



Contribution ID: 34

Type: Poster

Vibrational Relaxation of water at the Ice – Air Interface

Wednesday, January 10, 2018 6:10 PM (3h 5m)

The surface of ice is relevant for various important phenomena including glacier sliding, and (photo-) chemical conversion of molecules on that surface.

For chemical reactions occurring on the ice surface, the dynamics of the surface water molecules and energy flow pathways play an important role: following a chemical reaction, the rate of excess energy dissipation determines the probability of a back-reaction occurring. Here we elucidate the relaxation dynamics of water molecules at the surface of single crystalline hexagonal ice after exciting the O-H stretch vibration. We compare our results with the relaxation dynamics of the liquid water-air interface.

To study the vibrational dynamics we apply time-resolved sum frequency generation spectroscopy (SFG). With this method we can specifically study the outermost molecular water layer at the ice-air interface and follow the relaxation dynamics of those interfacial water molecules following vibrational excitation. Time-resolved SFG is a laser-based spectroscopic technique that involves vibrationally exciting a fraction of surface molecules with a femtosecond infrared excitation pulse, and probing the dynamics of the interfacial molecules with the sum frequency generation process using a visible and an infrared pulse resonant with the O-H stretch vibrations. This detection scheme ensures that only the interfacial water molecules contribute to the signal, and the use of ~ 50 fs laser pulses ensures high time resolution in the experiments.

Our results show that the relaxation dynamics of the O-H stretch mode after excitation at 3100 cm^{-1} at the basal plane of ice is around 70 fs. This is three times faster than for the water-air interface. A potential reason for the faster relaxation could be the stronger hydrogen bonding of water molecules on the ice surface, compared to water. We discuss the implications of this fast energy dissipation in terms of more effectively catalyzing reactions on ice than on water.

Significance statement

We determine the vibrational energy relaxation of water molecules at the ice-air interface and compare it to the water-air interface. Vibrational energy relaxation rates reflect the local H-bonding network of water molecules. Our results shed light on interfacial energy dissipation, relevant for atmospheric catalytic reactions.

Primary author: Ms SUDERA, Prerna (PhD student)

Co-authors: Dr SÁNCHEZ, Alejandra (Max Planck Institute for Polymer Research); BACKUS, Ellen (Max Planck Institute for Polymer Research); Dr CYRAN, Jenee (Max Planck Institute for Polymer Research); Prof. BONN, Mischa (Max Planck Institute for Polymer Research)

Presenter: Ms SUDERA, Prerna (PhD student)

Session Classification: Poster Session & Apéro Riche (apéro dîner)