

Water Hydrogen-Bonding Dynamics at the Lipid Multibilayer Surface Depending on Chemical Structure of Lipids: Femtosecond Mid-IR Pump-Probe Spectroscopy

Membrane water is a potential reaction site for many biochemical reactions. Therefore, a molecular level understanding of water structure and dynamics that strongly depend on the chemical structure of lipid is prerequisite for elucidating the role of water in biological reactions on membrane surface. Here, we study water structure and dynamics depending on head part of the lipid molecules by femtosecond mid-IR pump-probe spectroscopy with OD stretch mode of HOD molecules at the lipid multibilayer. We observed two significantly different vibrational lifetime components (very fast 0.5 ps and slow 1.9 ps) at the negatively charged lipid multibilayer whereas only one vibrational lifetime component (1.6 ps) was observed at the positively charged lipid multibilayer. From the detailed mechanistic analyses and comparisons of vibrational energy and rotational relaxations of water at multibilayers of lipids with and without phosphate group, the role of phosphate group in structuring water molecules at phospholipid membrane is revealed.