A new method of X-ray solvent contrast modulation was developed to visualize lipid bilayers in crystals of membrane proteins at a high enough resolution to resolve individual phospholipids molecules (~3.5 Å). Visualization of lipid bilayer has been escaping from conventional crystallographic methods due to its extreme flexibility, and our knowledge on the behavior of lipid bilayer is still very much limited. Here we applied the new method of X-ray solvent contrast modulation to crystals of Ca2+-ATPase in 4 different physiological states. As phospholipids have to be added to make crystals of Ca2+-ATPase, it is expected that lipid bilayers are present in the crystals. Moreover, transmembrane helices of Ca2+-ATPase rearrange drastically during the reaction cycle and some of them show substantial movements perpendicular to the bilayer plane. Thus these crystals provide a rare opportunity to directly visualize phospholipids interacting with a membrane protein in different conformations. Complete diffraction data covering from 200 to 3.2 Å resolution were collected at BL41XU, Spring-8, using an R-Axis V imaging plate detector for crystals soaked in solvent of different electron density. A new concept “solvent exchange probability”, which should be 1 in the bulk solvent, 0 inside the protein and an intermediate at interface, was introduced and used as a restraint for real space phase improvement. The electron density maps thus obtained clearly show that: (i) Phospholipid molecules surrounding the protein are fixed apparently by Arg/Lys-phosphate salt bridges or Trp-carbonyl hydrogen bonds and follow the movements of transmembrane helices. Movements of as large as 12 Å are allowed. (ii) If the movement of a transmembrane helix exceeds this limit, associated phospholipids change the partners for fixation or change the orientation of the entire protein molecule.

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