Abstract Submitted for the MAR06 Meeting of The American Physical Society

Ultrafast Vibrational Dynamics of Water Interfaces Probed by Sum-frequency Vibrational Spectroscopy¹ JOHN A. MCGUIRE, Y.R. SHEN, Department of Physics, University of California and Material Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720 — Using sumfrequency generation (SFG) as a surface-specific probe, we measured the ultrafast population and dephasing dynamics of excitations of OH-stretch modes of water at fused-silica/water interfaces. A tunable femtosecond IR pulse was used to pump the vibrational mode. Another independently tunable femtosecond IR pulse, after a time delay, was used to probe the vibrational excitation and then up-converted by SFG for detection. Experiments on the hydrophilic fused-silica/water interface reveal population relaxation of the hydrogen-bonded OH modes on time scales of 200-500 fs, more rapid on the red side than the blue side. The width of transient spectral holes produced by the IR pump pulse correspond to homogeneous dephasing times of ~ 50 fs. Measurements of the dangling OH bonds at the hydrophobic silica/n-octadecyltrichlorosilane/water interface reveal a population relaxation time of about 1.2 ps.

¹The work was supported by DOE

John A. McGuire Dept. of Physics, Univ. of California and LBNL

Date submitted: 06 Dec 2005

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