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Achintya Kundu, Fabian Dahms, Benjamin P. Fingerhut, Erik T.J. Nibbering, Ehud Pines, Thomas Elsaesser

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Ultrafast Vibrational Relaxation and Energy Dissipation of Hydrated Excess Protons in Polar Solvents

Achintya Kundu[†], Fabian Dahms[†], Benjamin P. Fingerhut[†], Erik T. J. Nibbering[†]

Ehud Pines[‡], and Thomas Elsaesser^{*,†}

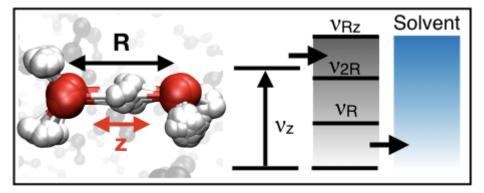
[†]Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Max-Born-Str. 2a, 12489 Berlin, Germany

[‡]Department of Chemistry, Ben Gurion University of the Negev, Beer-Sheva 84105, Israel

Vibrational relaxation and energy redistribution of Zundel cations $H_5O_2^+$ in acetonitrile and neat water are studied by femtosecond infrared pump-probe spectroscopy with resonant excitation of the 1200 cm⁻¹ proton transfer mode. Transient spectra in a range from 1000 to 2200 cm⁻¹ reveal intermode anharmonic couplings and a sub-100 fs decay of the proton transfer mode, followed by femtosecond vibrational redistribution within the cations' vibrational manifold and picosecond energy transfer to the solvent. Energy transfer to an aqueous environment occurs on the same time scale as within neat water, demonstrating the strong coupling of cations and water solvation shell.

Keywords: Hydrated excess proton, solvation, vibrational relaxation, femtosecond infrared spectroscopy

*Corresponding author. Phone +49 30 63921400, email address elsasser@mbi-berlin.de



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