

Photoelectron Diffraction on Laser-Aligned Molecules

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The possibility to obtain femtosecond time-resolved information on single molecules with Ångström spatial resolution (i.e. recording the so-called “molecular movie”) is one of the driving forces for the development of short-pulse VUV and X-ray sources such as Free-Electron Lasers (FELs). While coherent diffractive imaging, even though still facing tremendous challenges, certainly appears to be a promising avenue to study larger-scale structures, time-resolved photoelectron diffraction is another viable and, in many respects, complementary option. The site-selectivity of the inner-shell photoemission process allows selectively probing the local environment of the emitter, and Ångström resolution can already be obtained with photon energies a few hundred eV above a given inner-shell threshold. The photoelectron diffraction technique is especially valuable for investigating fundamental chemical reaction dynamics in small, polyatomic molecules, which are hard to probe by coherent imaging due to the low target density in a supersonic jet and the small X-ray scattering cross sections for low-Z molecular constituents.

Photoelectron angular distributions of adiabatically laser-aligned and mixed-field oriented p-fluorophenylacetylene molecules were measured at the AMO beamline of the Linac Coherent Light Source (LCLS) at SLAC National Accelerator Laboratory. A double-sided velocity map imaging (VMI) spectrometer was installed in the CFEL-ASG MultiPurpose (CAMP) chamber, which allowed recording electron and ion momentum distributions at the same time such that the degree of molecular alignment could be recorded simultaneously with the photoelectron diffraction patterns. The molecules were introduced in the chamber as a cold molecular beam produced by a pulsed Evan-Lavie valve and were adiabatically laser-aligned by a nanosecond Nd:YAG laser synchronized with the FEL pulses. One- and three-dimensional alignment as well as orientation of the molecules was measured at different photon energies to explore the sensitivity of the method to the (changing) molecular geometry.

By laser-aligning the molecules to a high degree, it is possible to obtain molecular-frame photoelectron distributions, which offer a structure-sensitive method to image molecules from inside. Through comparison with DFT and multiple scattering calculations, we can relate these measured photoelectron diffraction images to the geometric structure of the molecules.