MAPPING HYDRATION DYNAMICS AND COUPLED WATER-PROTEIN FLUCTUATIONS AROUND A PROTEIN SURFACE

LUYUAN ZHANG, LIJUAN WANG, YA-TING KAO, WEIHONG QIU, YI YANG, OGHAGHARE OKOBIAH, and DONGPING ZHONG, *Departments of Physics, Chemistry, and Biochemistry, Programs of Biophysics, Chemical Physics, and Biochemistry, 191 West Woodruff Avenue, The Ohio State University, Columbus, OH 43210*.

Elucidation of the molecular mechanism of water-protein interactions is critical to understanding many fundamental aspects of protein science, such as protein folding and misfolding and enzyme catalysis. We recently carried out a global mapping of protein-surface hydration dynamics around a globular α-helical protein apomyoglobin. The intrinsic optical probe tryptophan was employed to scan the protein surface one at a time by site-specific mutagenesis. With femtosecond resolution, we mapped out the dynamics of water-protein interactions with more than 20 mutants and for two states, native and molten globular. A robust bimodal distribution of time scales was observed, representing two types of water motions: local relaxation and protein-coupled fluctuations. The time scales show a strong correlation with the local protein structural rigidity and chemical identity. We also resolved two distinct contributions to the overall Stokes-shifts from the two time scales. These results are significant to understanding the role of hydration water on protein structural stability, dynamics and function.