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How Synchrotron Measurements Help Advance Attosecond Science

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VUV and soft X-ray synchrotron beamlines dedicated to molecular photoionization experiments have recently received strong competitors in the form of new advanced light sources, such as free electron lasers (FELs) and table-top high harmonic generation (HHG) laser systems. While the former provide access to significantly higher light intensities and to a time-resolution of tens of femtosecond, HHG sources offer the prospect to study electronic dynamics on the attosecond time scale. It will be shown, however, that the complementarity of synchrotron experiments, providing a high spectral resolution, and ultrafast time-resolved experiments with HHG sources can be exploited to get a profound understanding of molecular photoionization.
Molecular Frame Photoelectron Angular Distributions (MFPADs) from inner-valence shell ionization of the NO molecule were investigated employing 3D-momentum electron ion coincidence spectroscopy at the Pléiades and Désirs beamlines of Synchrotron SOLEIL as well as at the new 10 kHz HHG beamline of ATTOLab (Paris-Saclay). MFPADs describe the emission direction of the photoelectron in a photoionization reaction with respect to the orientation of the molecule. They encode information on the symmetry of the bound and continuum states, on quantum interferences between ionization channels and the phase and polarization state of the ionizing light. A molecular polarimetry based on reference MFPADs obtained with synchrotron radiation was employed to characterize the complete polarization state of the new HHG attosecond beamline at ATTOLab during commissioning. Ultrafast time-resolved experiments employing an XUV attosecond pulse train pump and a delayed IR probe were then performed to obtain first insights into the angular dependence of time-delay in molecular photoionization encoded in MFPADs.

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