Interfacial Structure and Hydration of Model Membranes Elucidated with Non-Linear Scattering Techniques

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Nature employs surfaces in contact with aqueous environments as a hub for a variety of biologically important chemical processes, from antibody-antigen recognition to various enzymatic reactions. The molecular mechanisms by which such processes take place have been an ongoing scientific challenge. Recently developed, state-of-the-art, non-linear scattering methods enable in situ and label-free monitoring of interfacial processes occurring at buried aqueous solutions. In this talk I will focus on two biologically relevant systems. In the first, the molecular structure of 3D phospholipid monolayers is elucidated as a model system for adiposome organelles. We find that acyl chain length, saturation, and number of tails per lipid have substantial influences on the molecular structure. Namely, shortening the lipid tail length, having unsaturated bonds, or using single chained lipids result in more disorder. Nevertheless, the phospholipid headgroup orientation seems to remain unchanged, and stays approximately parallel to the nanodroplet interface. Remarkably, the single tailed lyso-PC lipids form more diluted and “patchy” 3D monolayers.[1,2,3] In the second part, the transmembrane asymmetry of small unilamellar liposomes consisting of zwitterionic and charged lipids in aqueous solution is characterized. For single component liposomes, transmembrane asymmetry is present only for the charge distribution and lipid hydration, but the leaflets are not detectably asymmetric in terms of the number of lipids per leaflet, even though geometrical packing arguments would predict so. However, lipid transmembrane asymmetry is induced in binary lipid mixtures under conditions that enable intermolecular hydrogen-bonding interactions between phosphate and amine groups. In this case, the measured asymmetry consists of a different number of lipids in the outer and inner leaflet, a difference in transmembrane headgroup hydration, and a different headgroup orientation for the interacting lipid headgroups.[4]

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