Attosecond soft-x-ray spectroscopy of NO molecules using intense IR sources

Nobuhisa Ishii¹, Nariyuki Saito², Tomoya Mizuno², Teruto Kanai², Jiro Itatani²

¹Natinoal Institutes for Quantum and Radiological Science and Technology ²Institute for Solid State Physics, the University of Tokyo <u>IItatani@issp.u-tokyo.ac.jp</u>

Attosecond science has made tremendous progress in the past two decades with technological advancement of Ti:sapphire laser-based ultrafast technologies. Intense ultrashort pulses in visible and near infrared regions are now routinely produced, which allow to produce attosecond pulses in the extreme ultraviolet (EUV, <200 eV). Since the high harmonic cutoff is scaled by the ponderomotive potential that is proportional to the square of laser wavelength, intense ultrashort-pulse IR sources are required to extend the spectral range of attosecond pulses towards the soft X-ray range, where we can achieve element specific ultrafast spectroscopy using absorption edges [1]. Such atomic selectivity, photon-in/photon-out methodology, and the preservation of temporal resolution are attractive to construct attosecond spectroscopy that is generally applicable to atoms, molecules, and condensed matters even in the presence of strong external fields.

We develop carrier-envelope phase-stable ultrashort-pulse IR sources that uses BiB_3O_6 crystals as an optical parametric chirped-pulse amplification (OPCPA) medium. Because of the extremely broadband gain spectrum of BIBO-based OPCPA in 1.2-2.2 µm when pumped by Ti:sapphire lasers around 800 nm, we successfully produce CEP-stable sub-two-cycle IR pulses (1.5 mJ, 10 fs) at a repetition rate of 1 kHz [2]. The high pressure gas cell that is filled with Ne or He gases allow to generate attosecond continua that cover the entire water window. Attosecond transient spectroscopy of NO molecules reveals that he absorption spectra near the nitrogen *K*-edge (~400 eV) allow to capture electronic, vibrational, and rotational dynamics with a single spectrogram [3]. These results show that soft-x-ray absorption spectroscopy is a powerful technique to trace molecular dynamics of various freedoms in different time and energy scales.

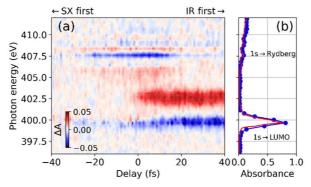


Fig. 1: (a) Measured transient absorbance change. (b) Measured absorbance without pump pulses (blue circles) and high-resolution absorbance from a synchrotron measurement.

References:

[1] Y. Pertot *et al.*, "Time-resolved x-ray absorption spectroscopy with a water window high-harmonic source," Science 355, 264 (2017); A. R. Attar *et al.*, "Femtosecond x-ray spectroscopy of an electrocyclic ring-opening reaction," Science 356, 54 (2017); A. Chew *et al.*, "Attosecond transient absorption spectrum of argon at the L2,3 edge," Phys. Rev. A 97, 031407(R) (2018).
[2] N. Ishii et al., "Generation of sub-two-cycle millijoule infrared pulses in an optical parametric chirped-pulse amplifier and their application to soft x-ray absorption spectroscopy with high-flux high harmonics," J. Opt. 20, 014003 (2018).

[3] N. Saito et al., ArXive:1904.10456 (2019).