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Spectrally- and Time-Resolved Sum Frequency Generation (STiR-SFG): a new tool for ultrafast hydrogen bond dynamics at interfaces. ALEXANDER BENDERSKII, ANDREY BORDENYUK, CHAMPIKA WEERAMAN, Wayne State University — The recently developed spectrally- and time-resolved Sum Frequency Generation (STiR-SFG) is a surface-selective 3-wave mixing (IR+visible) spectroscopic technique capable of measuring ultrafast spectral evolution of vibrational coherences. A detailed description of this measurement will be presented, and a noniterative method or deconvolving the laser pulses will be introduced to obtain the molecular response function. STiR-SFG, combined with the frequency-domain SFG spectroscopy, was applied to study hydrogen bonding dynamics at aqueous interfaces (D_2O/CaF_2). Spectral dynamics of the OD-stretch on the 50-150 fs time scale provides real-time observation of ultrafast H-bond rearrangement. Tuning the IR wavelength to the blue or red side of the OD-stretch transition, we selectively monitor the dynamics of different sub-ensembles in the distribution of the H-bond structures. The blue-side excitation (weaker H-bonding) shows monotonic red-shift of the OD-frequency. In contrast, the red-side excitation (stronger H-bonding structures) produces a blue-shift and a recursion, which may indicate the presence of an underdamped intermolecular mode of interfacial water. Effect of electrolyte concentration on the H-bond dynamics will be discussed.

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