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Frontiers in Geochemistry
Seminar Series

Ions and the ultrafast spectroscopy and dynamics at aqueous interfaces

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Interfacial water structure, which can be probed by vibrational sum-frequency generation (vSFG) spectroscopy, is key to many processes. Time-resolved vSFG shows that in the absence of surface charge (pH 2), water at silica surfaces exhibits significantly slower OH stretch vibrational relaxation (~600 fs) compared to bulk. However, at charged silica surfaces (e.g., pH 6), bulk-like fast dynamics (~200 fs) are observed at low ionic strength. This decelerates to ~600 fs, with the addition of NaCl. In parallel, vSFG results demonstrated that silica interfacial water structure is most sensitive to cations at pH=6-8. Consequently, it is unclear whether the observed slowing of the vibrational dynamics is due to the reduction in Debye length, or because of changes in the local hydrogen bonding environment caused by the electrolyte and how this might depend on the identity of the ions. Our results shed light on the ongoing debate on the role of ions in interfacial water structure and whether the observed behavior is specific to silica/water interfaces or can be generalized to other systems.

Host: Zheming Wang, 371-6349

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