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Ultrafast memory loss and energy redistribution in the hydrogen bond network of liquid H2O.

Cowan ML, Bruner BD, Huse N, Dwyer JR, Chugh B, Nibbering ET, Elsaesser T, Miller RJ.

Department of Chemistry, University of Toronto, 80 St George Street, Toronto, Ontario, Canada M5S3H6.

Abstract

Many of the unusual properties of liquid water are attributed to its unique structure, comprised of a random and fluctuating three-dimensional network of hydrogen bonds that link the highly polar water molecules. One of the most direct probes of the dynamics of this network is the infrared spectrum of the OH stretching vibration, which reflects the distribution of hydrogen-bonded structures and the intermolecular forces controlling the structural dynamics of the liquid. Indeed, water dynamics has been studied in detail, most recently using multi-dimensional nonlinear infrared spectroscopy for acquiring structural and dynamical information on femtosecond timescales. But owing to technical difficulties, only OH stretching vibrations in D_2O or OD vibrations in H_2O could be monitored. Here we show that using a specially designed, ultrathin sample cell allows us to observe OH stretching vibrations in H_2O. Under these fully resonant conditions, we observe hydrogen bond network dynamics more than one order of magnitude faster than seen in earlier studies that include an extremely fast sweep in the OH frequencies on a 50-fs timescale and an equally fast disappearance of the initial inhomogeneous distribution of sites. Our results highlight the efficiency of energy redistribution within the hydrogen-bonded network, and that liquid water essentially loses the memory of persistent correlations in its structure within 50 fs.

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