CHIRAL SUM FREQUENCY GENERATION SPECTROSCOPY REVEALS HOW MIRROR-IMAGE β-SHEETS ORGANIZE WATER SUPERSTRUCTURES WITH OPPOSITE CHIRALITY

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The structure and function of biomacromolecules depend on water structures in the hydration shell. However, elucidating the architectures of hydration shells around biomacromolecules has proven challenging. We used heterodyne chiral vibrational sum frequency generation (SFG) spectroscopy to study the structure of the solvation shell around an antiparallel β-sheet protein.

Heterodyne chiral SFG spectroscopy reveals that water molecules form chiral superstructures around the protein, and that these water superstructures are induced by the chirality of the protein. Enantiomeric (L-) or (D-) proteins give rise to enantiomeric water superstructures. Molecular dynamics simulations expose the chiral architecture of the water superstructures around the protein. Modeling the chiral SFG response of the hydration shell suggests that hydrogen bonding between water and protein is a driving force behind the formation of the chiral water superstructures.

Our work demonstrates the promise of chiral SFG spectroscopy to reveal structures and dynamics of hydration shells around biomacromolecules. We speculate on the involvement of chiral water superstructures in biomolecular recognition, and the role of chiral water superstructures in the origins of biological homochirality.