

Ultra-broadband & Ultrafast Vibrational Spectroscopy & Dynamics of Mineral/Aqueous Interfaces

Professor Eric Borguet (Department of Chemistry, Temple University, Philadelphia, USA)



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The properties and behavior of water at interfaces is important in many fields. However, 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 dynamics of the O-H stretch using IR pump-vibrational Sum Frequency Generation (vSFG) probe at the water/silica interface. 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. Charging of the interface appears to lead to a dramatic acceleration of vibrational relaxation. Experiments on the effect of ionic strength, at pH=6 suggest that the primary reason for the accelerated dynamics at the charged surface is the sampling of water that has bulk-like solvation within the Debye length.

A newly developed SFG spectrometer, based on a novel ultrabroadband optical parametric amplifier (OPA) generating IR pulses in the ~ 2800 - 6000 cm^{-1} range bandwidths >2000 cm^{-1} in the near-IR range will be presented. This allows vSFG spectroscopy of silica/water interfaces over a wide frequency range, including low-intensity features such as:

- 1) non-hydrogen bonded OH vibrations at hydrophilic silica/water interfaces.
- 2) combination [stretch+bend] bands of interfacial water appearing at ~ 5000 - 5200 cm^{-1} .
- 3) overtones of OH stretching modes at silica/water interfaces.

Access to these modes opens up new opportunities for investigations of a broad range of interfaces.

問合せ先: 触媒化学研究センター・叶深 (ye@cat.hokudai.ac.jp/011-706-9126)