Looking at molecules from two sides with table-top femtosecond soft-X-rays

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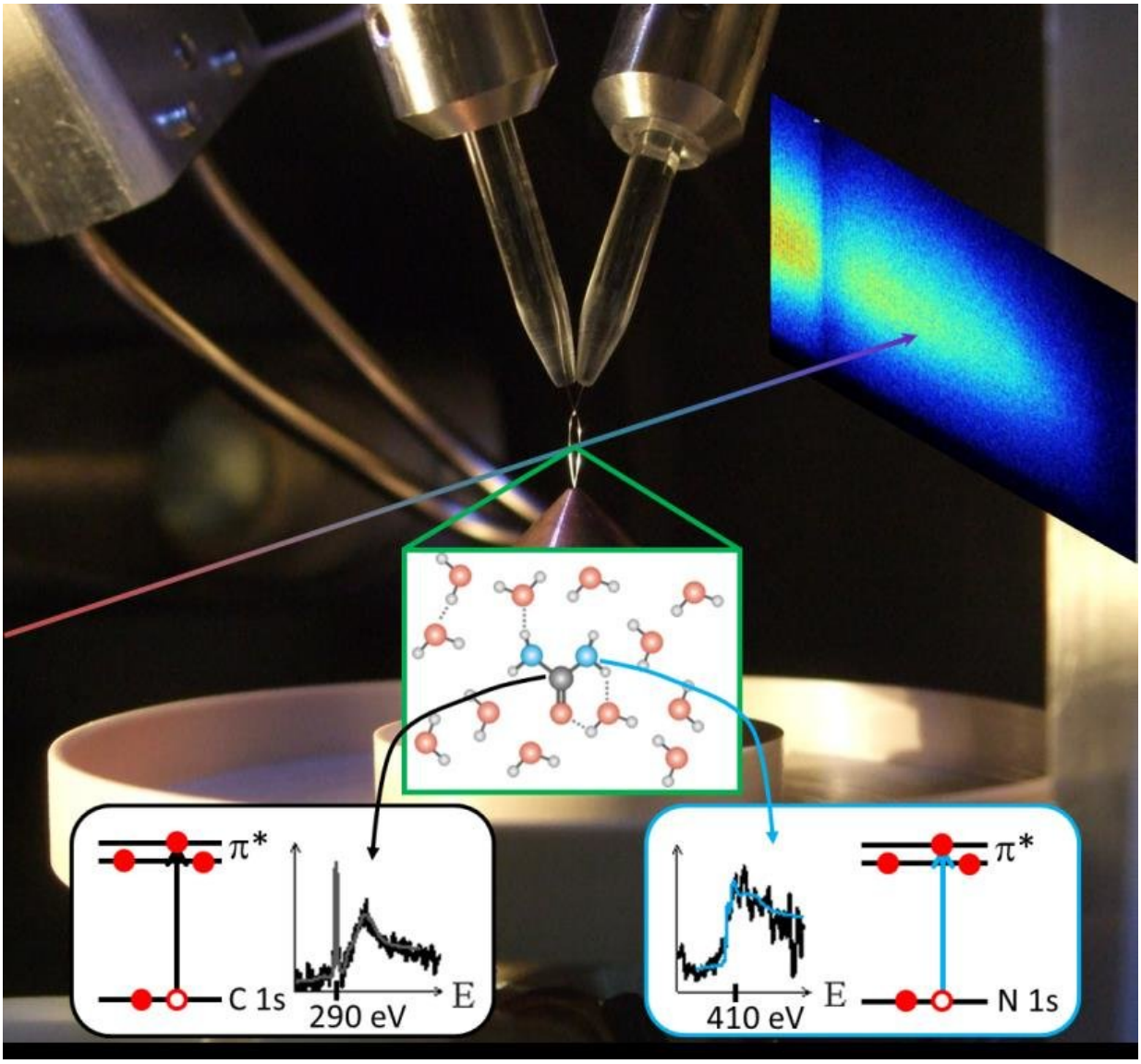

Liquid flatjet (solvated urea) illuminated by a broadband soft X-ray pulse

obtained by high-order harmonic generation. The insets show the steady-state absorption of Urea at the C and N K-edges extracted from the measurements. Credit: MBI Berlin

X-ray spectroscopy provides direct access into the nature of chemical bonds, from which the outcome of chemical reactions can be understood. Thus, researchers are exploring both X-ray source development and the implementation of new measurement methods. Researchers at the Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy (MBI) have now successfully combined a table-top laser-based extreme high-order harmonic source for short-pulse soft-X-ray absorption spectroscopy in the water window with novel flatjet technology. They are the first to demonstrate the simultaneous probing of carbon and nitrogen atoms in organic molecules in aqueous solution.

X-ray absorption spectroscopy (XAS) monitors unoccupied electronic orbitals with element specificity from which the electronic structure can be derived. For the majority of organic molecules, the soft-X-ray spectral region (100-1000 eV) is relevant, as K-edge transitions of low-Z elements (C, N, and O), and the L-edges of 3-D metals are found there. XAS is typically performed at large-scale facilities such as storage rings or free-electron lasers. Table-top laser-based sources have until now only been sparsely used to probe pure materials, e.g., metals and organic films. So far, measurements of the carbon or nitrogen K-edges of organic molecules in dilute aqueous solution have not been reported.

The research team at the MBI has now developed a bright source of femtosecond soft X-ray pulses by making use of the extreme high-order harmonic generation process. Long-wavelength (1.8 μm) driver pulses generated with an amplified Ti:sapphire laser system were used to generate high-order harmonics well above the conventional spectral

range, i.e., now extending up to 450 eV. They have combined this source with liquid flatjet technology fully functioning under vacuum conditions. Steady-state absorption spectra of organic molecules and inorganic salts in a thin (~ 1 μm) sheet of aqueous solution can now be measured throughout the so-called water window region between 200-540 eV (see Fig. 1). In particular, this technique enables the simultaneous local probing at both carbon and nitrogen sites within the molecules. With this the research, they have demonstrated the feasibility of following multiple sites within molecular systems, with the potential of probing possible correlations between these sites upon molecular rearrangements.

This investigation represents a major step towards the systematic investigation of ultrafast rearrangements of solution phase molecular systems with femtosecond soft X-ray spectroscopy. This could bring new insights into ultrafast charge transport processes and photo-induced reactions in chemistry and biology.

More information: Carlo Kleine et al, Soft X-ray Absorption Spectroscopy of Aqueous Solutions Using a Table-Top Femtosecond Soft X-ray Source, *The Journal of Physical Chemistry Letters* (2018). DOI: 10.1021/acs.jpclett.8b03420