varphi^b = 0.1$ for butanol, as no bilayer could be formed anymore, consistent with experimental facts that butanol may easily destroy a bilayer and put the system into a fluid isotropic phase (L_2)[?] . For DOPC we could reach higher levels of φ^b and see the curve reaching a plateau before the same happens. Again our interest is not in the disintegration of bilayers and therefore we have set the limit in added butanol for the remainder of our calculations at $\varphi^b = 0.1$.

In figure S2 we have plotted the absorption isotherms of all FA used. As can be seen from the figure, we observe relative linear positive trends. This indicates that FA inserts themselves into the bilayer and that we managed to stay away from the solubility limit or CMC of the fatty acids. Clearly FA partition strongly in the bi-

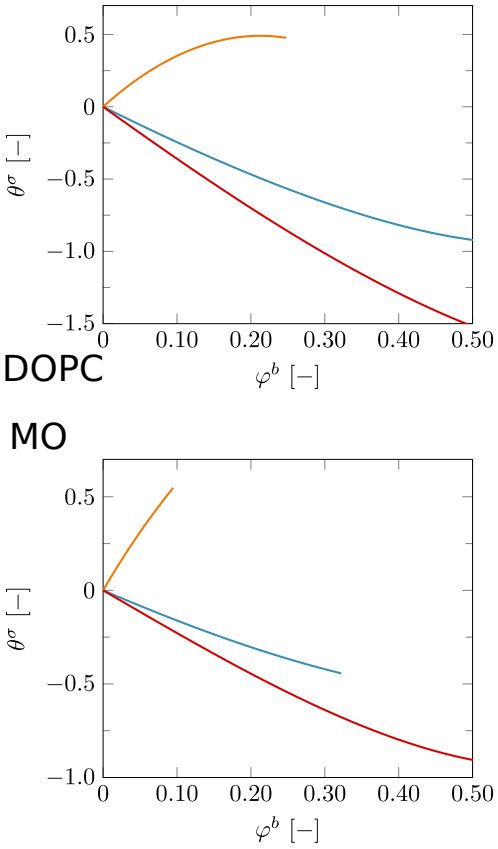

Figure S1 Isotherm (θ^σ as a function of φ^b) for DOPC bilayers (top) or MO bilayers (bottom) with added ethanol C_2O (blue), added t-butanol tC_4O (orange) or added butanediol (C_4O_2) (red). A) $\bar{\kappa}$; B) κ; C) J_0^m.

layers. This must of course be attributed to the hydrophobic tails that have a high affinity for the core. This preferential partitioning of the FA in the membrane is a strong function of the length of the tail and therefore the adsorption isotherms show an increase in adsorbed amount at progressively lower bulk concentrations the longer is the tail length.

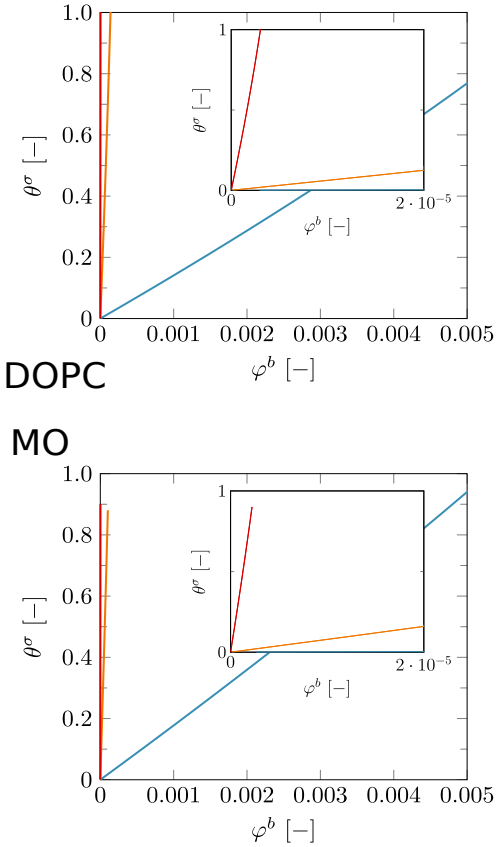

Figure S2 Isotherm (θ^σ as a function of φ^b) for DOPC bilayers (top) or MO bilayers (bottom) with added fatty acids. blue: C_8O_2; orange: $C_{12}O_2$; red: $C_{16}O_2$. The inset provides the same graph but in which the x-axis is zoomed