Real-Time Tracking of the intramolecular vibrational dynamics of liquid water

Gaia Giovannetti[1], Sergey Ryabchuk[1,2], Ammar Bin Wahid[1], Hui-Yuan Chen[3], Giovanni Batignani[4], Erik P. Månsson[1], Oliviero Cannelli[1], Emanuele Mai[4], Andrea Trabattoni[1,5], Ofer Neufeld[6], Angel Rubio[1,7], Vincent Wanie[1], Hugo Marroux[8,*], Tullio Scopigno[4,*], Majed Chergui[3,9,*], and Francesca Calegari[1,2,10,*]

[1]Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany

[2]The Hamburg Centre for Ultrafast Imaging, Universität Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany

[3]Lausanne Centre for Ultrafast Science (LACUS), Ecole Polytechnique Fédérale de Lausanne, ISIC, FSB, Station 6, CH 1015 Lausanne, Switzerland

[4]Dipartimento di Fisica, Università di Roma" La Sapienza", Roma, I-00185, Italy

[5]Institute of Quantum Optics, Leibniz Universität Hannover, 30167 Hannover, Germany

[6]Schulich Faculty of Chemistry, Technion – Israel Institute of Technology, 32000, Haifa, Israel

[7]Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg, Germany

[8]Laboratoire Interactions, Dynamiques et Lasers, CEA-Saclay, 91191 Gif-sur-Yvette, France

[9]Elettra - Sincrotrone Trieste S.C.p.A., S.S. 14 Km 163, 5 in Area Science Park, I 34149 Trieste, Italy

[10]Physics Department, Universität Hamburg, Luruper Chaussee 149, 22761, Hamburg, Germany

ABSTRACT

Water's polarity and hydrogen-bond network give rise to its unique chemical and biochemical behaviour. Its vibrational motions, occurring on a few-femtosecond timescale, govern ultrafast energy transfer within the hydrogen-bond network. However, direct real-time observation of these motions has remained elusive due to the extreme temporal resolution required.

In this contribution, we present the ground-state vibrational dynamics of liquid water initiated by a sub-5 fs near-infrared (NIR) pump pulse via Impulsive Stimulated Raman Scattering (ISRS). Using few-fs ultraviolet (UV) probe pulses transmitted through a 5 µm-thick liquid jet, we monitor the coherent vibrational wave packet dominated by the OH stretch mode, exhibiting a 10 fs oscillation period and a 25 fs damping time. These results reveal the rapid dephasing of the OH stretch mode preceding its relaxation through coupling to the bending vibrations, highlighting the importance of intermolecular couplings of liquid water in the high frequency vibrational dynamics.