Interfacial vibrational dynamics of ice I_h and liquid water

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Besides the potential importance for atmospheric chemistry, vibrational energy dynamics on aqueous surfaces are of fundamental interest for understanding the surface and interfaces of ice. Here, we study the vibrational dynamics of surface OH groups on single-crystalline hexagonal ice (Ice I_h). OH-oscillators are selectively excited using femtosecond infrared pulses at different frequencies. The effect of excitation of the interfacial molecules is probed using the surface-specific method sum-frequency generation (SFG) spectroscopy. A comparison to liquid water surfaces, reveals accelerated vibrational energy relaxation and dissipation at the ice surface for hydrogen-bonded OH groups. In contrast, free-OH groups sticking into the vapor phase exhibit substantially slower vibrational dynamics on ice.