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Lipid membrane phase transitions involve structural redistribution of interfacial water

Morphological transitions of lipid membranes are thought to depend primarily on the chain length diversity of its components. However, the role of different lipid head groups, their counter ions and hydrating water molecules in the close proximity of the membrane on the gel-to-liquid phase transition of lipid is mostly unknown. We employed second harmonic (SH) scattering measurements to probe asymmetric distribution of water molecules between the lipid leaflets. Measurements on the same acyl chain length and single lipid component liposomes consisting of 1,2-dimyristoyl-sn-glycero-3-phosphate (sodium salt) (DMPA), 1,2-dimyristoyl-sn-glycero-3-phospho-L-serine (sodium salt) (DMPS), 1,2-dimyristoyl-sn-glycero-3-phospho-(1'-rac-glycerol) (sodium salt) (DMPG), 1,2-dimyristoyl-sn-glycero-3-phosphocholine (DMPC) and 1,2-dimyristoyl-3-trimethylammonium-propane (chloride salt) (DMTAP) showed a significant lipid hydration alteration as a function of temperature. More specifically, we observed a $> 20\%$ increase in the second harmonic intensity at the phase transition temperature for liposomes made of charged lipids. Zwitterionic DMPC lipids on the other hand, display only smaller ($\sim 8\%$) changes. This data implied that an increasing number of water molecules anisotropically oriented towards surface normal above the phase transition and the charge state of the lipids play key role on the gel-to-liquid phase transition. Our data demonstrate that lipid phase transitions not only lead to structural changes in the bilayer itself but also in the adjacent aqueous phase.

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