Probing Chemical Systems with Two-Color High-Harmonic and X-ray Spectroscopy

Zhong Yin,[1] Tran Trung Luu,[1] Stephan Thürmer,[2] Sebastian Malerz,[3] Marvin Pohl,[3] Evanthia Chatzigeorgiou,[4] Yitao Cui,[5,6] Kosuke Yamazoe,[5,6] Jun Miyawaki, [5,6] Takashi Tokushima,[7] Conny Sathe,[7] Johan Grasjö,[4] Yoshihisa Harada,[5,6] Bernd Winter,[3] Jan-Erik Rubensson,[4] Hans Jakob Wörner[1]

[1]*Laboratorium für Physikalische Chemie, ETH Zürich, 8093 Zürich, Switzerland*
[2]*Department of Chemistry, Kyoto University, Kyoto 606-8502, Japan*
[3]*Molecular Physics Department, Fritz-Haber-Institute of the Max-Planck-Society, 14195 Berlin, Germany*
[4]*Department of Physics and Astronomy, Uppsala University, P.O. Box 516, SE-751 20 Uppsala, Sweden*
[5]*Institute for Solid State Physics, The University of Tokyo, Kashiwanoha, Chiba 277-8581, Japan*
[6]*Synchrotron Radiation Research Organization, The University of Tokyo, Hyogo 679-5198, Japan*
[7]*MAX IV Laboratory, Lund University, Box 118, SE-221 00 Lund, Sweden*
[8] Department of Pharmacy, Uppsala University, P.O. Box 580, SE-751 23 Uppsala, Sweden

Liquid water is essential for the existence of life. Despite enormous efforts, there is still an incomplete understanding of the interaction of water molecules with each other and with ions.[1] In this work, we present a combined study of liquid water and aqueous solutions utilizing state-of-the-art high harmonic and high-resolution Resonant Inelastic X-ray Scattering (RIXS) spectroscopy in liquid phase which might help to corroborate current understanding.

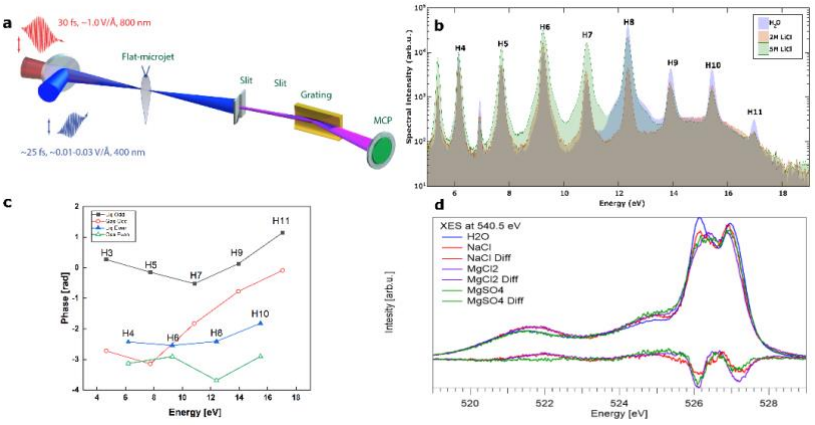

Fig. 1: a) Schematic experimental setup for two-color HHG from liquid samples. b) HHG spectrum of pure liquid water in comparison to $LiCl_{(aq)}$ solutions. c) Distinctively, relative phase difference for even and odd harmonics of liquid and gas phase water. d) RIXS data of liquid water and various aqueous salt solutions.

High harmonic generation (HHG) enabled the study of electronic dynamics on their natural time scale.[2] In conventional HHG from inversion-symmetric gases, the multi-cycle laser pulses give rise to only odd harmonics. Adding the second harmonic of the fundamental pulses perturbs the electron's trajectories, alters the interference between different cycles and thereby permits the emission of even harmonics. Thus, two-color experiments of this type have enabled the possibility of studying and controlling the HHG process in fine details.[3] Here, we show that this approach can be extended to the liquid phase using the flat micro-jet,[4] to elucidate electronic processes in liquids in real time. Preliminary analyses show that the relative phase between the odd and even harmonics in liquids is distinctively different from the gas phase see Fig. 1c, suggesting a complex mechanism compared to well-known gas phase behavior. This two-color approach allows mapping the dynamical electronic properties of the aqueous solutions with unprecedented time resolution. With RIXS, a photon-in-photon out method, the bonding characteristics of a system are probed. Our recent high-resolution RIXS results reveal strong perturbation of the electronic structure induced by the presence of specific ions, ranging from weak to strong hydrated ions. We anticipate that our works would contribute in elucidating the water–water and water–ion interaction on the molecular level.

[1] F. Perakis, et al, Chem. Rev. (2015). [2] P. B. Corkum et al. Nat. Phys. 3, 381 (2007). [3] N. Dudovich et al.,Nat. Phys., 2, 781 (2006). [4] T.T. Luu, Z. Yin et al., Nat. Com. 9, 3723 (2018).