

# **Molecular vibrational response of ice layers after ultrashort-laser excitation of metal surfaces**

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Electrons as excess charges in polar solvents result in molecular motion within the solvent to stabilize the new charge distribution known as electron solvation. We employ time-resolved broadband-IR-VIS SFG (sum frequency generation) spectroscopy as a powerful technique to study the changes in the vibrational spectra of D<sub>2</sub>O water layers upon electron injection from a Ru(001) surface. Unlike in time-resolved photoemission experiments where the electron binding energy is traced and the solvent response is inferred subsequently, SFG vibrational spectroscopy *directly* monitors the molecular reorientation induced by the excess charge. If the Ru surface is excited with 266-nm (4.65 eV) photons, excess electrons injected into the adsorbate layer induce –dependent on layer morphology and layer thickness– drastic changes in the vibrational spectra of the OD stretch vibration. While in amorphous ice layers only weak changes are observed, a SFG signal increase by several orders of magnitude is found in crystalline D<sub>2</sub>O layers (enhancement factor of 10<sup>3</sup> up to 10<sup>4</sup>). The physical and chemical processes involved will be discussed.