Development of Ultra-broadband Infrared Optical Parametric Sources and Applications in Ultrafast Vibrational Spectroscopy & Dynamics of Interfaces

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Ultrashort IR pulses are desirable for many scientific applications such as multidimensional IR spectroscopy, high-harmonic generation and the production of attosecond pulses, but also for ultrafast vibrational spectroscopy and coherent control of vibrationally mediated processes. Exploiting our understanding of non-collinear optical parametric amplification (NOPA), we have extended NOPA to new classes of optical materials and developed ultrabroadband sources with bandwidths approaching 90 THz (3000 cm⁻¹) in the near IR,[1] and are currently pushing this into the mid-IR.[2]

A primary application of ultrabroadband sources in our laboratory is to understand the properties and behavior of water at interfaces.[3, 4] In particular, it is not clear what effects the presence of the surface, the charge that can develop on the surface, the solution ionic strength, and the interfacial electric field, have on how interfacial water molecules communicate with each other, e.g., how thermal (vibrational) energy flows. To address these issues we have investigated the ultrafast vibrational population and dephasing dynamics of the O-H stretch using IR pump-vibrational Sum Frequency Generation (vSFG) probe at the water/mineral interfaces.[4] Contrary to previous reports, the vibrational lifetime of the O-H stretch at the silica/water interface is ~ 600 fs, a factor 2-3 slower than bulk water, when the surface is neutral.[5] Charging the SiO₂ interface appears to lead to a dramatic acceleration of vibrational relaxation. Experiments on the effect of ionic strength, suggest that the primary reason for accelerated dynamics at pH=6 is the sampling of water within the Debye length that has bulk-like solvation.[6]

References: