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Sensitivity of ultrafast vibrational dynamics of interfacial water to cations at silica/water interfaces SHALAKA DEWAN, AASHISH TULADHAR, ERIC BORGUET, Temple Univ — Interfacial water structure is a key player in the chemistry of environmentally relevant processes, e.g. the dissolution of mineral surfaces. Since the properties of water are dominated by its hydrogen bonding network, understanding the local changes in this structure at the mineral/water interface is important. Vibrational sum-frequency generation (vSFG) spectroscopy is a tool that can probe the hydrogen bonding environment at buried interfaces. Time-resolved vSFG of the OH stretch is further sensitive to the changes in hydrogen bonding at surfaces. vSFG has demonstrated that interfacial water structure at silica is most sensitive to cations at neat water pH 6-8. Further, at this pH, increasing the ionic strength slows the vibrational relaxation of O-H stretch of water from ~ 200 to ~ 600 fs. Here, we show that the slowing down of O-H dynamics on addition of cations is more pronounced at pH 6 than at pH 12, even though the surface is negatively charged in both cases. This suggests that, in addition to the salt screening the surface charge, the cations actually change the local H-bonding environment of water differently at distinct bulk pH. Our results shed light on the ongoing debate on the role of ions in altering the structure of water at an interface.

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