Ion specificity on the dynamics of ion-induced surface charge asymmetry of freestanding lipid membranes

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The distribution and flow of charges in biological membranes dictate the electrostatic environment essential for the activity of fundamental membrane processes such as signaling and transport in cells. These membranes reside in an aqueous environment and are stable because of the hydrating water that drives the membrane structure intact. The presence of charges on the membrane orients the hydrating water through electrostatic charge-dipole interaction. We use this universal phenomenon to image the orientational ordering of water molecules with a non-resonant optical second harmonic microscope[1]. Second harmonic generation is a coherent method that provides contrast only when a material possesses spatial non-centrosymmetry[2]. Differences in surface charge between the bilayer leaflets lead to non-centrosymmetric ordering of water molecules, thus, providing second harmonic contrast. As such, SHG imaging is sensitive to the asymmetric distribution of surface charges, and concomitantly, the asymmetric distribution of membrane potentials between bilayer leaflets. In this work, we demonstrate this imaging method and investigate the affinity of specific ions to induce asymmetric surface charge distribution in freestanding lipid bilayer membranes. The images provide insights in both the spatial and temporal distribution of ions, and on how different ions can affect the electrostatic membrane environment that cannot be explained by simple electrostatic interaction alone.

Keywords: interfaces, membrane potentials, surface charges, aggregation, membrane asymmetry, freestanding lipid membranes

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